

**Diffusivity Database (DDB) for Major Rocks:  
Database for the Second Progress Report**

October, 1999

Tokai Works

Japan Nuclear Cycle Development Institute

本資料の全部または一部を複写・複製・転載する場合は、下記にお問い合わせ  
ください。

〒319-1184 茨城県那珂郡東海村村松 4 番地 4 9

核燃料サイクル開発機構

技術展開部 技術協力課

Inquiries about copyright and reproduction should be addressed to:

Technical Cooperation Section,

Technology Management Division,

Japan Nuclear Cycle Development Institute

4-49 Muramatsu, Tokai-mura, Naka-gun, Ibaraki 319-1184,

Japan

© 核燃料サイクル開発機構 (Japan Nuclear Cycle Development Institute)

1999

# Diffusivity Database (DDB) for Major Rocks: Database for the Second Progress Report

Haruo Sato<sup>1</sup>

## Abstract

A database for diffusivity for a data setting of effective diffusion coefficients in rock matrices in the second progress report, was developed. In this database, 3 kinds of diffusion coefficients: effective diffusion coefficient ( $D_e$ ), apparent diffusion coefficient ( $D_a$ ) and free water diffusion coefficient ( $D_o$ ) were treated. The database, based on literatures published between 1980 and 1998, was developed considering the following points.

- (1) Since Japanese geological environment is focused in the second progress report, data for diffusion are collected focused on Japanese major rocks.
- (2) Although 22 elements are considered to be important in performance assessment for geological disposal, all elements and aquatic tracers are treated in this database development considering general purpose.
- (3) Since limestone, which belongs to sedimentary rock, can become one of the natural resources and is inappropriate as a host rock, it is omitted in this database development.

Rock was categorized into 4 kinds of rocks; acid crystalline rock, alkaline crystalline rock, sedimentary rock (argillaceous/tuffaceous rock) and sedimentary rock (psammitic rock/sandy stone) from the viewpoint of geology and mass transport. In addition, rocks around neutrality among crystalline rock were categorized into the alkaline crystalline rock in this database. The database is composed of sub-databases for 4 kinds of rocks. Furthermore, the sub-databases for 4 kinds of the rocks are composed of databases to individual elements, in which totally, 24 items such as species, rock name, diffusion coefficients ( $D_e$ ,  $D_a$ ,  $D_o$ ), obtained conditions (method, porewater, pH, Eh, temperature, atmosphere, etc.), etc. are input.

As a result of literature survey, for  $D_e$  values for acid crystalline rock, totally, 207 data for 18 elements and one tracer (hydrocarbon) have been reported and all data were for granitic rocks such as granite, granodiorite and biotitic granite. For alkaline crystalline rock, totally, 32  $D_e$  data for 6 elements have been reported for basalt, andesite and schist. For sedimentary rock (argillaceous/tuffaceous rock), totally, 54  $D_e$  data for 8 elements have been reported for mudstone, pelitic schist and tuff, and for sedimentary rock (psammitic rock/sandy stone), 11  $D_e$  data for 1 element have been reported for siliceous sedimentary rock. As is clear, much  $D_e$  data are found in granitic rocks. Whilst,  $D_e$  data for psammitic rocks are short comparing to those for the other kinds of rocks.

Correlativities have been approximately found between  $D_e$  values and rock porosities for all kinds of the rocks, but significant porosity dependencies have not been found for  $D_a$  values. In addition, correlations between various parameters such as that between formation factors and rock porosities, that between geometric factors and rock porosities, that between  $D_e$  values and ionic charge, that between  $D_e$  values and  $D_o$  values, and that between  $D_e$  values and element, were also discussed.

---

<sup>1</sup>Radiochemistry Group, Waste Isolation Research Division, Waste Management and Fuel Cycle Research Center, Tokai Works, Japan Nuclear Cycle Development Institute

# 主要岩石中の核種の拡散係数データベース -第2次取りまとめのためのデータベース- (研究報告)

佐藤治夫<sup>1</sup>

## 要 旨

第2次取りまとめにおいて、岩石マトリックス中の実効拡散係数設定のための拡散係数のデータベース（以下DBと略称）を整備した。本DBでは、実効拡散係数(De)、見掛けの拡散係数(Da)、自由水中の拡散係数(Do)の3種類の拡散係数を取り扱った。また、1980～1998年に公表された文献を対象として以下の点に留意して整備した。

- (1) 第2次取りまとめは、我が国の地質環境を対象としていることから、国内の岩石を対象として整備する。
- (2) 地層処分の性能評価では22元素を重要元素としているが、汎用性を考慮して全ての元素または水溶性トレーサを対象とする。
- (3) 対象岩石の内、堆積岩に含まれる石灰岩については、天然資源となり得ることから、DB整備の対象から除くものとする。

物質移行および地質学的観点から、岩種を結晶質岩(酸性)、結晶質岩(塩基性)、堆積岩(砂質岩類)、堆積岩(泥質・凝灰質岩類)の4種類に分類した。また、結晶質岩の内、中性付近の岩石については塩基性岩とした。DBは、各岩種単位で構成される。各岩種毎のDBはさらに各元素単位で整理し、各元素毎に化学種、岩石名、拡散係数(De, Da, Do)、取得条件(方法、間隙水、pH, Eh, 温度、雰囲気など)、文献など24項目の情報を入力した。

調査の結果、結晶質岩(酸性)に対するDeは、全部で18元素及びトレーサ(炭化水素)、207件のデータが報告されており、その全てが花崗岩、花崗閃緑岩、黒雲母花崗岩などの花崗岩類であった。結晶質岩(塩基性)に対しては、玄武岩、安山岩、片岩について、6元素、32件のDeデータが報告されていた。堆積岩(泥質・凝灰質岩類)に対しては、泥岩、泥質片岩、凝灰岩について、8元素、54件のDeデータが、また、堆積岩(砂質岩類)に対しては、珪質堆積岩について、1元素、11件のDeデータが報告されていた。これからも分かるように、データは花崗岩類に偏る傾向が見られた。一方、砂質岩類に対するデータが少ないことも分かった。各岩種ともDeは概ね間隙率と相関性が見られるものの、Daについては余り相関性が見られなかった。他に、形状因子や幾何学因子と間隙率との関係、Deとイオン電荷、Do、元素との関係など、様々なパラメータ間の関係についても議論した。

<sup>1</sup> 核燃料サイクル開発機構 東海事業所 環境保全・研究開発センター  
処分研究部 放射化学研究グループ

# Contents

## Chapter 1 General Introduction

<b>1.1 Introduction</b> .....	1
<b>1.2 Theoretical Background for Mass Transport</b> .....	2
1.2.1 Mass transport theory .....	2
1.2.2 Relations between diffusion coefficients and parameters expressing the pore structure of geological material .....	6
1.2.3 Free water diffusion coefficient ( $D_0$ ) .....	9
1.2.4 Porosity of geological material .....	11
<b>1.3 Methodology for Diffusivity Data Acquisition</b> .....	12
1.3.1 In-diffusion method .....	12
1.3.2 Through-diffusion method .....	14
1.3.3 Back-to-back method .....	18
1.3.4 Out-diffusion method .....	21
1.3.5 Reservoir-depletion method .....	24
1.3.6 Electrical conductivity method .....	26
<b>1.4 Basic Concept for Data Extraction for Performance Assessment</b> .....	28
1.4.1 Categorization of rock .....	28
1.4.2 Basic concept for data extraction .....	29
1.4.3 Basic structure of database .....	29
1.4.4 Literature survey .....	30
<b>1.5 References</b> .....	31

## Chapter 2 Diffusivity Data of Nuclides in Matrix Part of Domestic Major Rocks

<b>2.1 Sedimentary Rock: argillaceous/tuffaceous rock</b> .....	39
2.1.1 Chlorine (Cl) .....	39

2.1.2 Cesium (Cs)	-----	44
2.1.3 Iodine (I)	-----	48
2.1.4 Potassium (K)	-----	52
2.1.5 Sodium (Na)	-----	55
2.1.6 Nickel (Ni)	-----	59
2.1.7 Selenium (Se)	-----	63
2.1.8 Samarium (Sm)	-----	67
2.1.9 Uranium (U)	-----	71
2.1.10 Summary for difusivity data in argillaceous and tuffaceous rock	-----	74
<b>2.2 Sedimentary Rock: psammitic rock/sandy stone</b>	-----	<b>85</b>
2.2.1 Iodine (I)	-----	85
2.2.2 Summary for diffusivity data in psammitic rock/sandy stone	-----	89
<b>2.3 Alkaline Crystalline Rock</b>	-----	<b>100</b>
2.3.1 Chlorine (Cl)	-----	100
2.3.2 Cesium (Cs)	-----	103
2.3.3 Iodine (I)	-----	106
2.3.4 Potassium (K)	-----	109
2.3.5 Sodium (Na)	-----	111
2.3.6 Nickel (Ni)	-----	113
2.3.7 Summary for diffusivity data in alkaline crystalline rock	-----	117
<b>2.4 Acid Crystalline Rock</b>	-----	<b>128</b>
2.4.1 Barium (Ba)	-----	128
2.4.2 Beryllium (Be)	-----	132
2.4.3 Chlorine (Cl)	-----	135
2.4.4 Cobalt (Co)	-----	139
2.4.5 Cesium (Cs)	-----	144
2.4.6 Tritiated water (HTO)	-----	149
2.4.7 Iodine (I)	-----	153
2.4.8 Potassium (K)	-----	157
2.4.9 Magnesium (Mg)	-----	160
2.4.10 Sodium (Na)	-----	163

2.4.11 Nickel (Ni)	-----	167
2.4.12 Neptunium (Np)	-----	171
2.4.13 Plutonium (Pu)	-----	176
2.4.14 Selenium (Se)	-----	178
2.4.15 Samarium (Sm)	-----	183
2.4.16 Strontium (Sr)	-----	186
2.4.17 Technetium (Tc)	-----	191
2.4.18 Uranium (U)	-----	194
2.4.19 Uranine (Sodium Fluorescein: $C_{20}H_{10}O_5Na_2$ )	-----	199
2.4.20 Summary for diffusivity data in acid crystalline rock	-----	203

### Chapter 3 Conclusions

3.1 Summary	-----	214
3.2 Future Work	-----	215

### Chapter 4 Data Sheet

4.1 Data Sheet for Sedimentary Rock (argillaceous/tuffaceous rock)	-----	217
4.2 Data Sheet for Sedimentary Rock (psammitic rock/sandy stone)	-----	256
4.3 Data Sheet for Alkaline Crystalline Rock	-----	264
4.4 Data Sheet for Acid Crystalline Rock	-----	287

## Figures

### Chapter 1 General Introduction

Figure 1.2.2-1 Illustration for a categorization of pores	-----	7
---	-------	---

<b>Figure 1.2.2-2</b> Image of tortuosity -----	9
<b>Figure 1.3.1-1</b> Illustration of a diffusion experiment by in-diffusion method -----	13
<b>Figure 1.3.2-1</b> Illustration of a diffusion experiment by through-diffusion method -----	15
<b>Figure 1.3.2-2</b> Image of concentration profiles in bentonite and filters -----	17
<b>Figure 1.3.3-1</b> Illustration of a diffusion experiment by back-to-back method -----	19
<b>Figure 1.3.3-2</b> Illustration of a diffusion experiment by a variant of back-to-back method -----	20
<b>Figure 1.3.4-1</b> Illustration of a diffusion experiment by out-diffusion method -----	22
<b>Figure 1.3.5-1</b> Illustration of a diffusion experiment by reservoir-depletion method -----	24
<b>Figure 1.3.6-1</b> Illustration of a diffusion experiment by electrical conductivity method -----	28
<b>Figure 1.4.3-1</b> Basic structure of database for diffusion coefficient -----	30

## **Chapter 2 Diffusivity Data of Nuclides in Matrix Part of Domestic Major Rocks**

### **Sedimentary Rock: argillaceous/tuffaceous rock**

<b>Figure 2.1.1-1</b> Histogram for $D_e$ values of $\text{Cl}^-$ ion in sedimentary rock (mudstone, tuff, pelitic schist) -----	40
<b>Figure 2.1.1-2</b> Histogram for $D_a$ values of $\text{Cl}^-$ ion in sedimentary rock (mudstone, tuff, pelitic schist) -----	41
<b>Figure 2.1.1-3</b> $D_e$ values of $\text{Cl}^-$ ion as a function of rock porosity for sedimentary rock (mudstone, tuff, pelitic schist) -----	41
<b>Figure 2.1.1-4</b> $D_a$ values of $\text{Cl}^-$ ion as a function of rock porosity for sedimentary rock (mudstone, tuff, pelitic schist) -----	42
<b>Figure 2.1.2-1</b> Histogram for $D_e$ values of $\text{Cs}^+$ ion in sedimentary rock (mudstone) -----	45
<b>Figure 2.1.2-2</b> Histogram for $D_a$ values of $\text{Cs}^+$ ion in sedimentary rock (mudstone) -----	45



**Figure 2.1.2-3** De values of Cs<sup>+</sup> ion as a function of rock porosity for  
 sedimentary rock (mudstone) ----- 46

**Figure 2.1.2-4** Da values of Cs<sup>+</sup> ion as a function of rock porosity for  
 sedimentary rock (mudstone) ----- 46

**Figure 2.1.3-1** Histogram for De values of I<sup>-</sup> ion in sedimentary rock (tuff) ----- 49

**Figure 2.1.3-2** Histogram for Da values of I<sup>-</sup> ion in sedimentary rock (tuff) ----- 49

**Figure 2.1.3-3** De values of I<sup>-</sup> ion as a function of rock porosity for  
 sedimentary rock (tuff) ----- 50

**Figure 2.1.3-4** Da values of I<sup>-</sup> ion as a function of rock porosity for  
 sedimentary rock (tuff) ----- 50

**Figure 2.1.4-1** Histogram for De values of K<sup>+</sup> ion in sedimentary rock  
 (mudstone, tuff) ----- 53

**Figure 2.1.4-2** De values of K<sup>+</sup> ion as a function of rock porosity for  
 sedimentary rock (mudstone, tuff) ----- 53

**Figure 2.1.5-1** Histogram for De values of Na<sup>+</sup> ion in sedimentary rock  
 (mudstone, tuff) ----- 56

**Figure 2.1.5-2** Histogram for Da values of Na<sup>+</sup> ion in sedimentary rock (tuff) ----- 56

**Figure 2.1.5-3** De values of Na<sup>+</sup> ion as a function of rock porosity for  
 sedimentary rock (mudstone, tuff) ----- 57

**Figure 2.1.5-4** Da values of Na<sup>+</sup> ion as a function of rock porosity for  
 sedimentary rock (tuff) ----- 57

**Figure 2.1.6-1** Histogram for De values of Ni<sup>2+</sup> ion in sedimentary rock  
 (mudstone) ----- 60

**Figure 2.1.6-2** Histogram for Da values of Ni<sup>2+</sup> ion in sedimentary rock  
 (mudstone) ----- 60

**Figure 2.1.6-3** De values of Ni<sup>2+</sup> ion as a function of rock porosity for  
 sedimentary rock (mudstone) ----- 61

**Figure 2.1.6-4** Da values of Ni<sup>2+</sup> ion as a function of rock porosity for  
 sedimentary rock (mudstone) ----- 61

**Figure 2.1.7-1** Histogram for De values of SeO<sub>3</sub><sup>2-</sup> ion in sedimentary rock (tuff) ----- 64

**Figure 2.1.7-2** Histogram for Da values of SeO<sub>3</sub><sup>2-</sup> ion in sedimentary rock (tuff) ----- 64

**Figure 2.1.7-3** De values of  $\text{SeO}_3^{2-}$  ion as a function of rock porosity for  
 sedimentary rock (tuff) ----- 65

**Figure 2.1.7-4** Da values of  $\text{SeO}_3^{2-}$  ion as a function of rock porosity for  
 sedimentary rock (tuff) ----- 65

**Figure 2.1.8-1** Histogram for De values of  $\text{Sm}^{3+}$  ion in sedimentary rock  
 (mudstone) ----- 68

**Figure 2.1.8-2** Histogram for Da values of  $\text{Sm}^{3+}$  ion in sedimentary rock  
 (mudstone) ----- 68

**Figure 2.1.8-3** De values of  $\text{Sm}^{3+}$  ion as a function of rock porosity for  
 sedimentary rock (mudstone) ----- 69

**Figure 2.1.8-4** Da values of  $\text{Sm}^{3+}$  ion as a function of rock porosity for  
 sedimentary rock (mudstone) ----- 69

**Figure 2.1.9-1** Histogram for Da values of  $\text{UO}_2(\text{CO}_3)_3^{4-}$  ion in sedimentary rock  
 (tuff, tuffaceous sandstone) ----- 72

**Figure 2.1.10-1** Histogram for De values in sedimentary rock for 8 elements  
 (mudstone, pelitic schist, tuff) ----- 74

**Figure 2.1.10-2** Histogram for Da values in sedimentary rock for 8 elements  
 (mudstone, pelitic schist, tuff, tuffaceous sandstone) ----- 75

**Figure 2.1.10-3** De values for 8 elements as a function of porosity for  
 sedimentary rock (mudstone, pelitic schist, tuff) ----- 76

**Figure 2.1.10-4** Empirical equation for De values as a function of rock porosity  
 for sedimentary rock (mudstone, pelitic schist, tuff) ----- 77

**Figure 2.1.10-5** A width of variation in De values as a function of porosity for  
 sedimentary rock (mudstone, pelitic schist, tuff) ----- 78

**Figure 2.1.10-6** Da values as a function of rock porosity for sedimentary rock  
 (mudstone, pelitic schist, tuff, tuffaceous sandstone) ----- 79

**Figure 2.1.10-7** De values as a function of charge number of species ----- 80

**Figure 2.1.10-8** FF values as a function of rock porosity  
 (mudstone, tuff, pelitic schist) ----- 81

**Figure 2.1.10-9** Gf values as a function of rock porosity  
 (mudstone, tuff, pelitic schist) ----- 82

**Figure 2.1.10-10** A plot of De values for individual elements in sedimentary rock  
 (mudstone, tuff, pelitic schist) ----- 83

**Figure 2.1.10-11** A correlation of De values to Do values for sedimentary rock  
 (mudstone, tuff, pelitic schist) ----- 84

**Sedimentary Rock: psammitic rock/sandy stone**

**Figure 2.2.1-1** Histogram for De values of  $\Gamma^-$  ion in sedimentary rock  
 (siliceous sedimentary rock) ----- 86

**Figure 2.2.1-2** Histogram for Da values of  $\Gamma^-$  ion in sedimentary rock  
 (siliceous sedimentary rock) ----- 86

**Figure 2.2.1-3** De values of  $\Gamma^-$  ion as a function of rock porosity for  
 sedimentary rock (siliceous sedimentary rock) ----- 87

**Figure 2.2.1-4** Da values of  $\Gamma^-$  ion as a function of rock porosity for  
 sedimentary rock (siliceous sedimentary rock) ----- 87

**Figure 2.2.2-1** Histogram for De values in sedimentary rock for one element (for I)  
 (siliceous sedimentary rock) ----- 89

**Figure 2.2.2-2** Histogram for Da values in sedimentary rock for one element (for I)  
 (siliceous sedimentary rock) ----- 90

**Figure 2.2.2-3** De values for  $\Gamma^-$  ion as a function of porosity for sedimentary rock  
 (siliceous sedimentary rock) ----- 91

**Figure 2.2.2-4** Empirical equation for De values as a function of rock porosity  
 for sedimentary rock (siliceous sedimentary rock) ----- 92

**Figure 2.2.2-5** A width of variation in De values as a function of porosity for  
 sedimentary rock (siliceous sedimentary rock) ----- 93

**Figure 2.2.2-6** Da values as a function of rock porosity for sedimentary rock  
 (siliceous sedimentary rock) ----- 94

**Figure 2.2.2-7** De values as a function of charge number of species ----- 95

**Figure 2.2.2-8** FF values as a function of rock porosity for sedimentary rock  
 (siliceous sedimentary rock) ----- 96

**Figure 2.2.2-9** Gf values as a function of rock porosity for sedimentary rock  
 (siliceous sedimentary rock) ----- 97

**Figure 2.2.2-10** A plot of  $D_e$  values for individual elements in sedimentary rock  
 (siliceous sedimentary rock) ----- 98

**Figure 2.2.2-11** A correlation of  $D_e$  values to  $D_o$  values for sedimentary rock  
 (siliceous sedimentary rock) ----- 99

### Alkaline Crystalline Rock

**Figure 2.3.1-1** Histogram for  $D_e$  values of  $Cl^-$  ion in alkaline crystalline rock  
 (basalt) ----- 101

**Figure 2.3.1-2** Histogram for  $D_a$  values of  $Cl^-$  ion in alkaline crystalline rock  
 (basalt) ----- 101

**Figure 2.3.1-3**  $D_e$  values of  $Cl^-$  ion as a function of rock porosity for  
 alkaline crystalline rock (basalt) ----- 102

**Figure 2.3.1-4**  $D_a$  values of  $Cl^-$  ion as a function of rock porosity for  
 alkaline crystalline rock (basalt) ----- 102

**Figure 2.3.2-1** Histogram for  $D_e$  values of  $Cs^+$  ion in alkaline crystalline rock  
 (basalt) ----- 104

**Figure 2.3.2-2** Histogram for  $D_a$  values of  $Cs^+$  ion in alkaline crystalline rock  
 (basalt) ----- 104

**Figure 2.3.2-3**  $D_e$  values of  $Cs^+$  ion as a function of rock porosity for  
 alkaline crystalline rock (basalt) ----- 105

**Figure 2.3.2-4**  $D_a$  values of  $Cs^+$  ion as a function of rock porosity for  
 alkaline crystalline rock (basalt) ----- 105

**Figure 2.3.3-1** Histogram for  $D_e$  values of  $I^-$  ion in alkaline crystalline rock  
 (andesite, schist) ----- 107

**Figure 2.3.3-2** Histogram for  $D_a$  values of  $I^-$  ion in alkaline crystalline rock  
 (andesite, schist) ----- 107

**Figure 2.3.3-3**  $D_e$  values of  $I^-$  ion as a function of rock porosity for  
 alkaline crystalline rock (andesite, schist) ----- 108

**Figure 2.3.3-4**  $D_a$  values of  $I^-$  ion as a function of rock porosity for  
 alkaline crystalline rock (andesite, schist) ----- 108

**Figure 2.3.4-1** Histogram for  $D_e$  values of  $K^+$  ion in alkaline crystalline rock

(basalt) -----	110
<b>Figure 2.3.4-2</b> De values of $K^+$ ion as a function of rock porosity for alkaline crystalline rock (basalt) -----	110
<b>Figure 2.3.5-1</b> Histogram for De values of $Na^+$ ion in alkaline crystalline rock (basalt) -----	112
<b>Figure 2.3.5-2</b> De values of $Na^+$ ion as a function of rock porosity for alkaline crystalline rock (basalt) -----	112
<b>Figure 2.3.6-1</b> Histogram for De values of $Ni^{2+}$ ion in alkaline crystalline rock (basalt) -----	114
<b>Figure 2.3.6-2</b> Histogram for Da values of $Ni^{2+}$ ion in alkaline crystalline rock (basalt) -----	114
<b>Figure 2.3.6-3</b> De values of $Ni^{2+}$ ion as a function of rock porosity for alkaline crystalline rock (basalt) -----	115
<b>Figure 2.3.6-4</b> Da values of $Ni^{2+}$ ion as a function of rock porosity for alkaline crystalline rock (basalt) -----	115
<b>Figure 2.3.7-1</b> Histogram for De values in alkaline crystalline rock for 6 elements (basalt, andesite, schist) -----	117
<b>Figure 2.3.7-2</b> Histogram for Da values in alkaline crystalline rock (basalt, andesite, schist) -----	118
<b>Figure 2.3.7-3</b> De values for 6 elements as a function of porosity for alkaline crystalline rock (basalt, andesite, schist) -----	119
<b>Figure 2.3.7-4</b> Empirical equation for De values as a function of rock porosity for alkaline crystalline rock (basalt, andesite, schist) -----	120
<b>Figure 2.3.7-5</b> A width of variation in De values as a function of rock porosity for alkaline crystalline rock (basalt, andesite, schist) -----	121
<b>Figure 2.3.7-6</b> Da values as a function of rock porosity for alkaline crystalline rock (basalt, andesite, schist) -----	122
<b>Figure 2.3.7-7</b> De values as a function of charge number of species -----	123
<b>Figure 2.3.7-8</b> FF values as a function of rock porosity for alkaline crystalline rock (basalt, andesite, schist) -----	124
<b>Figure 2.3.7-9</b> Gf values as a function of rock porosity for alkaline crystalline rock	

(basalt, andesite, schist) -----	125
<b>Figure 2.3.7-10</b> A plot of De values for individual elements in alkaline crystalline rock	
(basalt, andesite, schist) -----	126
<b>Figure 2.3.7-11</b> A correlation of De values to Do values for alkaline crystalline rock	
(basalt, andesite, schist) -----	127

### Acid Crystalline Rock

<b>Figure 2.4.1-1</b> Histogram for De values of Ba <sup>2+</sup> ion in acid crystalline rock	
(granite) -----	130
<b>Figure 2.4.1-2</b> De values of Ba <sup>2+</sup> ion as a function of rock porosity for acid crystalline rock (granite)	
-----	130
<b>Figure 2.4.2-1</b> Histogram for De values of Be <sup>2+</sup> ion in acid crystalline rock	
(granite) -----	133
<b>Figure 2.4.2-2</b> De values of Be <sup>2+</sup> ion as a function of rock porosity for acid crystalline rock (granite)	
-----	133
<b>Figure 2.4.3-1</b> Histogram for De values of Cl <sup>-</sup> ion in acid crystalline rock	
(granite, granodiorite, altered granodiorite) -----	136
<b>Figure 2.4.3-2</b> Histogram for Da values of Cl <sup>-</sup> ion in acid crystalline rock	
(granodiorite, altered granodiorite) -----	136
<b>Figure 2.4.3-3</b> De values of Cl <sup>-</sup> ion as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite)	
-----	137
<b>Figure 2.4.3-4</b> Da values of Cl <sup>-</sup> ion as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)	
-----	137
<b>Figure 2.4.4-1</b> Histogram for De values of Co <sup>2+</sup> ion in acid crystalline rock	
(granite) -----	141
<b>Figure 2.4.4-2</b> Histogram for Da values of Co <sup>2+</sup> ion in acid crystalline rock	
(granite) -----	141
<b>Figure 2.4.4-3</b> De values of Co <sup>2+</sup> ion as a function of rock porosity for acid crystalline rock (granite)	
-----	142
<b>Figure 2.4.4-4</b> Da values of Co <sup>2+</sup> ion as a function of rock porosity for acid crystalline rock (granite)	
-----	142

**Figure 2.4.5-1** Histogram for De values of Cs<sup>+</sup> ion in acid crystalline rock  
 (granodiorite, altered granodiorite) ----- 146

**Figure 2.4.5-2** Histogram for Da values of Cs<sup>+</sup> ion in acid crystalline rock  
 (granite, granodiorite, altered granodiorite, biotitic granite) ----- 146

**Figure 2.4.5-3** De values of Cs<sup>+</sup> ion as a function of rock porosity for  
 acid crystalline rock (granodiorite, altered granodiorite) ----- 147

**Figure 2.4.5-4** Da values of Cs<sup>+</sup> ion as a function of rock porosity for acid crystalline  
 rock (granite, granodiorite, altered granodiorite, biotitic granite) ----- 147

**Figure 2.4.6-1** Histogram for De values of HTO in acid crystalline rock  
 (granodiorite, altered granodiorite) ----- 150

**Figure 2.4.6-2** Histogram for Da values of HTO in acid crystalline rock  
 (granodiorite, altered granodiorite) ----- 150

**Figure 2.4.6-3** De values of HTO as a function of rock porosity for  
 acid crystalline rock (granodiorite, altered granodiorite) ----- 151

**Figure 2.4.6-4** Da values of HTO as a function of rock porosity for acid crystalline  
 rock (granodiorite, altered granodiorite) ----- 151

**Figure 2.4.7-1** Histogram for De values of I<sup>-</sup> ion in acid crystalline rock  
 (granite) ----- 154

**Figure 2.4.7-2** Histogram for Da values of I<sup>-</sup> ion in acid crystalline rock  
 (granite) ----- 154

**Figure 2.4.7-3** De values of I<sup>-</sup> ion as a function of rock porosity for  
 acid crystalline rock (granite) ----- 155

**Figure 2.4.7-4** Da values of I<sup>-</sup> ion as a function of rock porosity for acid crystalline  
 rock (granite) ----- 155

**Figure 2.4.8-1** Histogram for De values of K<sup>+</sup> ion in acid crystalline rock  
 (granite) ----- 158

**Figure 2.4.8-2** De values of K<sup>+</sup> ion as a function of rock porosity for  
 acid crystalline rock (granite) ----- 158

**Figure 2.4.9-1** Histogram for De values of Mg<sup>2+</sup> ion in acid crystalline rock  
 (granite) ----- 161

**Figure 2.4.9-2** De values of Mg<sup>2+</sup> ion as a function of rock porosity for

acid crystalline rock (granite)	-----	161
<b>Figure 2.4.10-1</b> Histogram for De values of Na <sup>+</sup> ion in acid crystalline rock		
(granodiorite, altered granodiorite)	-----	164
<b>Figure 2.4.10-2</b> Histogram for Da values of Na <sup>+</sup> ion in acid crystalline rock		
(granodiorite, altered granodiorite)	-----	164
<b>Figure 2.4.10-3</b> De values of Na <sup>+</sup> ion as a function of rock porosity for		
acid crystalline rock (granodiorite, altered granodiorite)	-----	165
<b>Figure 2.4.10-4</b> Da values of Na <sup>+</sup> ion as a function of rock porosity for acid crystalline		
rock (granodiorite, altered granodiorite)	-----	165
<b>Figure 2.4.11-1</b> Histogram for De values of Ni <sup>2+</sup> ion in acid crystalline rock		
(granite, granodiorite)	-----	168
<b>Figure 2.4.11-2</b> Histogram for Da values of Ni <sup>2+</sup> ion in acid crystalline rock		
(granite, granodiorite)	-----	168
<b>Figure 2.4.11-3</b> De values of Ni <sup>2+</sup> ion as a function of rock porosity for		
acid crystalline rock (granite, granodiorite)	-----	169
<b>Figure 2.4.11-4</b> Da values of Ni <sup>2+</sup> ion as a function of rock porosity for acid crystalline		
rock (granite, granodiorite)	-----	169
<b>Figure 2.4.12-1</b> Histogram for De values of Np in acid crystalline rock (granite)	-----	173
<b>Figure 2.4.12-2</b> Histogram for Da values of Np in acid crystalline rock (granite)	-----	173
<b>Figure 2.4.12-3</b> De values of Np as a function of rock porosity for		
acid crystalline rock (granite)	-----	174
<b>Figure 2.4.12-4</b> Da values of Np as a function of rock porosity for acid crystalline		
rock (granite)	-----	174
<b>Figure 2.4.13-1</b> Histogram for De values of Pu in acid crystalline rock (granite)	-----	177
<b>Figure 2.4.14-1</b> Histogram for De values of SeO <sub>3</sub> <sup>2-</sup> ion in acid crystalline rock		
(granodiorite, altered granodiorite)	-----	180
<b>Figure 2.4.14-2</b> Histogram for Da values of SeO <sub>3</sub> <sup>2-</sup> or SeO <sub>4</sub> <sup>2-</sup> ion in acid crystalline rock		
(granodiorite, altered granodiorite)	-----	180
<b>Figure 2.4.14-3</b> De values of SeO <sub>3</sub> <sup>2-</sup> ion as a function of rock porosity for		
acid crystalline rock (granodiorite, altered granodiorite)	-----	181
<b>Figure 2.4.14-4</b> Da values of SeO <sub>3</sub> <sup>2-</sup> ion as a function of rock porosity for		



acid crystalline rock (granodiorite, altered granodiorite)	-----	181
<b>Figure 2.4.15-1</b> Histogram for De values of $\text{Sm}^{3+}$ ion in acid crystalline rock (granodiorite)	-----	184
<b>Figure 2.4.15-2</b> Histogram for Da values of $\text{Sm}^{3+}$ ion in acid crystalline rock (granodiorite)	-----	184
<b>Figure 2.4.15-3</b> De values of $\text{Sm}^{3+}$ ion as a function of rock porosity for acid crystalline rock (granodiorite)	-----	185
<b>Figure 2.4.15-4</b> Da values of $\text{Sm}^{3+}$ ion as a function of rock porosity for acid crystalline rock (granodiorite)	-----	185
<b>Figure 2.4.16-1</b> Histogram for De values of $\text{Sr}^{2+}$ ion in acid crystalline rock (granite)	-----	188
<b>Figure 2.4.16-2</b> Histogram for Da values of $\text{Sr}^{2+}$ ion in acid crystalline rock (granite)	-----	188
<b>Figure 2.4.16-3</b> De values of $\text{Sr}^{2+}$ ion as a function of rock porosity for acid crystalline rock (granite)	-----	189
<b>Figure 2.4.16-4</b> Da values of $\text{Sr}^{2+}$ ion as a function of rock porosity for acid crystalline rock (granite)	-----	189
<b>Figure 2.4.17-1</b> Histogram for De values of $\text{TcO}_4^-$ ion in acid crystalline rock (granite)	-----	192
<b>Figure 2.4.17-2</b> Histogram for Da values of $\text{TcO}_4^-$ ion in acid crystalline rock (granite)	-----	192
<b>Figure 2.4.17-3</b> De values of $\text{TcO}_4^-$ ion as a function of rock porosity for acid crystalline rock (granite)	-----	193
<b>Figure 2.4.17-4</b> Da values of $\text{TcO}_4^-$ ion as a function of rock porosity for acid crystalline rock (granite)	-----	193
<b>Figure 2.4.18-1</b> Histogram for De values of $\text{UO}_2^{2+}$ and $\text{UO}_2(\text{CO}_3)_3^{4-}$ ions in acid crystalline rock (granite, biotitic granite)	-----	196
<b>Figure 2.4.18-2</b> Histogram for Da values of $\text{UO}_2^{2+}$ ion in acid crystalline rock (granite, biotitic granite)	-----	196
<b>Figure 2.4.18-3</b> De values of $\text{UO}_2^{2+}$ and $\text{UO}_2(\text{CO}_3)_3^{4-}$ ions as a function of rock porosity for acid crystalline rock (granite, biotitic granite)	-----	197

**Figure 2.4.18-4** Da values of  $UO_2^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite, biotitic granite) ----- 197

**Figure 2.4.19-1** Histogram for De values of uranine in acid crystalline rock (granite) ----- 200

**Figure 2.4.19-2** Histogram for Da values of uranine in acid crystalline rock (granite) ----- 200

**Figure 2.4.19-3** De values of uranine as a function of rock porosity for acid crystalline rock (granite) ----- 201

**Figure 2.4.19-4** Da values of uranine as a function of rock porosity for acid crystalline rock (granite) ----- 201

**Figure 2.4.20-1** Histogram for De values in acid crystalline rock for 18 elements and one hydrocarbon (granite, granodiorite, biotitic granite) ----- 203

**Figure 2.4.20-2** Histogram for Da values in acid crystalline rock (granite, granodiorite, biotitic granite) ----- 204

**Figure 2.4.20-3** De values for 17 elements and one tracer (hydrocarbon) as a function of porosity for acid crystalline rock (granite, granodiorite, biotitic granite) ----- 205

**Figure 2.4.20-4** Empirical equation for De values as a function of rock porosity for acid crystalline rock (granite, granodiorite, biotitic granite) ----- 206

**Figure 2.4.20-5** A width of variation in De values as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite) ----- 207

**Figure 2.4.20-6** Da values as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite) ----- 208

**Figure 2.4.20-7** De values as a function of charge number of species ----- 209

**Figure 2.4.20-8** FF values as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite) ----- 210

**Figure 2.4.20-9** Gf values as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite) ----- 211

**Figure 2.4.20-10** A plot of De values for individual elements in acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite) ----- 212

**Figure 2.4.20-11** A correlation between De values and Do values for  
acid crystalline rock (granite, granodiorite, altered granodiorite,  
biotitic granite) ----- 213

## Tables

**Table 3.1-1** Summary of De and Da values for each element and each kind of rock  
treated in this database ----- 216

# Chapter 1 General Introduction

## 1.1 Introduction

The diffusion of nuclides in rock matrix is considered to be important for performance assessment of the geological disposal of high-level radioactive waste (HLW). Effective diffusion coefficient ( $D_e$ ) is particularly listed up as one of the essential parameters to evaluate flux, which expresses the amount of nuclides passed per unit area and unit time, and the release rates of nuclides [e.g. Sato, 1998].

For diffusion in rock matrix, nuclides are considered to migrate by a combination of concentration gradient, electric field gradient and the Brownian motion of molecular in grain-boundaries, micro fissures and cleavages, sorbing on minerals composing the rock matrix and occurring electro-physical interactions with the rock surfaces. It has been clarified by a natural analogue study for  $^{238}\text{U}$ -series disequilibrium at the Kamaishi In Situ Test Site, Iwate, Japan that matrix diffusion actually had occurred from a fracture surface into the matrix part [Ota et al., 1998].

Repository site for HLW in Japan has not been decided including candidate site yet. The kind of host rock has not been also therefore fixed. From this matter, the diffusion of nuclides in major rocks found in Japan must be evaluated in performance assessment. It is known that  $D_e$  values of nuclides in rock matrix generally depend on diffusion species [e.g. Sato and Shibutani, 1994]. It is therefore ideal that  $D_e$  is determined every species, but  $D_e$  values of nuclides in rock matrix obtained considered repository relevant conditions are quite limited.

Effective diffusion coefficients of nuclides in rock matrix were simply determined based on the formation factors (FF) of the rock matrices, which were derived from  $D_e$  values of HTO [Umeki et al., 1986] obtained by through-diffusion method [e.g. Eriksen et al., 1981; Lever, 1986; Muurinen et al., 1987; Cheung and Gray, 1989; Muurinen et al., 1989; Kita et al., 1989; Nishiyama et al., 1990; Park et al., 1991; Sato et al., 1992a; Yamaguchi et al., 1993],  $D_e$  values of  $\text{K}^+$  [Conca and Wright, 1992; Conca et al., 1993] obtained from electrical conductivity measurements [e.g. Conca et al., 1991, 1993; Conca and Wright, 1992; Okamoto et al., 1994, 1995; Nakazawa et al., 1997] and free water diffusion coefficients of nuclides ( $D_o$ ) in the first progress report for performance assessment, what is called, H-3 report [PNC, 1992]. In the H-3 report, 4 kinds of rocks; granite, basalt, mudstone and tuff, were treated as host rocks. In that model, FF of rock matrix was assumed to be constant for all elements (totally 22 elements<sup>1</sup> [e.g. PNC, 1991; Tochiyama et al., 1998]) to the same kind of rock, and  $D_e$  values were determined from the product between FF and  $D_o$  values of nuclides [Sato et al., 1992a]. The FF was determined every

---

<sup>1</sup> Key elements for performance assessment: Sn, Zr, Ni, Pd, Tc, Se, Sm, Nb, Sb, Cs, Cm, Am, Pu, Np, U, Th, Pa, Ac, Ra, Bi, Po, Pb

kind of rock. However, De and FF values have not been arranged as a database so far, FF values of individual rocks were determined from a small number of data. Thus the development of database for diffusion has been initiated after the H-3 report.

The effect of temperature on De was also evaluated for 25 and 60°C in the H-3 report, and De values were corrected based on the activation energy of Do [Sato et al., 1992a]. The De values of individual nuclides are however actually different depending on diffusion species, which individual nuclides take in the porewater. Some studies indicate that De is affected by surface diffusion of cationic species such as Cs<sup>+</sup> and Sr<sup>2+</sup> [e.g. Skagius and Neretnieks, 1982a; Yamaguchi et al., 1992, 1993]. However, anion exclusion of anionic species in rock matrix has not been discussed. The probable reason is due to anionic species such as I<sup>-</sup> and Cl<sup>-</sup> are generally used as a reference or a standard to evaluate the surface diffusion [e.g. Skagius and Neretnieks, 1982a], because they are non-sorptive onto bentonite and rocks. For anion exclusion of anionic species in rock matrix, Kumpulainen et al. [1991] have discussed to be caused by the change in the thickness of electric double layer near the mineral surface composing the rock matrix from the results of through-diffusion experiments of Cl<sup>-</sup> conducted as a function of the ionic strength of porewater (porewater was adjusted by NaCl concentration).

According to the Long-Term Program for Research, Development and Utilization of Atomic Energy issued on 24th, June in 1994 by the Atomic Energy Commission, it is planned that the second progress report for performance assessment of the geological disposal of HLW is published and submitted to the government before the year 2000 [Atomic Energy Commission, 1994]. For the sake of that, various studies and reportings concerning performance assessment have been carried out based on this schedule. In addition, implementing body will be also established around the year 2000.

The primary purpose of this report is to develop a database for matrix diffusion of nuclides in Japanese major rocks for the second progress report in performance assessment of geological disposal. Since the geological disposal is focused on Japanese geological environment, the database was developed focused on data obtained for Japanese major rocks.

This report introduces data integrated based on literature survey for matrix diffusion of nuclides in domestic major rocks, which were individually categorized.

## **1.2 Theoretical Background for Mass Transport**

### **1.2.1 Mass transport theory**

Before introduction and discussion for this database, theoretical background for mass transport and some definitions are explained here. Diffusion equation for three-dimensional non-steady state for a nuclide including advection, dispersion, sorption and decay is generally given by the following equation based on Fickian second law [e.g. RWMC, 1990].

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial X} \left( D_{ax} \cdot \frac{\partial C}{\partial X} \right) + \frac{\partial}{\partial Y} \left( D_{ay} \cdot \frac{\partial C}{\partial Y} \right) + \frac{\partial}{\partial Z} \left( D_{az} \cdot \frac{\partial C}{\partial Z} \right) - \frac{1}{R} \cdot \left( U_x \cdot \frac{\partial C}{\partial X} + U_y \cdot \frac{\partial C}{\partial Y} + U_z \cdot \frac{\partial C}{\partial Z} \right) - \lambda_d \cdot C \quad \text{----- (1.2.1-1)}$$

Where

C = concentration of an element (a nuclide) per unit volume on the (X, Y, Z) axis at an arbitrary time

t = time

D<sub>ax</sub> = apparent diffusion coefficient of an element (a nuclide) in the direction of the X axis

D<sub>ay</sub> = apparent diffusion coefficient of an element (a nuclide) in the direction of the Y axis

D<sub>az</sub> = apparent diffusion coefficient of an element (a nuclide) in the direction of the Z axis

X = distance in the direction of the X axis

Y = distance in the direction of the Y axis

Z = distance in the direction of the Z axis

R = retardation factor

λ<sub>d</sub> = decay constant

U<sub>x</sub> = flow velocity of the porewater in the direction of the X axis

U<sub>y</sub> = flow velocity of the porewater in the direction of the Y axis

U<sub>z</sub> = flow velocity of the porewater in the direction of the Z axis

If the diffusion coefficients D<sub>ax</sub>, D<sub>ay</sub> and D<sub>az</sub> are independent of distance X, Y and Z, equation (1.2.1-1) is rewritten as follows.

$$\frac{\partial C}{\partial t} = D_{ax} \cdot \frac{\partial^2 C}{\partial X^2} + D_{ay} \cdot \frac{\partial^2 C}{\partial Y^2} + D_{az} \cdot \frac{\partial^2 C}{\partial Z^2} - \frac{1}{R} \cdot \left( U_x \cdot \frac{\partial C}{\partial X} + U_y \cdot \frac{\partial C}{\partial Y} + U_z \cdot \frac{\partial C}{\partial Z} \right) - \lambda_d \cdot C \quad \text{----- (1.2.1-2)}$$

If the apparent diffusion coefficients D<sub>ax</sub>, D<sub>ay</sub> and D<sub>az</sub> are all the same and the flow velocities of porewater U<sub>x</sub>, U<sub>y</sub> and U<sub>z</sub> are also all the same, equation (1.2.1-2) is rewritten as follows.

$$\frac{\partial C}{\partial t} = D_a \cdot \left( \frac{\partial^2 C}{\partial X^2} + \frac{\partial^2 C}{\partial Y^2} + \frac{\partial^2 C}{\partial Z^2} \right) - \frac{U}{R} \cdot \left( \frac{\partial C}{\partial X} + \frac{\partial C}{\partial Y} + \frac{\partial C}{\partial Z} \right) - \lambda_d \cdot C \quad \text{----- (1.2.1-3)}$$

Where

D<sub>a</sub> = apparent diffusion coefficient of an element (a nuclide) (D<sub>a</sub>=D<sub>ax</sub>=D<sub>ay</sub>=D<sub>az</sub>)

U = flow velocity of the porewater (U=U<sub>x</sub>=U<sub>y</sub>=U<sub>z</sub>)

In performance assessment, mass transport in bentonite and in rock matrix is generally treated as diffusion without advection (U=0), because their hydraulic conductivities are quite low [e.g. Sato

et al., 1993]. In this case, equation (1.2.1-3) is rewritten as follows.

$$\frac{\partial C}{\partial t} = Da \cdot \left( \frac{\partial^2 C}{\partial X^2} + \frac{\partial^2 C}{\partial Y^2} + \frac{\partial^2 C}{\partial Z^2} \right) - \lambda_d \cdot C \quad \text{----- (1.2.1-4)}$$

Expressing equation (1.2.1-4) using Laplacian  $\nabla^2$ , we have the following equation.

$$\frac{\partial C}{\partial t} = Da \cdot \nabla^2 C - \lambda_d \cdot C \quad \text{----- (1.2.1-5)}$$

Where

$$\nabla^2 = \text{Laplacian} (\nabla^2 = \text{div grad} = \partial^2/\partial X^2 + \partial^2/\partial Y^2 + \partial^2/\partial Z^2)$$

Diffusive problem is generally treated in one-dimension. Retardation factor (R) is defined as follows [e.g. Sato et al. 1992b].

$$R = \frac{\phi + \rho \cdot Kd}{\phi} = 1 + \frac{\rho \cdot Kd}{\phi} \quad \text{----- (1.2.1-6)}$$

Where

$\phi$  = porosity

Kd = distribution coefficient

$\rho$  = dry bulk density

Equation (1.2.1-5) is therefore rewritten as the following equation using equation (1.2.1-6).

$$\begin{aligned} (\phi + \rho \cdot Kd) \cdot \frac{\partial C}{\partial t} &= \phi \cdot Dp \cdot \nabla^2 C - \lambda_d \cdot (\phi + \rho \cdot Kd) \cdot C \quad \text{----- (1.2.1-7)} \\ &= De \cdot \nabla^2 C - \lambda_d \cdot (\phi + \rho \cdot Kd) \cdot C \end{aligned}$$

Where

Dp = diffusion coefficient of an element (a nuclide) in the porewater

De = effective diffusion coefficient of an element (a nuclide)

The Da is disintegrated into some parameters as follows [e.g. Muurinen, 1994].

$$Da = \frac{\phi \cdot Dp}{\phi + \rho \cdot Kd} = \frac{De}{\phi + \rho \cdot Kd} \quad \text{----- (1.2.1-8)}$$

The R is also derived from equations (1.2.1-6) and (1.2.1-8) as follows.

$$R = \frac{D_p}{D_a} \quad \text{-----} \quad (1.2.1-9)$$

The  $D_a$  is further modelled as the following equation taking into account both pore diffusion and surface diffusion [e.g. Muurinen et al., 1985a, 1985b, 1987; Muurinen, 1994; Olin, 1994].

$$D_a = \frac{\phi \cdot D_p}{\phi + \rho \cdot K_d} + \frac{\rho \cdot K_d \cdot D_s}{\phi + \rho \cdot K_d} \quad \text{-----} \quad (1.2.1-10)$$

Where

$D_s$  = surface diffusion coefficient of an element (a nuclide)

For instance, equation (1.2.1-10) is used to estimate  $D_s$  values for cations such as  $\text{Cs}^+$  and  $\text{Sr}^{2+}$  [Eriksen and Jacobsson, 1996]. From equations (1.2.1-8) and (1.2.1-10),  $D_e$  is therefore expressed by the following equation [e.g. Muurinen, 1994; Olin, 1994].

$$D_e = \phi \cdot D_p + \rho \cdot K_d \cdot D_s \quad \text{-----} \quad (1.2.1-11)$$

The relational equation between concentration of an element (a nuclide) in the porewater  $C_p$  and concentration per unit volume  $C$  is given by the following equation.

$$C_p = \frac{C}{\phi + \rho \cdot K_d} = \frac{C}{\alpha} \quad \text{-----} \quad (1.2.1-12)$$

Where

$C_p$  = concentration of an element (a nuclide) in the porewater

$\alpha$  = rock capacity factor

The  $D_e$  is the parameter to directly affect flux, which expresses the amount of an element (a nuclide) passed per unit area and unit time. For instance, the flux is expressed by the following equation using  $D_e$  based on Fickian first law [Crank, 1975].

$$\vec{J} = -D_e \cdot \nabla C_p \quad \text{-----} \quad (1.2.1-13)$$

Where

$\nabla$  = nabla ( $\nabla = \text{grad} = \mathbf{i} \cdot \partial C_p / \partial X + \mathbf{j} \cdot \partial C_p / \partial Y + \mathbf{k} \cdot \partial C_p / \partial Z$ )

$\mathbf{i}, \mathbf{j}, \mathbf{k}$  = unit vectors in the directions of the X, Y and Z axes, respectively

$\vec{J}$  = mass flux vector of an element (a nuclide)

Though equation (1.2.1-13) shows the flux for three-dimensions, diffusive problem is often treated for homogeneous media and is generally solved in one-dimension. The flux in one-



dimensional diffusion equation is given by the following equation [Crank, 1975].

$$J = -De \cdot \left( \frac{\partial C_p}{\partial X} \right) \text{----- (1.2.1-14)}$$

Where

J = mass flux of an element (a nuclide) in the direction of one-dimension

As shown in equations (1.2.1-13) and (1.2.1-14), flux directly depends on De. The De is therefore regarded as an important parameter to affect the release rates of nuclides from repository.

The De is also generally affected by some parameters such as the charge number of species and Do of diffusion species [e.g. Sato and Shibutani, 1994]. It is known from existing results that cations and anions respectively show high and low De values compared with neutral species such as HTO [e.g. Sato and Shibutani, 1994]. Moreover, it is generally said that surface diffusion occurs for cations [Muurinen et al., 1985a, 1985b, 1987; Cheung and Gray, 1989; Yamaguchi et al., 1993; Olin, 1994] in the porewater of compacted bentonite and rock matrix, and to the contrary, that anions are retarded by ion exclusion (in this case anion exclusion)[McKinley and Hadermann, 1984], since the surface of bentonite and rock matrix is totally negatively charged around pH8-9 which is generally said to take in the porewater of compacted bentonite [e.g. Sasaki et al., 1995] and rock matrix.

## 1.2.2 Relations between diffusion coefficients and parameters expressing the pore structure of geological material

Though how to express De is not necessarily united, De is generally expressed by the following equation [e.g. Skagius and Neretnieks, 1982a, 1982b; Yui et al., 1994; Olin, 1994; Muurinen, 1994].

$$De = \phi \cdot D_p = \phi \cdot \left( \frac{\delta}{\tau^2} \right) \cdot D_o \text{----- (1.2.2-1)}$$

Where

$\phi$  = porosity

$\delta$  = constrictivity

$\tau^2$  = tortuosity ( $\tau$  is also called so)

$D_o$  = free water diffusion coefficient of a species

Where the porosity  $\phi$  should be diffusion or transport porosity actually contributed, what is called, effective porosity [e.g. Brandberg and Skagius, 1991], but it is quite difficult to estimate

porosity actually contributing for diffusion, because the diffusion or transport porosity actually depends on diffusion species. Based on equation (1.2.1-8), the porosity actually contributed for diffusion can be however estimated by using non-sorbing and non-interactive nuclide.

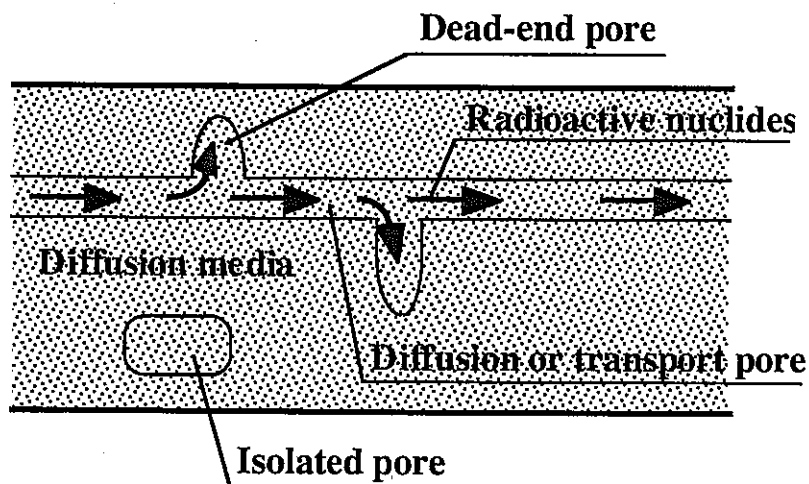
In equation (1.2.1-8), if  $K_d=0$ , it becomes that  $D_a=D_p$ . The pore is categorized into diffusion or transport pore, isolated pore and dead-end pore, and total porosity is expressed by the following equation.

$$\phi_{total} = \phi_{diff} + \phi_{iso} + \phi_{dead} \quad \text{----- (1.2.2-2)}$$

Where

- $\phi_{total}$  = total porosity
- $\phi_{diff}$  = diffusion or transport porosity
- $\phi_{iso}$  = isolated porosity
- $\phi_{dead}$  = dead-end porosity

The porosity is generally treated as  $\phi = \phi_{total}$ . **Figure 1.2.2-1** shows an illustration for a categorization of the pores.



**Figure 1.2.2-1** Illustration for a categorization of pores

- Diffusion or transport pore:** pores contributing for diffusion or transport and interconnected pores.
- Isolated pore:** pores existing independent of the other pores and not connected with any pores.
- Dead-end pore:** blind pores and also called storage pores [e.g. Brandberg and Skagius, 1991] because the pores don't directly contribute for transport, but can keep some amount of nuclides in them.

In equation (1.2.2-1),  $\phi \cdot \delta / \tau^2$  connecting  $D_e$  and  $D_o$  is called a formation factor, and is expressed by the following equation [e.g. Skagius and Neretnieks, 1982a, 1982b; Albinsson and Engkvist, 1991; Yui et al., 1994; Olin, 1994; Muurinen, 1994].

$$FF = \frac{D_e}{D_o} = \phi \cdot \left( \frac{\delta}{\tau^2} \right) = \phi \cdot Gf \quad \text{----- (1.2.2-3)}$$

Where

FF = formation factor

Gf = geometric factor (or also called tortuosity factor)

The Gf is here a physical parameter expressing the pore structure of geological material and is independent of porosity. The Gf is defined by the following equation [e.g. Olin, 1994; Muurinen, 1994].

$$Gf = \frac{D_p}{D_o} \quad \text{----- (1.2.2-4)}$$

The parameters  $\phi$ ,  $\delta$ ,  $\tau^2$ , FF and Gf described above are generally all physical parameters, and can not explain well that  $D_e$  values of elements (nuclides) depend on the charge of species such as surface diffusion or ion exclusion. The  $\delta$  expresses the effect of the change in pore form on diffusion and is originally characterized by the ratio of the minimum cross section to the maximum one [Van Brakel and Heertjets, 1974]. This definition is however for soil which has relatively large pore sizes and is considered to be inappropriate for highly compacted bentonite with ultra narrow pores which sizes are nearly ionic size level. Electrostatic interaction of species with the surface of bentonite can not be ignored in such case. The  $\delta$  therefore expresses not only physical effect, but also includes the effect of electrostatic interaction. Sato et al. [Sato and Shibutani, 1994; Sato et al., 1995; Sato, 1997, 1998] and Ochs et al. [Ochs et al., 1998] have modelled this parameter based on electric double layer theory. Moreover, Kato et al. [Kato et al., 1995] have modelled  $D_p$  based on the same theory. Though both models were originally developed to explain  $D_e$  values of nuclides in compacted bentonite, the availability and applicability are presently under discussion. Therefore, these models have not been applied for actual performance assessment.

Whilst the  $\tau$  stands for the tortuous degree of pores and is expressed by the following equation [e.g. Nakano, 1991; Shackelford and Daniel, 1991].

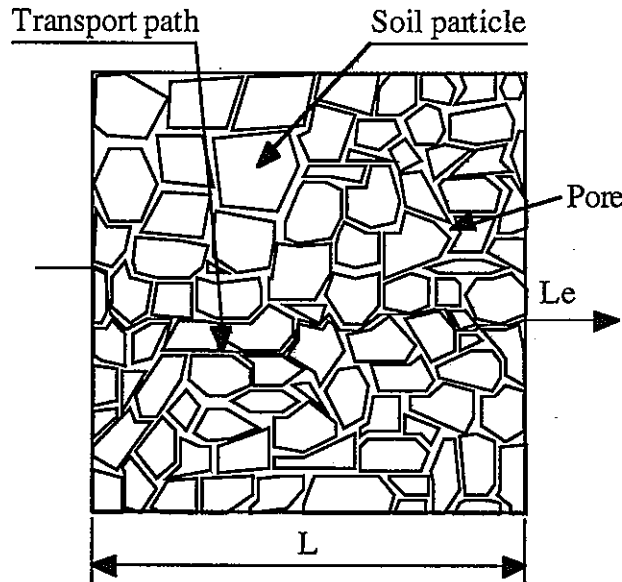
$$\tau = \frac{L_e}{L} \quad \text{----- (1.2.2-5)}$$

Where

$L_e$  = length of effective diffusion path

$L$  = length of straight path (corresponding to the thickness of sample)

**Figure 1.2.2-2** shows an image of the tortuosity. Nuclides can generally not go straight in sample as shown in **Figure 1.2.2-2**. The tortuosity is the factor to express this degree.



**Figure 1.2.2-2** Image of tortuosity

### 1.2.3 Free water diffusion coefficient ( $D_o$ )

By the way, it is well known that  $D_o$  generally depends on species. The  $D_o$  is calculated by the following Nernst-Einstein equation (Nernst expression)[e.g. Robinson and Stokes, 1959; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991].

$$D_o = \frac{R \cdot T \cdot \Lambda_o}{F^2 \cdot |Z|} \text{----- (1.2.3-1)}$$

Where

- R = gas constant (8.314 J/mol/K)
- T = absolute temperature
- $\Lambda_o$  = limiting ionic equivalent conductivity
- F = Faraday constant ( $F=e \cdot N$ )(96,493 Coulomb/mol)
- N = Avogadro number (6.02E23/mol)
- e = elementary charge
- |Z| = absolute value of the ionic charge

For an uncharged particle,  $D_o$  can be also calculated by the following Einstein-Stokes formula [Robinson and Stokes, 1959; Olin, 1994; Otaki, 1990].

$$D_o = \frac{k \cdot T}{6\pi \cdot \eta \cdot r} = \frac{R \cdot T}{6\pi \cdot \eta \cdot r \cdot N} \quad \text{----- (1.2.3-2)}$$

Where

- $\kappa$  = Boltzmann's constant ( $\kappa=R/N=1.3803E-23$  J/K)
- $\eta$  = viscosity of the porewater
- $r$  = radius of the particle

The  $D_o$  is often taken conservatively as  $2.0E-9$  m<sup>2</sup>/s for all species in performance assessment [e.g. Brandberg and Skagius, 1991]. A variation of more than one order of magnitude is however actually found depending on the nature of the chemical species [Sato et al., 1996]. For example,  $D_o$  of  $Th^{4+}$  at 18°C is  $1.53E-10$  m<sup>2</sup>/s [Brandberg and Skagius, 1991] and that of  $Cs^+$  is  $1.77E-9$  m<sup>2</sup>/s [Li and Gregory, 1974]. The highest  $D_o$  of those reported to the present is self-diffusion coefficient of water, which  $D_o$  is  $2.14E-9$  m<sup>2</sup>/s at 25 °C [Chemical Handbook, 1975].

With respect to a dual electrolyte such as NaCl,  $D_o$  is given by the Nernst-Hartley expression [Tamamushi, 1974].

$$D_o = \frac{R \cdot T}{F^2} \cdot \left\{ \left( \frac{v_+ + v_-}{v_+ \cdot |Z_+|} \right) \cdot \left( \frac{\Lambda_+ \cdot \Lambda_-}{\Lambda_+ + \Lambda_-} \right) \right\} \quad \text{----- (1.2.3-3)}$$

Where

- $v_+, v_-$  = activities for cations and anions respectively
- $|Z_+|$  = absolute value of the charge number of cation
- $\Lambda_+, \Lambda_-$  = limiting ionic equivalent conductivities for cations and anions respectively

The  $D_o$  of dual electrolyte can be also calculated by the following equation [Matsuo, 1990].

$$D_o = \frac{(|Z_+| + |Z_-|) \cdot D_{o+} \cdot D_{o-}}{|Z_+| \cdot D_{o+} + |Z_-| \cdot D_{o-}} \quad \text{----- (1.2.3-4)}$$

Where

- $D_o$  = free water diffusion coefficient of the electrolyte
- $|Z_+|, |Z_-|$  = absolute values of the charge numbers of cations and anions respectively
- $D_{o+}, D_{o-}$  = free water diffusion coefficients for cations and anions respectively

## 1.2.4 Porosity of geological material

The porosity is an important parameter as described in detail before and is given as the ratio of pore volume to total sample volume [Monicard, 1980]. The porosity of the sample is generally calculated by the following equation [e.g. Sato et al., 1993].

$$\phi = 1 - \frac{\rho}{\rho_{th}} \quad \text{----- (1.2.4-1)}$$

Where

$\rho$  = dry bulk density

$\rho_{th}$  = pure density of the sample (theoretical density)

The porosity in this case means the total porosity and does not mean transport porosity. However, it is difficult to measure actual transport porosity and the total porosity is generally used.

The pure density of a bentonite, Kunigel-V1<sup>②</sup> is given to be 2.7 Mg/m<sup>3</sup> from the pure density measurements and an estimation from molecular structure and molecular weight [Suzuki et al., 1992]. The pure densities of both Na-bentonites MX-80<sup>③</sup> and Kumipia-F<sup>④</sup> are also estimated to be both 2.7 Mg/m<sup>3</sup> [Suzuki et al., 1992].

The porosity calculated by equation (1.2.4-1) makes good for engineered barrier such as bentonite, but it is not generally used for natural rocks, because the porosity can not be theoretically calculated. The porosity besides depends on the kind of rock. Therefore, the porosity of rock is directly measured in general. The porosity is defined by the following equation [Monicard, 1980].

$$\phi = \frac{V_p}{V_s} = \frac{V_s - V_{th}}{V_s} \quad \text{----- (1.2.4-2)}$$

Where

$V_p$  = pore volume of the sample

<sup>②</sup> Kunigel-V1 is a product of the Kunimine Industries Co. Ltd. The mineralogy of the bentonite is described in detail in the literatures of Ito et al. [Ito et al., 1993, 1994]. Major constituent clay mineral is Na-montmorillonite, which mode is 46-49 wt%, and chalcedony, quartz, plagioclase, calcite, dolomite, analcite and pyrite are contained in the bentonite as impurities.

<sup>③</sup> MX-80 is a product of the American Colloid Company. The constituent content of Na-montmorillonite is 75 wt% [Müller and Kahr, 1983].

<sup>④</sup> Kunipia-F is a product of the Kunimine Industries Co. Ltd. This bentonite is the purified one which mode of Na-montmorillonite is over 99 wt% [Sato et al., 1992b; Kunimine Industries Co. Ltd., Private communication].

$V_s$  = total sample volume

$V_{th}$  = pure solid volume of the sample

The porosity is generally determined by liquid intrusion method (e.g. water saturation method, mercury intrusion method, kerosine intrusion method, etc.) and gas intrusion method (e.g. helium intrusion). These are the methods to measure porosity by compensating sample pores with fluid. Moreover, the porosity can be also determined by a leaching method, in which method the porosity is estimated from an equilibrium concentration of a non-sorbing tracer leached out from a sample saturated with the tracer in advance [Skagius and Neretnieks, 1985]. Then the porosity is determined by the following equation.

$$\phi = \frac{C_o \cdot V_s - C_p \cdot V_r}{C_p \cdot V_s} = \frac{C_o}{C_p} - \frac{V_r}{V_s} \quad \text{----- (1.2.4-3)}$$

Where

$C_o$  = initial concentration of the non-sorbing tracer contained per unit sample volume

$C_p$  = concentration of the non-sorbing tracer in the reservoir at the equilibrium

$V_s$  = total sample volume

$V_r$  = solution volume in the reservoir

### 1.3 Methodology for Diffusivity Data Acquisition

This section mainly explains 6 methods for diffusivity data acquisition including the theoretical derivation of diffusivity.

#### 1.3.1 In-diffusion method

From this method,  $D_a$  can be basically obtained [e.g. Eriksen et al., 1981; Torstenfelt et al., 1982a, 1982b; Nowak, 1983; Muurinen et al., 1983; Torstenfelt and Allard, 1986; Lever, 1986; Christiansen and Torstenfelt, 1988; Sharma and Oscarson, 1989; Idemitsu et al., 1990; Sato et al., 1991, 1992a, 1992b; Ota and Yoshida, 1994]. **Figure 1.3.1-1** shows an illustration of a diffusion experiment by the in-diffusion method. The end of a cylindrical sample contacts with a solution containing tracer for certain time period, allowing the tracer to diffuse into the sample. Then the concentration distribution (concentration profile) of the tracer in the sample is analysed after the sample was sliced or scraped, and  $D_a$  is obtained based on Fickian second law from the concentration profile. The  $D_a$  can be estimated from the following equation [Crank, 1975].

**Initial condition**

$$C=0, t=0, |X| \neq 0$$

**Boundary condition**

$$C=C_o, t > 0, X=0$$

$$C=0, t>0, |X|=\infty$$

$$C = C_0 \cdot \operatorname{erfc}\left(\frac{|X|}{2\sqrt{Da \cdot t}}\right) = \left\{ 1 - \operatorname{erf}\left(\frac{|X|}{2\sqrt{Da \cdot t}}\right) \right\} \quad \text{----- (1.3.1-1)}$$

$$\operatorname{erfc}(w) = 1 - \operatorname{erf}(w) = 1 - \frac{2}{\sqrt{\pi}} \int_0^w \exp(-\eta^2) d\eta$$

$$\lim_{w \rightarrow \infty} \left\{ \int_0^w \exp(-\eta^2) d\eta \right\} = \frac{\sqrt{\pi}}{2}$$

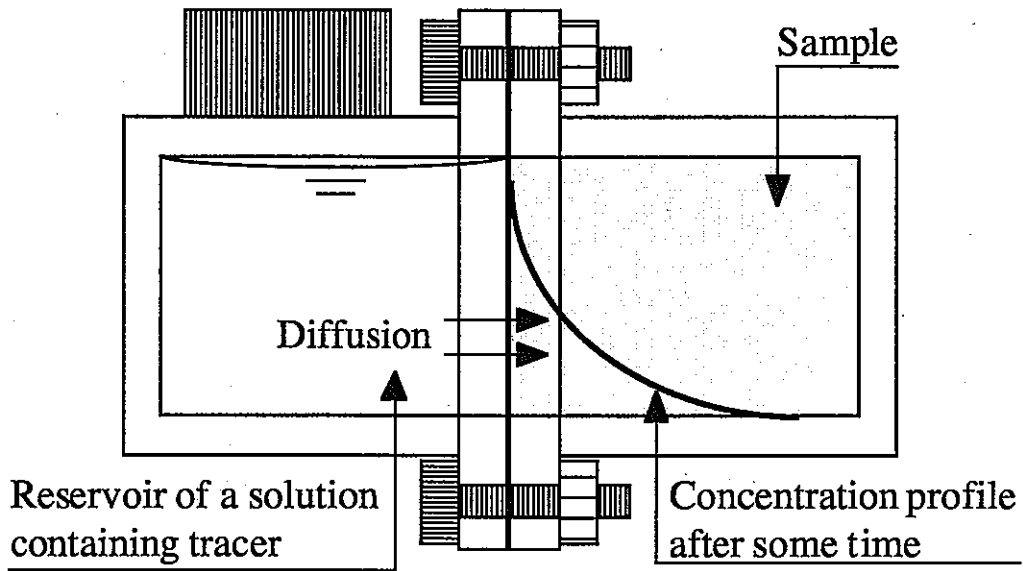
Where

C = concentration of the tracer per unit sample volume

C<sub>0</sub> = concentration of the tracer per unit sample volume at the end of the sample which the tracer solution contacted

X = distance from the source

t = time



**Figure 1.3.1-1** Illustration of a diffusion experiment by in-diffusion method

This is a popular method for relatively soft rocks such as tuff [Kosaka et al., 1984; Ota and Yoshida, 1994](though few studies have been reported for crystalline rock, some studies are found for crystalline rock such as granite [e.g. Tsukamoto and Ohe, 1988]) and for bentonite [e.g. Eriksen et al., 1981; Torstenfelt et al., 1982a, 1982b; Nowak, 1983; Muurinen et al., 1983; Torstenfelt and Allard, 1986; Lever, 1986; Christiansen and Torstenfelt, 1988; Sharma and Oscarson, 1989; Sato et al., 1991, 1992a, 1992b; 1993, 1995; Sato and Shibutani, 1994], being



adopted in many studies. The concentration of the tracer solution contacting with sample must be maintained constant during the experiment. The concentration profile of the tracer in the sample must be also obtained to determine  $Da$ . As described above, this method is popular for diffusion of nuclides in bentonite, but not so many studies have been reported for rocks, because it is generally a little difficult to carry out slicing or scraping depending on the kind of rock. It is listed up as one of the reasons that crystalline rocks such as granitic rocks and basalt are generally too hard to slice and to scrape.

Not only  $Da$ , but also rock capacity factor can be obtained from this method. In this method, the concentration of tracer in the reservoir is kept constant during the experiment. The concentration at the end of the sample, which contacted with the tracer solution is also obtained from concentration profile. From both concentrations, the rock capacity factor can be estimated from the following relation.

$$\alpha = \phi + \rho \cdot Kd = \phi + \rho_{th} \cdot (1 - \phi) \cdot Kd = \frac{C}{C_p} \Big|_{x=0} \text{-----} (1.3.1-2)$$

Where

$\alpha$  = rock capacity factor

$\phi$  = porosity of the sample

$\rho$  = dry bulk density of the sample

$\rho_{th}$  = pure density of the sample (theoretical density)

$Kd$  = distribution coefficient

$C$  = concentration of the tracer element per unit sample volume at the end of the sample where the tracer solution contacted

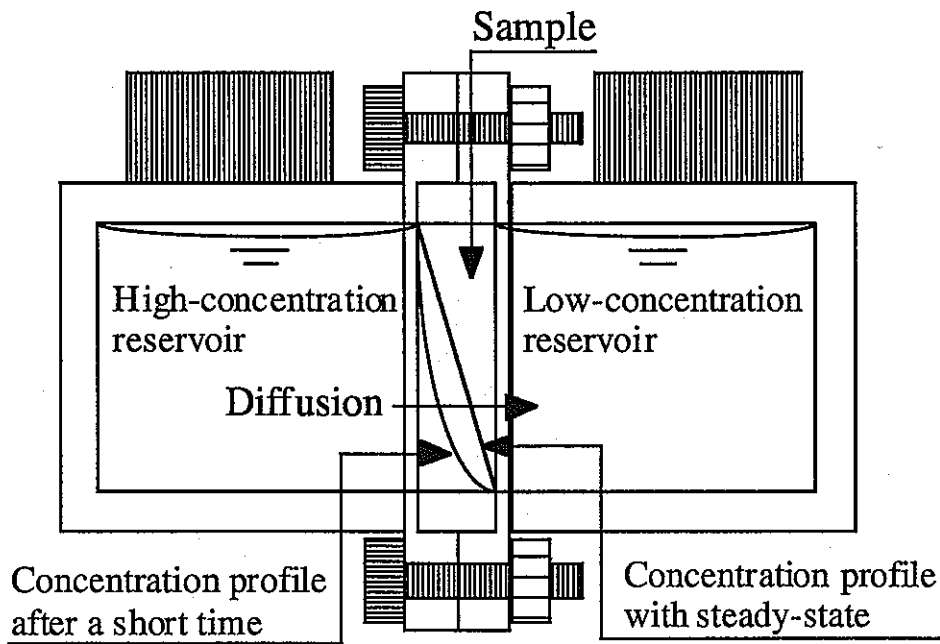
$C_p$  = concentration of the tracer element in the porewater at the end of the sample which the tracer solution contacted

In this case,  $C_p$  at the end of the sample, which contacted with the tracer solution ( $X=0$ ) is equal to the concentration of tracer in the reservoir.

### 1.3.2 Through-diffusion method

From this method,  $De$  can be basically obtained [e.g. Eriksen et al., 1981; Lever, 1986; Muurinen et al., 1987; Cheung and Gray, 1989; Muurinen et al., 1989; Kita et al., 1989; Park et al., 1991; Ishida et al., 1991; Sato et al., 1992a, 1994, 1997; Sato, 1996; Yamaguchi et al., 1993, 1997]. **Figure 1.3.2-1** shows an illustration of a diffusion experiment by the through-diffusion method. A sample of rock is sandwiched between two reservoirs, of which one contains tracer (tracer cell or reservoir cell) and tracer free porewater is injected into the other cell (measurement cell or low concentration reservoir). The tracer diffuses into and through the rock sample. The concentration of the tracer in the tracer cell initially remains much higher than that in the measurement cell,

where the concentration is monitored. The  $D_e$  can be obtained from the change in concentration of the tracer in the measurement cell as a function of time in steady state.



**Figure 1.3.2-1** Illustration of a diffusion experiment by through-diffusion method

The concentration of the tracer in the measurement cell at an arbitrary time based on initial and boundary conditions, is written as follows [Crank, 1975]:

**Initial condition**

$$C_p=0, t=0, 0 \leq X \leq L$$

**Boundary condition**

$$C_p=C_0, t>0, X=0$$

$$C_p=0, t>0, X=L$$

$$C_t = \frac{A \cdot L \cdot C_0}{V_m} \cdot \left[ \frac{D_e}{L^2} \cdot t - \frac{\alpha}{6} - \frac{2\alpha}{\pi^2} \sum_{n=1}^{\infty} \left\{ \frac{(-1)^n}{n^2} \cdot \exp\left(-\frac{D_e \cdot n^2 \cdot \pi^2 \cdot t}{L^2 \cdot \alpha}\right) \right\} \right] \quad \text{----- (1.3.2-1)}$$

Where

- $C_t$  = concentration of the tracer in the measurement cell at an arbitrary time
- $A$  = cross-section area of the sample
- $L$  = thickness of the sample
- $C_0$  = concentration of the tracer in the tracer cell
- $V_m$  = solution volume in the measurement cell
- $D_e$  = effective diffusion coefficient

t = time

$\alpha$  = rock capacity factor

At long time such as steady state, the exponentials fall away to zero. Therefore, equation (1.3.2-1) is approximately written for steady state as follows [Crank, 1975]:

$$C_t = \frac{A \cdot L \cdot C_o}{V_m} \cdot \left( \frac{De}{L^2} \cdot t - \frac{\alpha}{6} \right) \quad \text{----- (1.3.2-2)}$$

The De can be estimated from the slope of Ct as a function of time in steady state based on equation (1.3.2-2). Furthermore, Da can be also derived by the time-lag method [e.g. Muurinen et al., 1987]. The Da is derived from an intercept on the t-axis on the basis of equation (1.3.2-2), the so-called time-lag method [e.g. Muurinen et al., 1987]:

$$Da = \frac{L^2}{6T_{int}} \quad \text{----- (1.3.2-3)}$$

Where

T<sub>int</sub> = intercept on the t-axis

This method is popular to directly obtain De, but there are not so many reports comparing with those in in-diffusion method. It is relatively difficult to carry out, because the chemical conditions of solutions in both cells must be basically maintained constant during the experiment. If both sides of a sample are sandwiched by filters, the correction of De for tracer in the filters is needed, because concentration gradient is formed in the filters. For bentonite, sintered metal filter is generally used between bentonite and solution to restrain the swelling pressure. However, filter is not generally used for rock.

Figure 1.3.2-2 shows an image of the concentration profiles in the bentonite and filters. The concentration gradients in the filters depends on tracer. The true concentration gradient in such case is (C<sub>p1</sub>-C<sub>p2</sub>)/L. Since C<sub>p1</sub> and C<sub>p2</sub> are not directly obtained, De is corrected as follows.

Since all fluxes through filters and bentonite are the same, the following simultaneous equations make good.

$$J = De_f \cdot \left( \frac{C_o - C_{p1}}{L_f} \right) \quad \text{----- (1.3.2-4)}$$

$$J = De \cdot \left( \frac{C_{p1} - C_{p2}}{L} \right) \quad \text{----- (1.3.2-5)}$$

$$J = De_f \cdot \left( \frac{C_{p2} - C_t}{L_f} \right) \quad \text{----- (1.3.2-6)}$$

Where

J = mass flux of the tracer

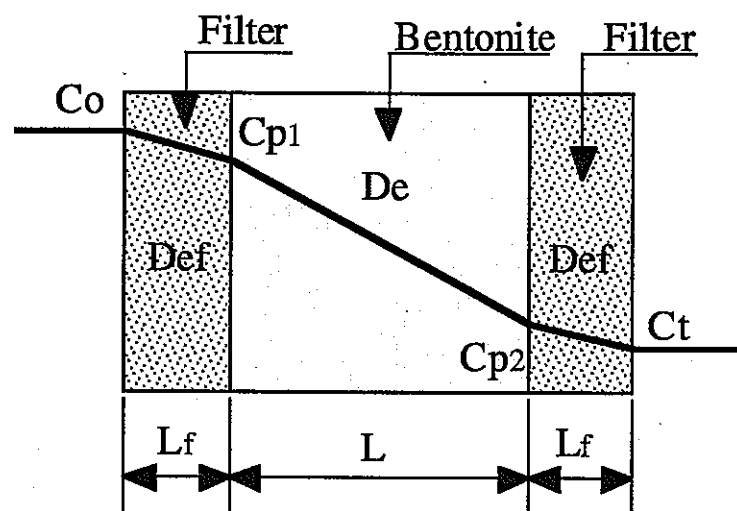
De<sub>f</sub> = effective diffusion coefficient in the filter

C<sub>o</sub> = concentration of the tracer in the tracer cell

C<sub>p1</sub>, C<sub>p2</sub> = concentration of the tracer in the porewater at the border between bentonite and filter

L<sub>f</sub> = thickness of the filter

C<sub>t</sub> = concentration of the tracer in the measurement cell



**Figure 1.3.2-2** Image of concentration profiles in bentonite and filters

In this case, since C<sub>t</sub> is quite lower than C<sub>o</sub>, it can be regarded as zero. Solving these simultaneous equations, De is finally corrected as follows.

$$De = \frac{2De_f \cdot L_f \cdot J - Co}{J \cdot L} \quad \text{----- (1.3.2-7)}$$

In actual experiment, it seems that this correlation is generally not carried out.

Through-diffusion method is of use, because De, Da and α can be simultaneously obtained, but it is inappropriate and impractical for elements which are high sorptive onto geological materials. It takes long times for breakthrough curve to reach steady state for such high sorptive elements.

If concentration profile in the sample can be obtained, both Da and a can be directly calculated.

The  $\alpha$  can be obtained by equation (1.3.1-2) from the concentration at the end of the sample, which contacted with tracer solution in the tracer cell and the concentration of the tracer in the tracer cell. The  $D_a$  can be estimated by the following equation from the slope in concentration distribution of the tracer in the sample and the flux, which can be obtained from the change in concentration of the tracer in the measurement cell in steady state.

$$J = D_a \cdot \left( \frac{\Delta C}{L} \right) \text{-----} (1.3.2-8)$$

Where

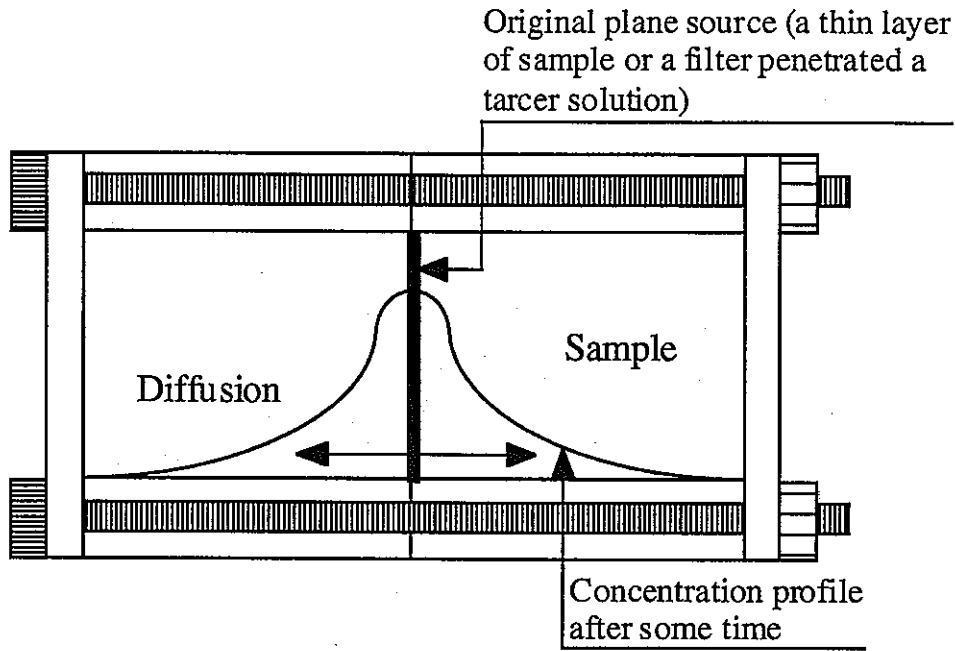
$J$  = mass flux of the tracer

$\Delta C$  = average concentration gradient of the tracer in the sample per unit volume

$L$  = thickness (length) of the sample

### 1.3.3 Back-to-back method

From this method,  $D_a$  can be obtained [e.g. Miyahara et al., 1991; Kozaki et al., 1996; Sato and Yui, 1997]. **Figure 1.3.3-1** shows an illustration of a diffusion experiment by the back-to-back method. A thin layer of sample such as filter penetrated a solution containing a tracer is sandwiched between two cylindrical samples. A small amount of tracer is sometimes pipetted on the surface of the cylindrical sample instead of the thin layer of sample [e.g. Sato et al., 1992b]. After a certain time period, the concentration profile of the tracer in the sample is measured. From this profile,  $D_a$  is obtained based on Fickian second law. For bentonite, the concentration profile is obtained by slicing the bentonite and measuring the concentration of the tracer in each slice. For hard material such as rock, the concentration profile is obtained by scraping the rock and measuring the concentration of the tracer in each scraped material.



**Figure 1.3.3-1** Illustration of a diffusion experiment by back-to-back method

This method is basically the same as in-diffusion method. The difference between both methods is only boundary condition. The later (in-diffusion method) keeps a constant concentration at the boundary. Whilst the concentration of the tracer gradually decreases in the former (back-to-back method), and the tracer acts as a planer source. The  $Da$  can be obtained by the following equation in this case.

**Initial condition**

$$C=0, t=0, |X| \neq 0$$

**Boundary condition**

$$C=0, t > 0, |X| = \infty$$

$$M = \int_{-\infty}^{\infty} C \, dx = \int_{-\infty}^0 C \, dx + \int_0^{\infty} C \, dx = 2 \int_0^{\infty} C \, dx$$

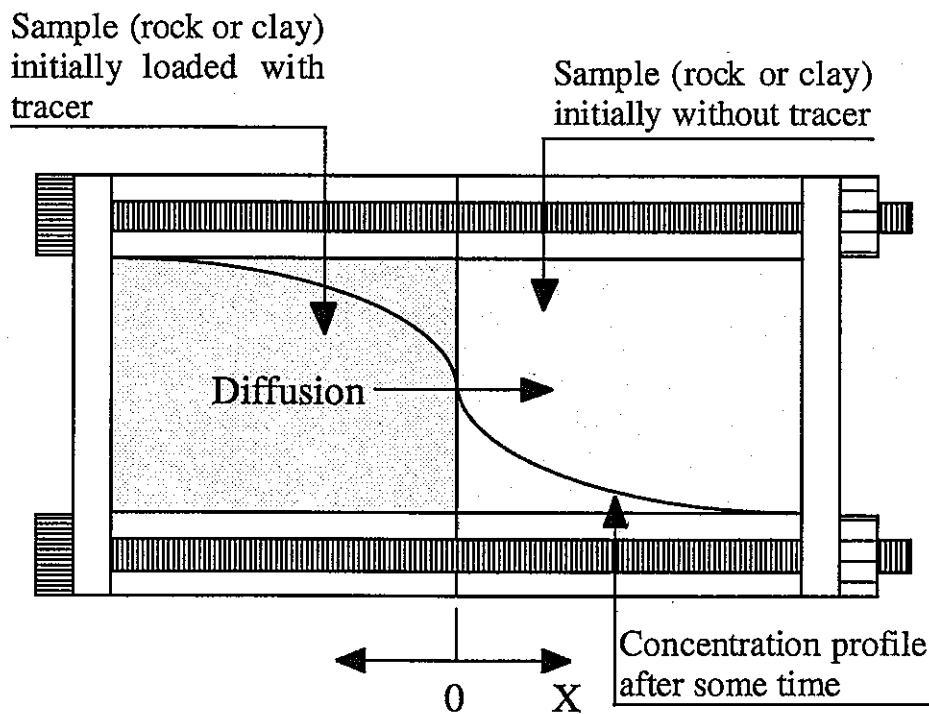
$$C = \frac{M}{2A\sqrt{\pi Da \cdot t}} \cdot \exp\left(-\frac{X^2}{4Da \cdot t}\right) \quad \text{----- (1.3.3-1)}$$

Where

- C = concentration of the tracer per unit sample volume
- M = total quantity of the tracer initially penetrated as a source
- A = cross-section area of the sample
- t = time
- X = distance from the source

As described above, this method is the same as in-diffusion method from the viewpoint of experimental process. It is sometimes inappropriate to distinct between boundary conditions in both experiments. For example, if precipitation of tracer is formed on the surface of sample after the tracer was pipetted, the boundary condition is controlled by the solubility of the tracer. In this case, boundary condition becomes constant,  $Da$  can not be simply estimated by equation (1.3.3-1). The  $Da$  should be estimated by equation (1.3.1-1) in such case, and it is considered to be rather appropriate to regard that the  $Da$  was obtained by in-diffusion method than back-to-back method in this case. Thus it is considered to be nonsense to distinguish between in-diffusion method and back-to-back method. Since the tracer also diffuses into the sample from the outside in the back-to-back method, it is considered that the back-to-back method may be included in the in-diffusion method.

A variant of this method has been also reported. **Figure 1.3.3-2** shows an illustration of a diffusion experiment by a variant of the back-to-back method. In both in-diffusion and back-to-back method, a solution containing a tracer as a source or a thin layer of sample containing a tracer (sometimes a small amount of tracer) is used. However, rock and clay uniformly loaded with a tracer is used as a source in the variant of the back-to-back method. Two samples are firstly prepared. One of the samples is uniformly loaded with a tracer in advance, and the other sample is free of the tracer. When both samples are connected, the diffusion experiment starts. The tracer in the sample containing the tracer gradually diffuses into the other sample and a concentration profile is formed after a certain time period.



**Figure 1.3.3-2** Illustration of a diffusion experiment by a variant of back-to-back method

The Da of the tracer can be estimated by the following equations from the concentration profile in the connected samples [Crank, 1975].

**Initial condition**

$C=C_0, t=0, X<0$

$C=0, t=0, X>0$

**Boundary condition**

$C=0, t>0, X=\infty$

The concentration of the tracer at a distance of  $\epsilon$  from a source with a thickness of  $d\epsilon$  is expressed as follows:

$$C_\epsilon = \frac{C_0 \cdot d\epsilon}{2\sqrt{\pi Da \cdot t}} \cdot \exp\left(-\frac{\epsilon^2}{4Da \cdot t}\right) \quad \text{----- (1.3.3-2)}$$

Therefore, the concentration distribution is calculated as follows.

$$C = \frac{C_0}{2\sqrt{\pi Da \cdot t}} \int_x^\infty \exp\left(-\frac{\epsilon^2}{4Da \cdot t}\right) d\epsilon$$

$$= \frac{C_0}{2\sqrt{\pi Da \cdot t}} \cdot \left\{ \int_x^0 \exp\left(-\frac{\epsilon^2}{4Da \cdot t}\right) d\epsilon + \int_0^\infty \exp\left(-\frac{\epsilon^2}{4Da \cdot t}\right) d\epsilon \right\} \quad \text{----- (1.3.3-3)}$$

Hence, the concentration distribution is finally derived as follows.

$$C = \frac{1}{2} C_0 \cdot \operatorname{erfc}\left(\frac{X}{2\sqrt{Da \cdot t}}\right) = \frac{1}{2} C_0 \cdot \left\{ 1 - \operatorname{erf}\left(\frac{X}{2\sqrt{Da \cdot t}}\right) \right\} \quad \text{----- (1.3.3-4)}$$

Where

$C_\epsilon$  = concentration of the tracer at a distance of  $\epsilon$  from the source with a thickness of  $d\epsilon$

$C$  = concentration of the tracer per unit sample volume

$C_0$  = initial concentration of the tracer in the sample containing the tracer

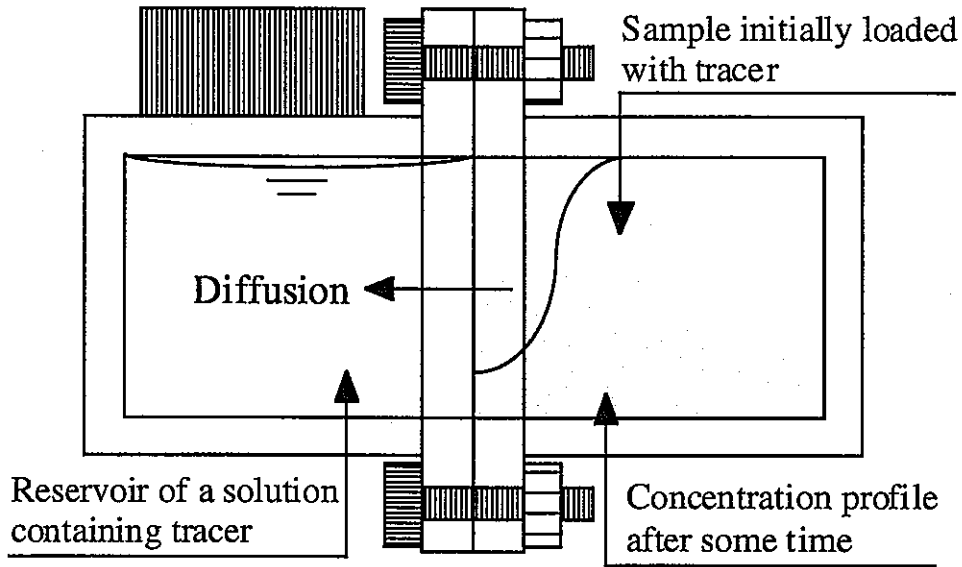
$X$  = distance from the connected plane of the samples

$t$  = time

This is useful for soft materials such as bentonite and clay, but it is difficult to carry out for hard rocks such as granite, because it is too hard to slice or to scrape to obtain concentration profile.



### 1.3.4 Out-diffusion method



**Figure 1.3.4-1** Illustration of a diffusion experiment by out-diffusion method

From this method,  $D_a$  can be obtained. **Figure 1.3.4-1** shows an illustration of a diffusion experiment by the out-diffusion method. This is basically the inverse technique to in-diffusion method [Lever, 1986]. A cylindrical core of sample, which is uniformly loaded with tracer, is contacted with a reservoir of water, which is free of the tracer. The tracer then diffuses out of the sample, and the concentration of the tracer in the reservoir gradually increases with increasing elapsed time. A breakthrough curve can be obtained by monitoring the concentration of the tracer in the reservoir as a function of time.

If the sample is effectively infinite, then the concentration of the tracer in the reservoir at an arbitrary time is expressed by the following equation.

$$\begin{aligned}
 C_t &= \frac{C_o}{\alpha} \cdot \left\{ 1 - \exp\left(\frac{A^2 \cdot De \cdot \alpha \cdot t}{V_r^2}\right) \cdot \operatorname{erfc}\left(\sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{V_r^2}}\right) \right\} \\
 &= \frac{C_o}{\alpha} \cdot \left[ 1 - \exp\left(\frac{A^2 \cdot De \cdot \alpha \cdot t}{V_r^2}\right) \cdot \left\{ 1 - \operatorname{erf}\left(\sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{V_r^2}}\right) \right\} \right] \quad \text{----- (1.3.4-1)}
 \end{aligned}$$

Where

- $C_t$  = concentration of the tracer in the reservoir at an arbitrary time
- $C_o$  = initial concentration of the tracer contained per unit sample volume
- $A$  = cross-section area of the sample
- $t$  = time
- $V_r$  = solution volume in the reservoir

When the concentration in the reservoir at short times is still small (i.e.  $A^2 \cdot De \cdot \alpha \cdot t / Vr^2 \ll 1$ ), equation (1.3.4-1) is approximately expressed by the following equation.

$$C_t = \frac{C_o}{\alpha} \cdot \left\{ 1 - \operatorname{erfc} \left( \sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{Vr^2}} \right) \right\} = \frac{C_o}{\alpha} \cdot \left[ 1 - \left\{ 1 - \operatorname{erf} \left( \sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{Vr^2}} \right) \right\} \right] \quad \text{----- (1.3.4-2)}$$

$$= \frac{C_o}{\alpha} \cdot \operatorname{erfc} \left( \sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{Vr^2}} \right)$$

By the way,

$$\operatorname{erfc}(w) = 1 - \operatorname{erf}(w) = 1 - \frac{2}{\sqrt{\pi}} \int_0^w \exp(-\eta^2) d\eta. \quad \text{----- (1.3.4-3)}$$

The  $\operatorname{erf}(w)$  is expressed as follows from Maclaurin's expansion [e.g. Sato, 1997]:

$$\operatorname{erf}(w) = \frac{2}{\sqrt{\pi}} \cdot \sum_{n=1}^{\infty} \left\{ \frac{(-1)^{n-1}}{(2n-1) \cdot (n-1)!} \cdot w^{2n-1} \right\}. \quad \text{----- (1.3.4-4)}$$

Where

$$w = \sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{Vr^2}}$$

From equation (1.3.4-4), the  $\operatorname{erf}(w)$  is approximately calculated as follows:

$$\operatorname{erf}(w) = \frac{2}{\sqrt{\pi}} \cdot w = \frac{2}{\sqrt{\pi}} \cdot \left( \sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{Vr^2}} \right). \quad \text{----- (1.3.4-5)}$$

Hence, at  $A^2 \cdot De \cdot \alpha \cdot t / Vr^2 \ll 1$ , equation (1.3.4-1) is approximately expressed by the following equation by substituting equation (1.3.4-5) into equation (1.3.4-2).

$$C_t = \frac{2C_o \cdot A}{Vr} \cdot \left( \sqrt{\frac{De \cdot t}{\pi \alpha}} \right) = \frac{2C_o \cdot A}{Vr} \cdot \left( \sqrt{\frac{Da \cdot t}{\pi}} \right) \quad \text{----- (1.3.4-6)}$$

From equation (1.3.4-6),  $Da$  can be estimated. At long times, the concentration of the tracer in the reservoir  $C_t$  as a function of time depends on  $De$  and  $a$  or  $Da$ .

At long times, equilibrium is reached between the tracer in the water in both reservoirs and the

sample and the tracer sorbed to the sample. Then the value of  $\alpha$  can be estimated by the following equation.

$$\alpha = \frac{C_o \cdot V_s - C_p \cdot V_r}{C_p \cdot V_s} = \frac{C_o}{C_p} - \frac{V_r}{V_s} \text{----- (1.3.4-7)}$$

Where

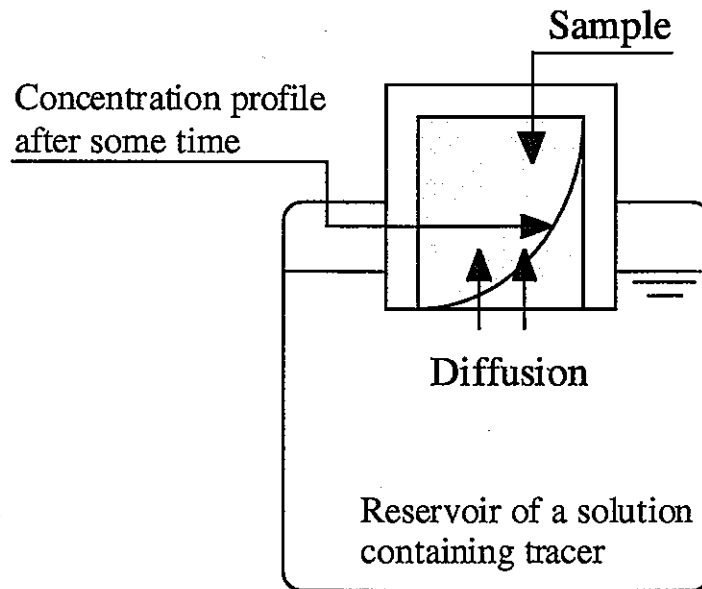
$C_p$  = concentration of the tracer in the reservoir at the equilibrium

$V_s$  = total sample volume

This is a minor method. In the preparation of initial sample, if the tracer is highly sorptive onto the sample, it takes long times for the preparation of sample.

### 1.3.5 Reservoir-depletion method

This method is basically similar to in-diffusion method. **Figure 1.3.5-1** shows an illustration of a diffusion experiment by the reservoir-depletion method. A reservoir of water containing a tracer contacts with one end of a cylindrical core of sample. The other sides are sealed. The tracer diffuses into the sample, and the quantity in the reservoir decreases as a function of time. This decrease is monitored as a function of time.



**Figure 1.3.5-1** Illustration of a diffusion experiment by reservoir-depletion method

If the length of the cylindrical core of the sample is effectively infinite, then the concentration of the tracer in the reservoir satisfies the following equation [Lever, 1986].

$$\begin{aligned}
 C_t &= C_o \cdot \exp\left(\frac{A^2 \cdot De \cdot \alpha \cdot t}{V_r^2}\right) \cdot \operatorname{erfc}\left(\sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{V_r^2}}\right) \\
 &= C_o \cdot \exp\left(\frac{A^2 \cdot De \cdot \alpha \cdot t}{V_r^2}\right) \cdot \left\{1 - \operatorname{erf}\left(\sqrt{\frac{A^2 \cdot De \cdot \alpha \cdot t}{V_r^2}}\right)\right\}
 \end{aligned}
 \tag{1.3.5-1}$$

Where

$C_t$  = concentration of the tracer in the reservoir at an arbitrary time

$C_o$  = initial concentration of the tracer in the reservoir

$A$  = cross-section area of the sample

$t$  = time

$V_r$  = solution volume in the reservoir

In this case, the change in concentration of the tracer depends on the product  $De \cdot \alpha$ . This is because the distance which it has diffused into the sample is proportional to  $(De \cdot t / \alpha)^{0.5}$ , and the quantity of the tracer in the sample is related to  $\alpha$  times as this, i.e. it is determined by  $(De \cdot \alpha \cdot t)^{0.5}$ . If the cylindrical core of the sample is not effectively infinite, then the solution is more complicated. If the sorption is linear, a Laplace transform method can be used. In this case, the solution doesn't just depend on the product  $De \cdot \alpha$  [Lever, 1986].

The concentration of the tracer doesn't continue to decrease indefinitely, and at long times equilibrium is reached between the tracer in the water in both reservoir and sample and the tracer sorbed to the sample. Then the value of  $\alpha$  can be estimated by the following equation from this equilibrium.

$$\alpha = \frac{(C_o - C_p) \cdot V_r}{C_p \cdot V_s} = \left(\frac{C_o}{C_p} - 1\right) \cdot \frac{V_r}{V_s}
 \tag{1.3.5-2}$$

Where

$C_p$  = concentration of the tracer in the reservoir at the equilibrium

$V_s$  = total sample volume

From this method, although neither  $De$  nor  $Da$  can be directly obtained,  $De$  and  $Da$  can be analytically estimated from equations (1.3.5-1) and (1.3.5-2).

### 1.3.6 Electrical conductivity method

From this method,  $D_e$  can be basically obtained. **Figure 1.3.6-1** shows an illustration of a diffusion experiment by the electrical conductivity method. A core of sample or a block of sample is firstly saturated with an electrolyte solution such as NaCl, of which electrolyte concentration is known. Then the electrical conductivity between two opposite planes (electrodes) in the sample is measured. For this measurement, the Wheatstone bridge circuit is generally used. When the Wheatstone bridge circuit gives the smallest capacitance and the smallest electric current, the relation between resistances is given by the following equation.

$$\frac{R}{R_2} = \frac{R_1}{R_3} \quad \text{----- (1.3.6-1)}$$

Where

$R$  = resistance of the conductance cell

$R_1, R_2, R_3$  = resistances in the bridge circuit

From the measured resistance, the electrical conductivity is calculated by dividing the cell constant by the measured resistance. The equivalent conductivity is calculated from the obtained electrical conductivity, and then  $D_e$  can be estimated by the Nernst expression [Robinson and Stokes, 1959]. In this case,  $D_e$  is also called a simple diffusion coefficient ( $D_{si}$ ) [e.g. Conca et al., 1991]. If the porosity  $\phi$  is 100%,  $D_e$  (or  $D_{si}$ ) is equal to  $D_0$ . The  $D_e$  in this method is estimated by the following equation [e.g. Conca and Wright, 1990].

$$D_e = \frac{R \cdot T \cdot K \cdot t_{ni}}{F^2 \cdot |Z_i| \cdot C_i} \quad \text{----- (1.3.6-2)}$$

Where

$R$  = gas constant

$K$  = specific conductance

$t_{ni}$  = transport or transference number of the  $i$ -th ion

$|Z_i|$  = absolute value of the charge number of the  $i$ -th ion in the electrolyte

$C_i$  = molar concentration of the  $i$ -th ion in the electrolyte

The specific conductance  $K$  is disintegrated as follows.

$$K = \Theta \cdot G \quad \text{----- (1.3.6-3)}$$

Where

$\Theta$  = cell constant for the conductivity sample holder

$G$  = measured conductance on the conductivity bridge

Therefore, finally, equation (1.3.6-2) is expressed as follows from equation (1.3.6-3).

$$De = \frac{R \cdot T \cdot \Theta \cdot G \cdot t_{ni}}{F^2 \cdot |Z_i| \cdot C_i} \text{----- (1.3.6-4)}$$

There is an activity correction factor that corrects for solution nonideality at higher ionic strengths. The  $De$  in this case is corrected by the following equation [e.g. Conca and Wright, 1990].

$$De = Deo \cdot \left( 1 + \frac{d \ln a}{d \ln C} \right) \text{----- (1.2.6-5)}$$

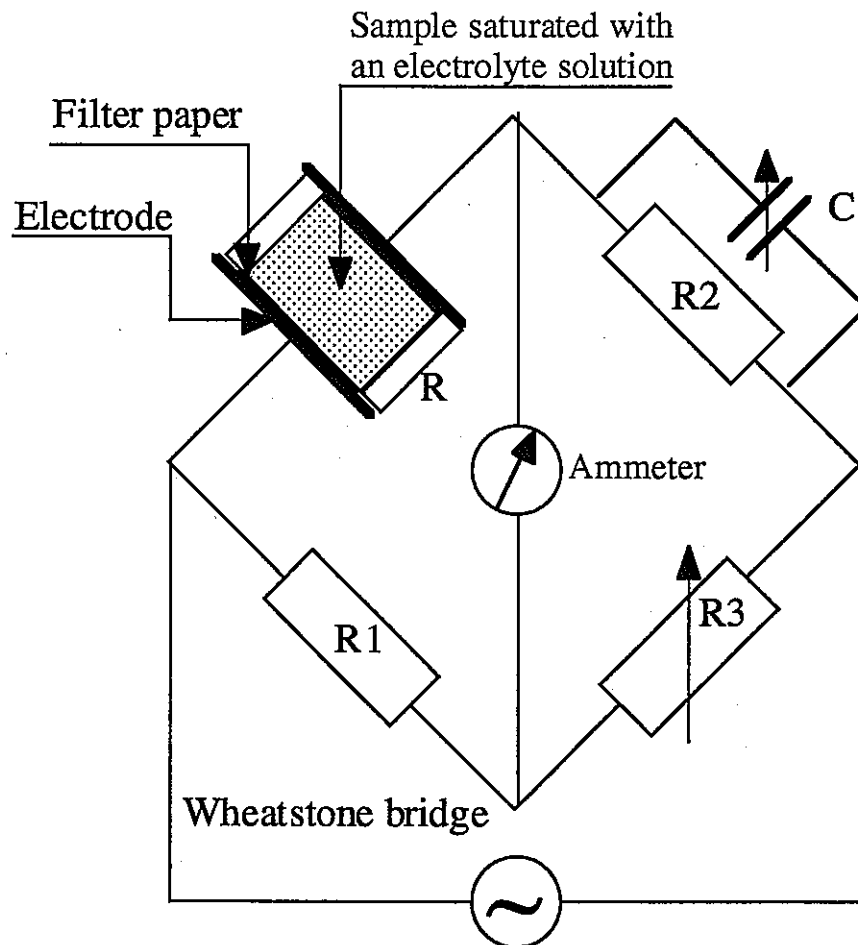
Where

$Deo$  = effective diffusion coefficient at infinite dilution

$a$  = mean molar activity coefficient

$C$  = solute concentration

This correction factor is also calculated for simple electrolytes from data given in standard references. For example, for a 0.1M KCl solution, the activity coefficient was 0.76 using the extended Debye-Hückel approximation [Conca and Wright, 1990], giving a correction factor of approximately 1.12 based on equation (1.2.6-5), which is applied to the diffusion coefficients calculated from equation (1.3.6-4). For a 0.01M KCl solution, the correction factor is calculated 1.04 [Conca and Wright, 1990].



**Figure 1.3.6-1** Illustration of a diffusion experiment by electrical conductivity method

## 1.4 Basic Concept for Data Extraction for Performance Assessment

### 1.4.1 Categorization of rock

Rock has been mainly categorized into 4 kinds of rocks; acid crystalline rock, alkaline crystalline rock, sedimentary rock (argillaceous/tuffaceous rock) and sedimentary rock (psammitic rock/sandy stone) from the viewpoints of geology, hydrogeology [PNC, 1998] and mass transport. The crystalline rock is generally regarded as fractured media and the sedimentary rock is generally regarded as porous media. Furthermore, the sedimentary rock has been separated into two ages; rocks before paleogene and those after neogene, because their physical properties such as porosity and the degree of consolidation significantly depend on age, which those rocks were formed. However, in actual separation for the sedimentary rock, it is difficult to distinctly distinguish between both ages, because few reports describe detailed information for the age of the

rock sample used in the experiment, and it is too short in information to judge. Therefore, the distinction of age in the sedimentary rock is not carried out in this report.

For categorized rocks, granite, granodiorite, rhyolite, etc. are included in acid crystalline rock. Basalt, andesite, schist, diorite, gabbro, etc. are included in alkaline crystalline rock. Mudstone, tuff, pelitic schist, shale, etc. are included in sedimentary rock (argillaceous/tuffaceous rock), and sandstone, siliceous sedimentary rock, etc. are included in sedimentary rock (psammitic rock/sandy stone) in this report.

Limestone is also included in sedimentary rock, but it is omitted from the development of diffusivity database, because limestone can become one of the natural resources and is inappropriate as a host rock.

## 1.4.2 Basic concept for data extraction

- (1) Since geological disposal is focused on Japanese geological environment, data for diffusion are collected focused on Japanese major rocks.
- (2) Not only 22 elements which are considered to be important in performance assessment of the geological disposal of HLW, but also all elements and aquatic tracers are treated in the development of database considering general purpose, because it is considered that rock pore sizes are quite large compared with ionic sizes and that electrostatic interaction of diffusion species with the rock surfaces is small. In this case,  $D_e$  strongly depends on  $D_0$  of species and the formation factor of rock, which quantitatively expresses geometric pore structure.
- (3) Since it is difficult to distinguish between rocks before paleogene and those after neogene for sedimentary rock, in which matrix diffusion data have been reported, the distinction of age is not carried out. Therefore, the development of database is basically focused on 4 kinds of rocks; acid crystalline rock, alkaline crystalline rock, sedimentary rock (argillaceous/tuffaceous rock) and sedimentary rock (psammitic rock/sandy stone).
- (4) Since limestone which belongs to sedimentary rock can become one of the natural resources and is inappropriate as a host rock, it is omitted in the development of database.

## 1.4.3 Basic structure of database

Figure 1.4.3-1 shows the basic structure of the database for diffusion coefficient. The whole database is composed of sub-databases of 4 kinds of rocks. Furthermore, the sub-databases of 4



kinds of the rocks are composed of databases to individual elements, in which each datum, literature information, remarks and so on are input. Totally, 24 parameters (or items) such as element, species, rock name, diffusion coefficients (De, Da, Do) and obtained conditions (method, porewater, pH, Eh, temperature, atmosphere, etc.) are input.

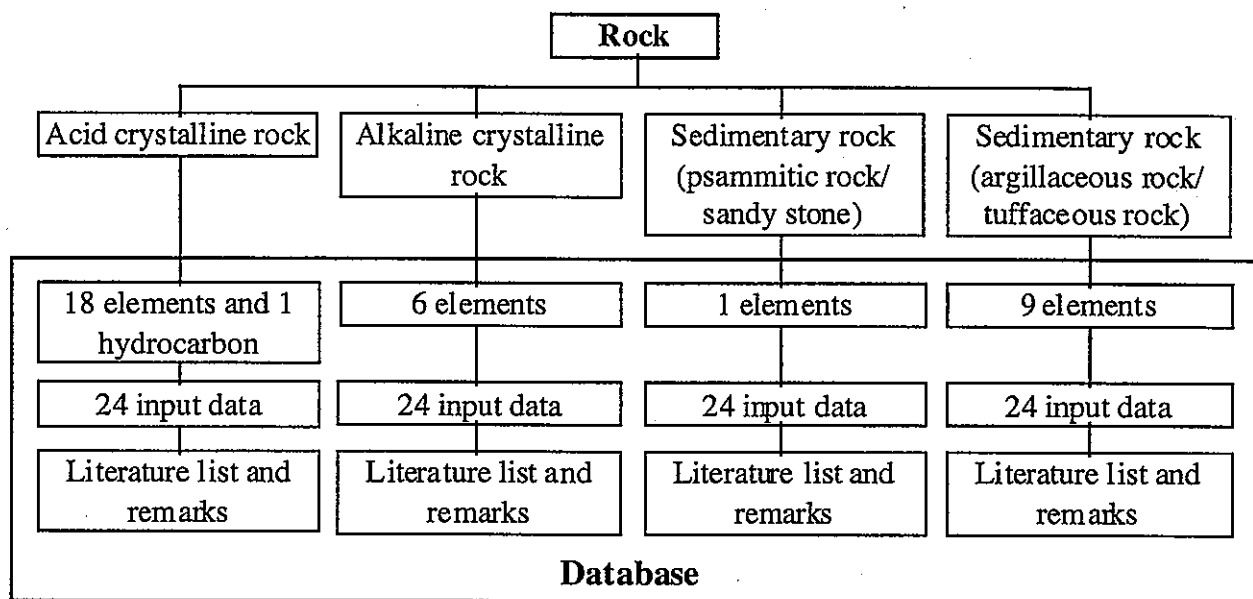


Figure 1.4.3-1 Basic structure of database for diffusion coefficient

### 1.4.4 Literature survey

Data regarding diffusion (De, Da, obtained conditions, etc.) in domestic rocks were collected based on literature survey. A review of literatures was carried out focused on literatures such as journals (domestic and international), proceedings (domestic and international) and technical reports (such as JAERI, PNC, etc.) already published between 1980 and 1998.

## 1.5 References

- Albinsson, Y. and Engkvist, I. (1991): Diffusion of Am, Pu, U, Np, Cs, I and Tc in Compacted Sand-Bentonite Mixture, *Radioactive Waste Management Nuclear Fuel Cycle*, **15** (4), pp.221~239.
- Atomic Energy Commission (1994): Long-Term Program for Research, Development and Utilization of Atomic Energy.
- Brandberg, F. and Skagius, K. (1991): Porosity, Sorption and Diffusivity Data Compiled for the SKB91-16 Study, SKB Technical Report, 91-16.
- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Cheung, S. C. and Gray, M. N. (1989): Mechanism of Ionic Diffusion in Dense Bentonite, *Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XII* (Lutze, W. and Ewing, R. C. eds.), Vol.127, pp.677~681.
- Christiansen, B. and Torstenfelt, B. (1988): Diffusion of Nickel, Strontium, Iodine, Cesium and Americium in Loosely Compacted Bentonite at High pH, *Radiochimica Acta*, **44/45**, pp.219~223.
- Conca, J. L. and Wright, J. (1990): Diffusion Coefficients in Gravel Under Unsaturated Conditions, *Water Resources Research*, Vol.26, No.5, pp.1055~1066 (May 1990).
- Conca, J. L., Ashida, T. and Sato, H. (1991): Apparent and Simple Diffusion Coefficients in Compacted Bentonite, *Proceedings of the Second Annual International Conference on the High Level Radioactive Waste Management*, Vol.2, pp.1382~1389, Las Vegas, Nevada, U.S.A.
- Conca, J. L. and Wright, J. (1992): Direct Determinations of Unsaturated Flow and Transport, *Proceedings of the Twelfth Annual American Geophysical Union Hydrology Days Meeting*, Fort Collins, March 1992, pp.103~116.
- Conca, J. L., Apted, M. J. and Arthur, R. C. (1993): Direct Determinations of Transport Parameters in Repository Materials, *Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVI* (Interrante, C. G. and Pabalan, R. T. eds.), Vol.294, pp.839~844.
- Crank, J. (1975): *The Mathematics of Diffusion*, 2nd ed., Pergamon Press, Oxford, UK.
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only

by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.

- Eriksen, T., Jacobsson, A. and Pusch, R. (1981): Ion Diffusion through Highly Compacted Bentonite, SKBF/KBS TR 81-05.
- Eriksen, T. and Jacobsson, A. (1996): Diffusion of  $I^-$ ,  $Cs^+$  and  $Sr^{2+}$  in Compacted Bentonite - Anion Exclusion and Surface Diffusion-, SKB Technical Report, 96-16.
- Idemitsu, K., Ishiguro, K., Yusa, Y., Sasaki, N. and Tsunoda, N. (1990): Plutonium Diffusivity in Compacted Bentonite, Engineering Geology, 28, pp.455~462.
- Ishida, H., Yamada, N., Ueta, S., Muroi, M., Sato, H. and Yui, M. (1991): Estimation of Effective Diffusivities in Buffer Material based on a Formation Factor, 1991 Fall Meeting of the Atomic Energy Society of Japan, J24 [In Japanese].
- Ito, M., Okamoto, M., Shibata, M., Sasaki, Y., Danbara, T., Suzuki, K. and Watanabe, T. (1993): "Bentonaito-no-kobutsu-osei-bunseki" (Mineral Composition Analysis of Bentonite), PNC Technical Report, PNC TN8430 93-003 [In Japanese].
- Ito, M., Okamoto, M., Suzuki, K., Shibata, M. and Sasaki, Y. (1994): "Bentonaito-no-kobutsu-osei-bunseki" (Mineral Composition Analysis of Bentonite), Journal of the Atomic Energy Society of Japan, Vol.36, No.11, pp.1055~1058 [In Japanese].
- Kato, H., Muroi, M., Yamada, N., Ishida, H. and Sato, H. (1995): Estimation of Effective Diffusivity in Compacted Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVIII (Murakami, T. and Ewing, R. C. eds.), Vol.353, pp.277~284.
- Kita, H., Iwai, T. and Nakashima, S. (1989): Diffusion Coefficient Measurement of an Ion in Pore Water of Granite and Tuff, Journal of the Japan Society of Engineering Geology, 30 (2), pp.26~32 [In Japanese].
- Kosaka, T., Sato, S., Furuya, H., Araya, S. and Matsuo, K. (1984): Diffusion Behavior of Uranium in a Rock Block, 1984 Fall Meeting of the Atomic Energy Society of Japan, G82 [In Japanese].
- Kozaki, T., Sato, H., Fujishima, A., Sato, S. and Ohashi, H. (1996): Activation Energy for Diffusion of Cesium in Compacted Sodium Montmorillonite, Journal of Nuclear Science and Technology, Vol.33, No.6, pp.522~524.
- Kumpulainen, H., Muurinen, A. and Uusheimo, K. (1991): Diffusion Porosity and Diffusivity of Radionuclides in Granitic Rocks. Part II: Diffusivity and Electrical Resistivity Measurements in Rock Matrix, Materials Research Society Symposium Proceedings on the

- Scientific Basis for Nuclear Waste Management XIV (Abrajano, Jr., T. and Johnson, L. H. eds.), Vol.212, pp.655~660.
- Kunimine Industries Co. Ltd. (private communication): "Bentonaito-no-kihontokusei-ichiranhyo" (A Table for Basic Properties of Bentonites).
- Lever, D. A. (1986): Some Notes on Experiments Measuring Diffusion of Sorbed Nuclides through Porous Media, Technical Report of the United Kingdom Atomic Energy Authority Harwell, AERE R 12321.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, *Geochimica et Cosmochimica Acta*, Vol.38, pp.703~714.
- Matsuo, S. ed. (1990): "Chikyu-kagaku" (Geochemistry), Kodansya [In Japanese].
- McKinley, I. G. and Hadermann, J. (1984): Nuclide Sorption Database for Swiss Safety Assessment, NAGRA Technical Report, NTB 84-40.
- Miyahara, K., Ashida, T., Kohara, Y., Yusa, Y. and Sasaki, N. (1991): Effect of Bulk Density on Diffusion for Cesium in Compacted Sodium Bentonite, *Radiochimica Acta*, 52/53, pp.293~297.
- Monicard, R. P. (1980): Properties of Reservoir Rocks: Core Analysis, Editions Technip, Paris, France.
- Müller-Vonmoos, M. and Kahr, G. (1983): Mineralogische Untersuchungen Von Wyoming Bentonit MX-80 und Montigel, NAGRA Technical Report, NTB83-12 [In German].
- Muurinen, A., Rantanen, J., Ovaskainen, R. and Heinonen, O. J. (1983): Diffusion Measurements in Concrete and Compacted Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management VI (Brookins, F. G. ed.), Vol.15, pp.777~784.
- Muurinen, A., Rantanen, J. and Penttilä-Hiltunen, P. (1985a): Diffusion Mechanisms of Strontium, Cesium and Cobalt in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management IX (Werme, L. O. ed.), Vol.50, pp.617~624.
- Muurinen, A., Rantanen, J. and Penttilä-Hiltunen, P. (1985b): Diffusion of Cesium and Strontium in Compacted and in Mixture of Sodium Bentonite and Crushed Rock, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management VIII (Jantzen, C. J., Stone, J. A. and Ewing, R. C. eds.), Vol.44, pp.883~890.

- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Muurinen, A., Penttilä-Hiltunen, P. and Uusheimo, K. (1989): Diffusion of Chloride and Uranium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XII (Lutze, W. and Ewing, R. C. eds.), Vol.127, pp.743~748.
- Muurinen, A. (1994): Diffusion of Anions and Cations in Compacted Sodium Bentonite, VTT Publications 168, Technical Research Center of Finland.
- Nakano, M. (1991): "Tsuchi-no-bussitsu-idogaku" (Mass Transport of Soil), The University of Tokyo, Japan [In Japanese].
- Nakazawa, T., Sato, S. and Ohashi, H. (1997): Electrical Conductivity of Ions in Comacted Sodium Montmorillonite, 1997 Fall Meeting of the Atomic Energy Society of Japan, L13 [In Japanese].
- Nishiyama, K., Nakashima, S., Tada, R. and Uchida, T. (1990): Diffusion of an Ion in Rock Pore Water and Its Relation to Pore Characteristics, Mining Geology, 40 (5), pp.323~336.
- Nowak, E. J. (1983): Diffusion of Radionuclides in Brine-saturated Backfill Barrier Materials, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management VI (Brookins, D. G. ed.), Vol.15, pp.735~744.
- Ochs, M., Boonekamp, M., Wanner, H., Sato, H. and Yui, M. (1998): A Quantitative Model for Ion Diffusion in Compacted Bentonite, Radiochimica Acta, 82, pp.437~443.
- Okamoto, M., Takase, T. and Higashi, K. (1994): Experimental Study on Cation Diffusivities in Homoionized Montmorillonite by Electrical Conductance Model, 1994 Fall Meeting of the Atomic Energy Society of Japan, K40 [In Japanese].
- Okamoto, M., Takase, T. and Higashi, K. (1995): Experimenta Study on Cation Diffusivities in Homoionized Montmorillonite by Electrical Conductance Model, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVIII (Murakami, T. and Ewing, R. C. eds.), Vol.353, pp.285~290.
- Olin, M. (1994): Diffusion in Crushed Rocks and in Bentonite Clay, VTT Publications 175, Technical Research Centre of Finland.
- Ota, K. and Yoshida, H. (1994): Influence of Microscopic Heterogeneity on Diffusion for Sedimentary Rocks, Proceedings of an NEA/SKI Symposium, Paris, France, October 11-14,

1994, pp.237~243.

- Ota, K., Amano, K. and Ando, T. (1998): In situ Nuclide Retardation in a Fractured Crystalline Rock, Kamaishi In situ Test Site, Kamaishi International Workshop Proceedings, Kamaishi, Japan, August 24-25, 1998, pp.III-2-1~III-2-8.
- Park, C. K., Han, K. W. and Park, H. H. (1991): Determination of Diffusivities of Sorbing and Non-sorbing species in Rocks, High Level Radioactive Waste Management, Proceedings of the Second Annual International Conference, Vol.1, pp.156~160.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1991): Current Status for Research and Development of Geological Disposal -FY1989-, PNC Technical Report, PNC TN1410 91-009 [In Japanese].
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1992): Research and Development on Geological Disposal of High-Level Radioactive Waste -First Progress Report-, PNC Technical Report, PNC TN1410 93-059.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1998): The First Draft of the Second Progress Report, PNC Technical Report, PNC TN1412 98-013 (September 2, 1998) [In Japanese].
- Radioactive Waste Management Center (RWMC) (1990): Radionuclides Distribution Coefficient of Soil-Environmental Parameters Series 2 [In Japanese].
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Sasaki, Y., Shibata, M., Yui, M. and Ishikawa, H. (1995): Experimental Studies on the Interaction of Some Radionuclides in Compacted Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVIII (Murakami, T. and Ewing, R. C. eds.), Vol.353, pp.337~344.
- Sato, H., Ashida, T., Shibutani, T., Kohara, Y., Uchidate, N. and Yui, M. (1991): Studies on Diffusion and Sorption Mechanism of Radioactive Nuclides in Kunigel-V1, 1991 Fall Meeting of the Atomic Energy Society of Japan, J25 [In Japanese].
- Sato, H., Ashida, T., Kohara, Y., Yui, M., Umeki, H. and Ishiguro, K. (1992a): Effective Diffusion Coefficients of Nuclides in Bentonite and Rocks, PNC Technical Report, PNC TN8410 92-164 [In Japanese].
- Sato, H., Ashida, T., Kohara, Y., Yui, M. and Sasaki, N. (1992b): Effect of Dry Density on Diffusion of Some Radionuclides in Compacted Sodium Bentonite, Journal of Nuclear

- Science and Technology, 29 (9), pp.873~882.
- Sato, H., Ashida, T., Kohara, Y. and Yui, M. (1993): Study on Retardation Mechanism of  $^3\text{H}$ ,  $^{99}\text{Tc}$ ,  $^{137}\text{Cs}$ ,  $^{237}\text{Np}$  and  $^{241}\text{Am}$  in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVI (Interrante, C. G. and Pabalan, R. T. eds.), Vol.294, pp.403~408.
- Sato, H. and Shibutani, T. (1994): Study on Adsorption and Diffusion Mechanism of Nuclides in Buffer Material and Geosphere, PNC Technical Review, No.91, PNC TN8410 94-284, pp.71~89 [In Japanese].
- Sato, H. and Yui, M. (1995): Diffusion Behaviour for Se and Zr in Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVIII (Murakami, T. and Ewing, R. C. eds.), Vol.353, pp.269~276.
- Sato, H. (1996): Through-Diffusion Experiments on NaCl through Tuff -Measurements of Tortuosity-, PNC Technical Report, PNC TN8410 96-111.
- Sato, H., Yui, M. and Yoshikawa, H. (1996): Ionic Diffusion Coefficients of  $\text{Cs}^+$ ,  $\text{Pb}^{2+}$ ,  $\text{Sm}^{3+}$ ,  $\text{Ni}^{2+}$ ,  $\text{SeO}_4^{2-}$  and  $\text{TcO}_4^-$  in Free Water Determined from Conductivity Measurements, Journal of Nuclear Science and Technology, Vol.33, No.12, pp.950~955.
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119~133, Elsevier Science B. V.
- Sato, H. and Yui, M. (1997): Diffusion of Ni in Compacted Sodium Bentonite, Journal of Nuclear Science and Technology, Vol.34, No.3, pp.334~336.
- Sato, H. (1997): A Study on Nuclide Migration in Buffer Materials and Rocks for Geological Disposal of Radioactive Waste, Doctoral Thesis of Akita University [In Japanese].
- Sato, H. (1998): Data Setting for Effective Diffusion Coefficients ( $D_e$ ) of Nuclides in the Buffer for Reference Case in Performance Assessment of the Geological Disposal of High-Level Radioactive Waste (I) -A Dataset of  $D_e$  Values for Fresh-Reducing-High pH Groundwater (FRHP)-, PNC Technical Report, PNC TN8410 98-097.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.
- Sharma, H. and Oscarson, D. W. (1989): Diffusion of Plutonium (IV) in Dense Bentonite-Based Materials, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XII (Lutze, W. and Ewing, R. C. eds.), Vol.127, pp.735~741.

- Skagius, K. and Neretnieks, I. (1982): Diffusion in Crystalline Rocks of Some Sorbing and Nonsorbing Species, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management V (Lutze, W. ed.), Vol.11, pp.509~518.
- Skagius, K. and Neretnieks, I. (1985a): Porosities and Diffusivities of Some Non-Sorbing Species in Crystalline Rocks, SKB Technical Report, 85-03, Swedish Nuclear Fuel and Waste Management Co., Stockholm.
- Skagius, K. and Neretnieks, I. (1985b): Diffusion in Crystalline Rocks of Some Sorbing and Nonsorbing Species, KBS 82-12.
- Suzuki, H. Shibata, M., Yamagata, J., Hirose, I. and Terakado, K. (1992): The Tests of Properties for Buffer Materials (I), PNC Technical Report, PNC TN8410 92-057 [In Japanese].
- Tamamushi, R. (1974): "Denki-kagaku" (Electrochemistry), Tokyo-kagaku-dojin [In Japanese].
- Tochiyama, O., Takasu, A., Ikeda, T., Kimura, H., Sato, S., Nagasaki, S., Nkayama, S., Niibori, Y., Furuya, H., Mitsugashira, T. and Yamaguchi, T. (1998): Concept and Applicability of Sorption Distribution Coefficients in the Radionuclide Transport Model, Journal of Nuclear Fuel Cycle and Environment, Vol.5, No.1, pp.3~19 [In Japanese].
- Torstenfelt, B., Andersson, K., Kipatsi, H., Allard, B. and Olofsson, U. (1982a): Diffusion Measurements in Compacted Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Waste Management (Topp, S. G. ed.), Vol.6, pp.295~302.
- Torstenfelt, B., Andersson, K., Kipatsi, H., Allard, B. and Olofsson, U. (1982b): Transport of Actinides through a Bentonite Backfill, Materials Research Society Symposium Proceedings on the Scientific Basis for Waste Management V (Lutze, W. ed.), Vol.11, pp.659~668.
- Torstenfelt, B. and Allard, B. (1986): Migration of Fission Products and Actinides in Compacted Bentonite, SKBF/KBS TR 86-14.
- Tsukamoto, M. and Ohe, T. (1988): Diffusion of Radioactive Cesium into Granitic Core Sample, 1988 Fall Meeting of the Atomic Energy Society of Japan, J45, p.169 [In Japanese].
- Umeki, H., Suzuki, A. and Kiyose, R. (1986): Nuclide Migration in Granitic Rock, 1986 Fall Meeting of the Atomic Energy Society of Japan, October 16-18, 1986, Kyusyu University, H63, p.137 [In Japanese].
- Van Brakel, J. and Heertjens, P. M. (1974): Analysis of Diffusion in Macroporous Media in terms



of a Porosity, a Tortuosity and a Constrictivity Factor, International Journal of Heat Mass Transfer, Vol.17, pp.1093~1103, Pergamon Press, UK.

Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1992): Surface Diffusion of Strontium in Rocks, 1992 Annual Meeting of the Atomic Energy Society of Japan, C19, p.119 [In Japanese].

Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1993): Consideration on Effective Diffusivities of Strontium in Granite, Journal of Nuclear Science and Technology, **30** (8), pp.796~803.

Yamaguchi, T., Sakamoto, Y., Nakayama, S. and Vandergraaf, T. T. (1997): Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, Journal of Contaminant Hydrology, **26**, pp.109~117, Elsevier Science B. V.

Yui, M., Sasaki, Y., Shibutani, T. and Sato, H. (1994): An Approach to Understanding Bentonite-Water Interaction and Nuclides Migration in Bentonite, Radioactive Waste Research, Vol.1, No.1, pp.33~42 [In Japanese].

# Chapter 2 Diffusivity Data of Nuclides in Matrix Part of Domestic Major Rocks

## 2.1 Sedimentary Rock: argillaceous/tuffaceous rock

Totally, 54 De and 40 Da data have been found for argillaceous or tuffaceous rock among sedimentary rock to date based on a review of literatures. Nine elements<sup>1</sup> have been totally discussed for argillaceous and tuffaceous rock. In this section, data regarding matrix diffusion obtained for individual elements are introduced, as well as are discussed.

### 2.1.1 Chlorine (Cl)

The De values for Cl (Cl<sup>-</sup>) in sedimentary rock have been reported for mudstone [Sato and Yui, 1992; Sato et al., 1992; PNC, 1996; Sato et al., 1997], pelitic schist [Sato, 1991] and tuff [Sato, 1996]. All measurements have been carried out using non-radioactive tracer. Chlorine is not directly included in important elements for performance assessment of the geological disposal of HLW, but it is generally regarded as a non-sorbing element. Chlorine (<sup>36</sup>Cl) is accounted as one of the important elements for performance assessment in low-level radioactive waste (LLW). The Da values have been also derived based on the time-lag method [e.g. Muurinen et al., 1987] or based on De and rock capacity factor.

For mudstone, tuff and pelitic schist, totally, 13 De values, obtained by through-diffusion experiments, have been reported up to the present. All measurements have been carried out under aerobic conditions, because Cl is not redox sensitive. The porewater initially saturated is all distilled water. All measurements have been performed at room temperature or 25 °C. No study for systematic temperature dependency on De has been reported. The reported porewater pH was in a range of 5.5 to 9.54. Since Cl simply takes chloride ion in a wide pH range, the change of the porewater pH is not directly concerned with chemical behaviour.

Figures 2.1.1-1 and 2.1.1-2 show histograms for De and Da values of Cl<sup>-</sup> in sedimentary rock reported to date, respectively. Totally, 13 De data have been reported for mudstone, tuff and pelitic schist. Relatively a wide range of variation is found in De. This is considered to be due to the difference of porosity. The De was in the order from highest to lowest, tuff, pelitic schist and mudstone. For Da, 7 data have been reported in total. A wide range of variation is not found for Da different from De. This is presumed to be due to the difference of sorptivity onto rocks.

---

<sup>1</sup> 9 elements to be introduced: Cl, Cs, I, K, Na, Ni, Se, Sm and U.

Figures 2.1.1-3 and 2.1.1-4 show plots of  $D_e$  and  $D_a$  values of  $Cl^-$  as a function of rock porosity for sedimentary rock, respectively. The correlations were plotted only for data reported both porosity and  $D_e$  or  $D_a$ . For  $D_e$ , 12 plots have been totally reported and 6 plots have been totally reported for  $D_a$ . The  $D_e$  clearly shows a tendency to increase with increasing rock porosity. While significant porosity dependence is not found for  $D_a$ , but it seems that the lower porosity is, the lower  $D_a$  is.

The  $D_o$  of  $Cl^-$  is estimated to be  $2.0 \times 10^{-9} \text{ m}^2/\text{s}$  at  $25^\circ \text{C}$  by the Nernst-Einstein equation (it is also called the Nernst expression)[Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991; Marcus, 1997]. Formation factor (FF) was calculated based on the equation:  $FF = D_e/D_o$ . Moreover, geometric factor (Gf) was also calculated based on the equation:  $Gf = D_p/D_o = D_e/(\phi \cdot D_o) = FF/\phi$ . Where  $D_p$  is the porewater diffusion coefficient, and  $\phi$  is the porosity. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.1.10.

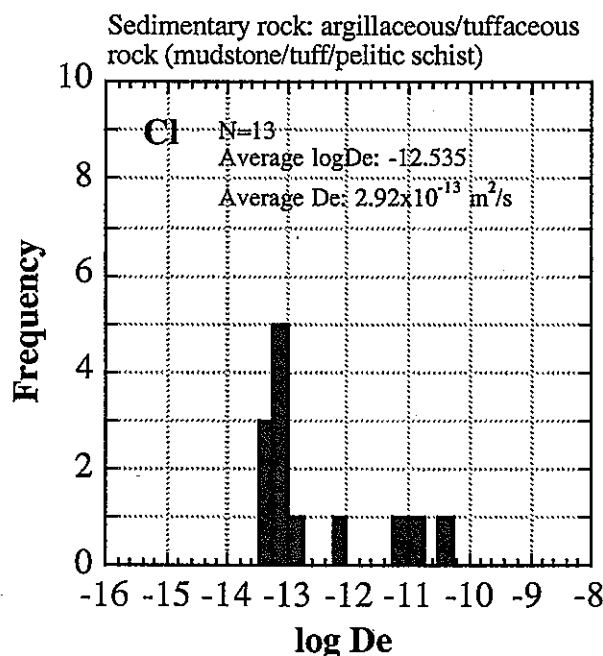
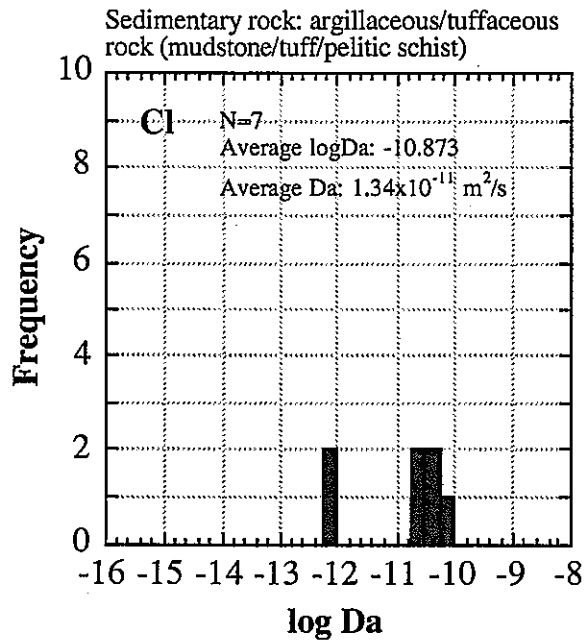
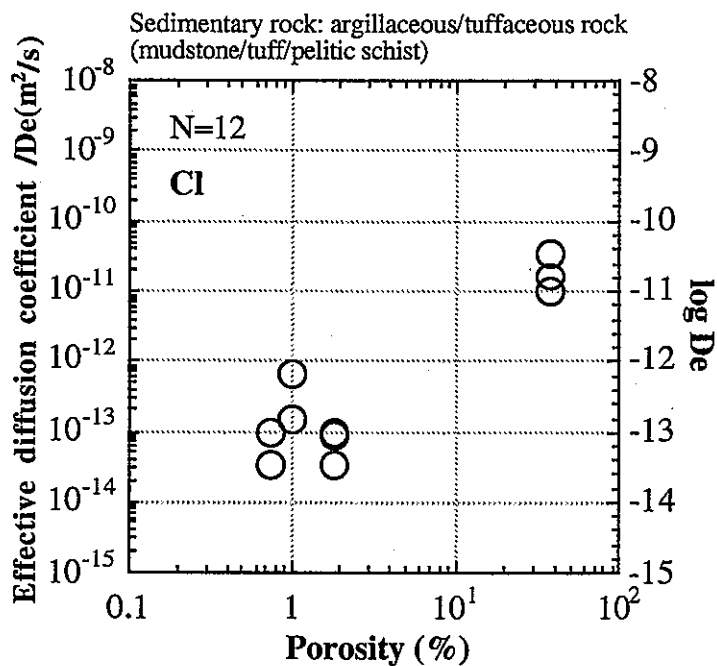


Figure 2.1.1-1 Histogram for  $D_e$  values of  $Cl^-$  ion in sedimentary rock (mudstone, tuff, pelitic schist)

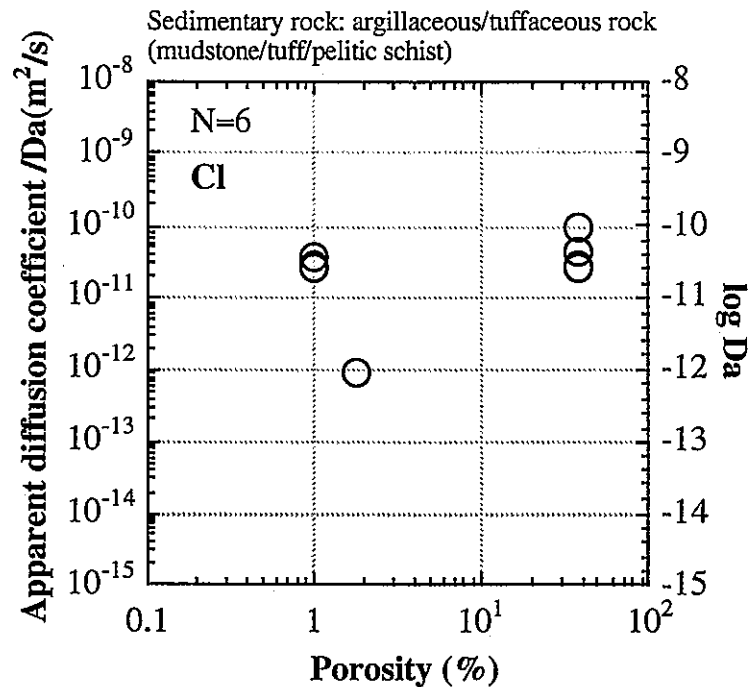
The average  $D_e$  of  $Cl^-$  in sedimentary rock was estimated to be  $2.92 \times 10^{-13} \text{ m}^2/\text{s}$ . Though a wide range of variation is found in  $D_e$ , this is considered to be due to a wide porosity range in the rock.



**Figure 2.1.1-2** Histogram for Da values of Cl<sup>-</sup> ion in sedimentary rock (mudstone, tuff, pelitic schist)  
The average Da of Cl<sup>-</sup> in sedimentary rock was estimated to be 1.34x10<sup>-11</sup> m<sup>2</sup>/s.



**Figure 2.1.1-3** De values of Cl<sup>-</sup> ion as a function of rock porosity for sedimentary rock (mudstone, tuff, pelitic schist)  
The correlations were plotted only for data reported both porosity and De. Totally, 12 plots have been reported for Cl<sup>-</sup>. The De shows a tendency to increase with increasing rock porosity.



**Figure 2.1.1-4** Da values of  $\text{Cl}^-$  ion as a function of rock porosity for sedimentary rock (mudstone, tuff, pelitic schist)

The correlations were plotted only for data reported both porosity and Da. Totally 6 plots have been reported for  $\text{Cl}^-$ . No significant porosity dependency is found for Da, but it seems that the lower porosity is, the lower Da is.

## References

- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.169.
- Muurinen, A., Penttiä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.

- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Sato, H. (1991): PNC Technical Report "Report of Information Exchange Meeting on the Performance Assessment Research", PNC TN1410 91-079, pp.275~286 [In Japanese].
- Sato, H. and Yui, M. (1992): Measurement of Ionic Diffusion Coefficients in Rocks, 1992 Fall of the Atomic Energy Society of Japan, F15, p.323 [In Japanese].
- Sato, H., Yui, M. and Ishikawa, H. (1992): A Study on Pore Structure of Rocks, PNC Technical Report, PNC TN8410 92-222 [In Japanese].
- Sato, H. (1996): Through-Diffusion Experiments on NaCl through Tuff -Measurement of Tortuosity-, PNC Technical Report, PNC TN8410 96-111.
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion on Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119~133, Elsevier Science B.V.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.

## 2.1.2 Cesium (Cs)

The De values for Cs ( $\text{Cs}^+$ ) in sedimentary rock have been reported only for mudstone [PNC, 1996; Sato, 1997]. Since Cs is not redox sensitive, all measurements have been carried out under aerobic conditions. Cesium is one of the important elements for performance assessment of the geological disposal of HLW. In the performance assessment,  $^{135}\text{Cs}$  is especially regarded as an important nuclide ( $\beta^-$  decay property, half-life:  $2.0 \times 10^6$  y) because of its long half-life [PNC, 1992]. This is produced as one of the fission products. Much  $^{137}\text{Cs}$  is initially included in HLW, but this is rapidly decayed because of its short half-life ( $\beta^-$  decay property, half-life:  $30.17$  y)<sup>2</sup> [PNC, 1992]. Cesium is soluble in solution and it is presumed that the chemical behaviour doesn't also change in reducing conditions.

The De data have been based on the time-lag method [e.g. Muurinen et al., 1987] or based on De and rock capacity factor. The porewater initially saturated is all distilled water and no experiment carried out as a parameter for the ionic strength of porewater is found. All measurements have been performed at 25 °C and no measurement for temperature dependence on De has been also especially carried out. The reported porewater pH was 6 for all measurements.

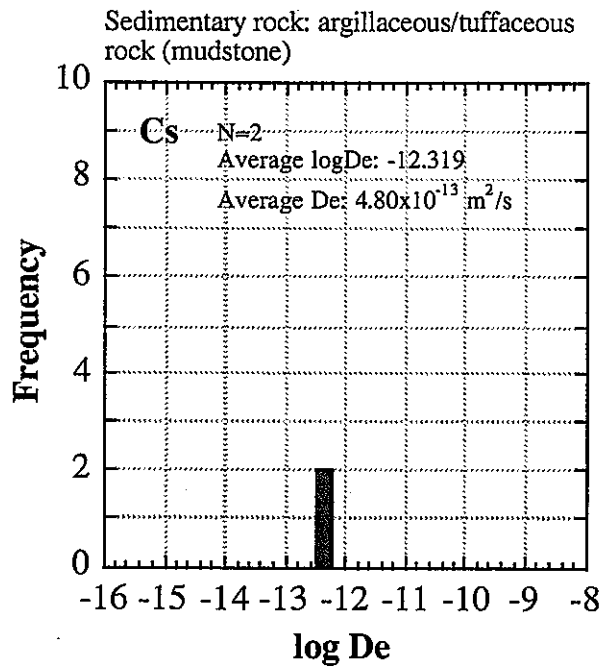
Figures 2.1.2-1 and 2.1.2-2 show histograms for De and Da values of  $\text{Cs}^+$  in sedimentary rock reported to date, respectively. The De values of  $\text{Cs}^+$  in sedimentary rock have been reported only for mudstone. Totally, 2 De data have been reported. Also for Da, totally, 2 data have been reported for mudstone.

Figures 2.1.2-3 and 2.1.2-4 show plots of De and Da values as a function of rock porosity for sedimentary rock, respectively. The correlations were plotted only for data reported both porosity and De or Da. Only one plot has been totally reported for both De and Da. Porosity dependency for diffusion can not be therefore discussed here.

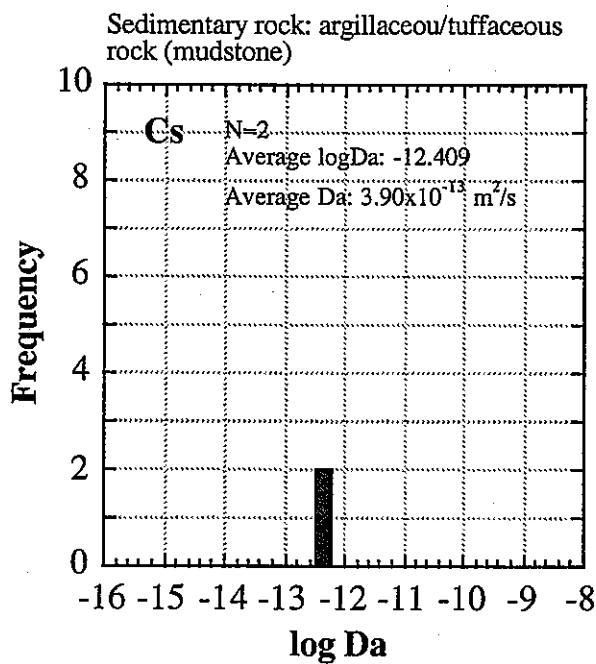
The Do of  $\text{Cs}^+$  is estimated to be  $2.1 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C by the Nernst-Einstein equation (it is also called the Nernst expression)[Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991; Marcus, 1997]. Formation factor (FF) and geometric factor (Gf) were calculated based on Do, De and porosity in the same way as Cl. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.1.10.

---

<sup>2</sup> Cesium-137 itself is a  $\beta^-$  decay nuclide (energy: 0.512 MeV), but the daughter,  $^{137\text{m}}\text{Ba}$  emits  $\gamma$ -ray by nuclear isomer transfer (IT), which has a strong energy of 0.662 MeV. Namely,  $^{137}\text{Cs}$  is generally called  $\gamma$  emitter.

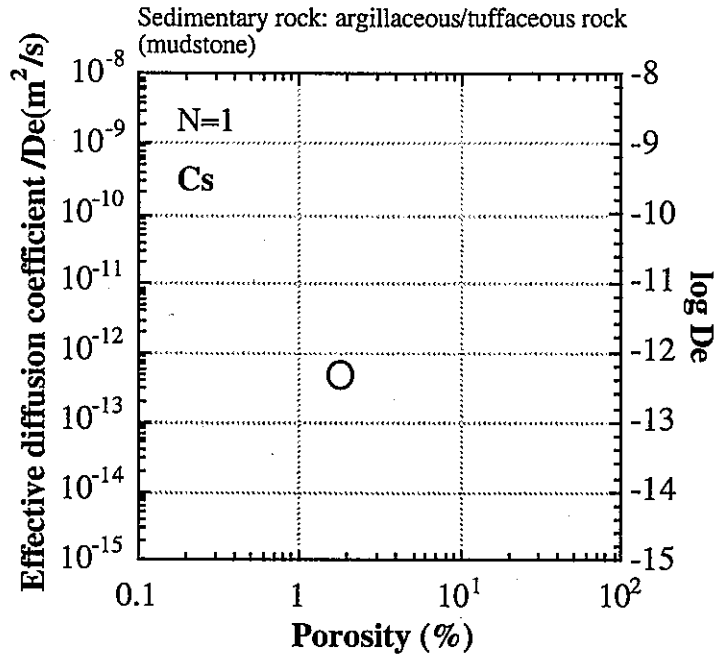


**Figure 2.1.2-1** Histogram for  $De$  values of  $\text{Cs}^+$  ion in sedimentary rock (mudstone)  
The average  $De$  of  $\text{Cs}^+$  in sedimentary rock was estimated to be  $4.80 \times 10^{-13} \text{ m}^2/\text{s}$ .



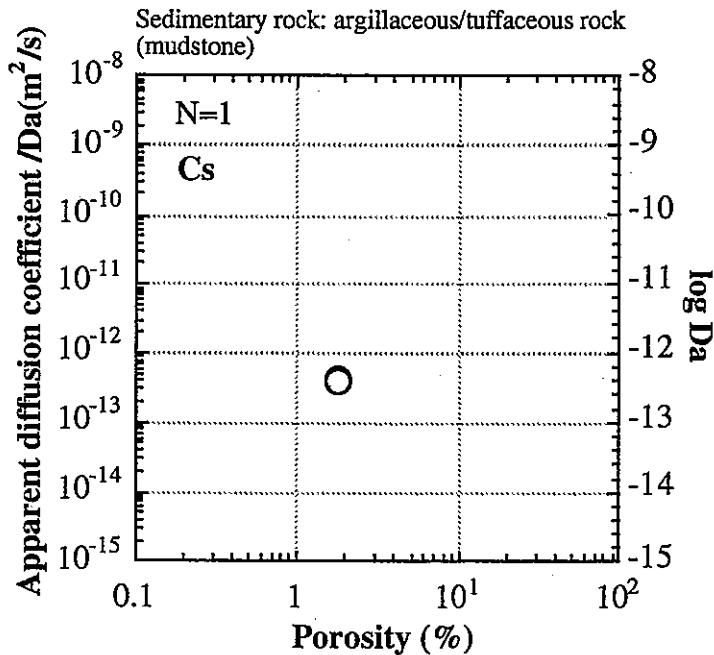
**Figure 2.1.2-2** Histogram for  $Da$  values of  $\text{Cs}^+$  ion in sedimentary rock (mudstone)  
The average  $Da$  of  $\text{Cs}^+$  in sedimentary rock was estimated to be  $3.90 \times 10^{-13} \text{ m}^2/\text{s}$ .





**Figure 2.1.2-3**  $D_e$  values of  $Cs^+$  ion as a function of rock porosity for sedimentary rock (mudstone)

Totally, only one plot has been reported for  $Cs^+$ . Porosity dependency for  $D_e$  can not be therefore discussed.



**Figure 2.1.2-4**  $D_a$  values of  $Cs^+$  ion as a function of rock porosity for sedimentary rock (mudstone)

Totally, only one plot has been reported for  $Cs^+$ . Porosity dependency for  $D_a$  can not be therefore discussed.

## References

- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1992): Research and Development on Geological Disposal of High-Level Radioactive Waste -First Progress Report-, PNC Technical Report, PNC TN1410 93-059.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119~133, Elsevier Science B. V.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.

### 2.1.3 Iodine (I)

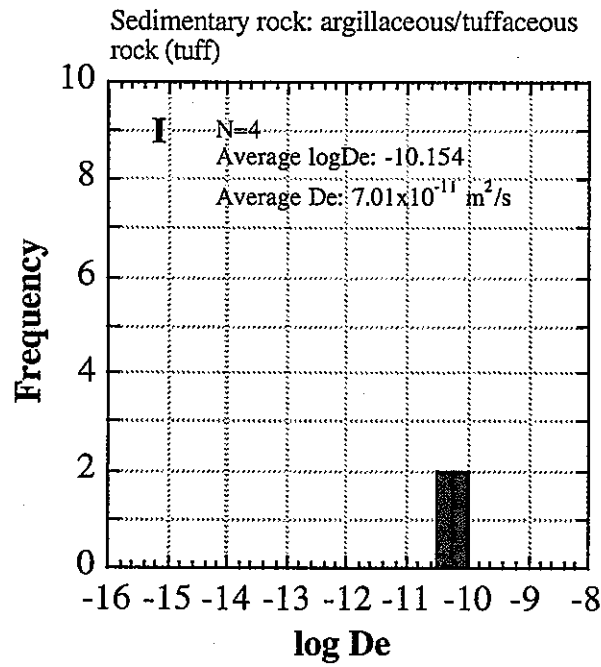
The  $D_e$  values for I ( $I^-$ ) in sedimentary rock have been reported for tuff [Nishiyama et al., 1990; Kita et al., 1989]. Since I (iodine) is not redox sensitive, all measurements have been carried out under aerobic conditions. Iodine is not directly included in important elements for performance assessment of the geological disposal of HLW, but it ( $^{129}I$ ) is accounted as one of the important elements in LLW. It is generally regarded that I (iodine) is a non-sorbing element, because it forms anionic species such as  $I^-$  and  $IO_3^-$  in solution. Though the dominant species of I depends on redox and pH conditions,  $I^-$  is predominant under aerobic conditions [Brookins, 1988]. The chemical behaviour of I doesn't also change in reducing conditions.

The  $D_e$  data have been obtained by through-diffusion method and  $D_a$  data have been derived based on the time-lag method [Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor. The porewater initially saturated is distilled water or deionized water and no experiment carried out as a parameter for the ionic strength of the porewater is found. All measurements have been performed at room temperature and temperature is not particularly reported. No measurement for temperature dependence on  $D_e$  has been carried out. The pH of the porewater is not reported.

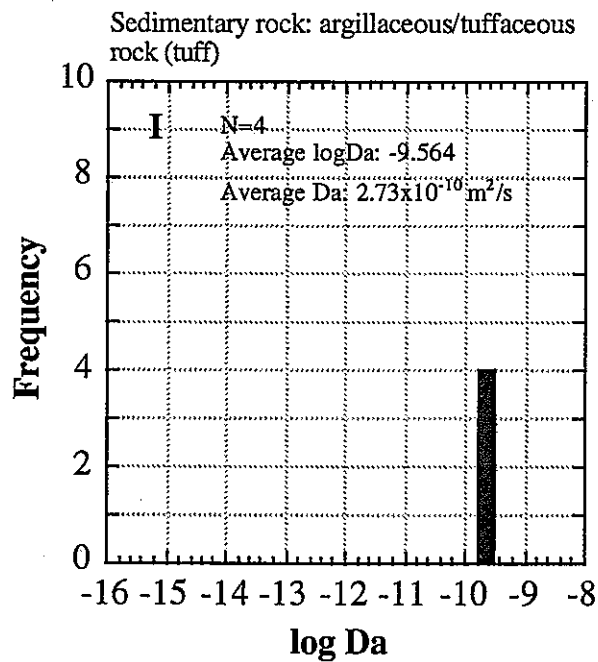
**Figures 2.1.3-1 and 2.1.3-2** show histograms for  $D_e$  and  $D_a$  values of  $I^-$  in sedimentary rock reported up to the present, respectively. The  $D_e$  values of  $I^-$  in sedimentary rock have been reported only for tuff. Four  $D_e$  data have been totally reported for tuff. For  $D_a$ , 4 data have been reported in total for tuff.

**Figures 2.1.3-3 and 2.1.3-4** show plots of  $D_e$  and  $D_a$  values of  $I^-$  as a function of rock porosity for sedimentary rock, respectively. The correlations were plotted only for data reported both porosity and  $D_e$  or  $D_a$ . Four plots have been totally reported for both  $D_e$  and  $D_a$ . Since rock porosity is relatively high (reported porosity: about 25 %), both  $D_e$  and  $D_a$  values are also high. Data enough to be able to discuss porosity dependency for diffusion are not reported.

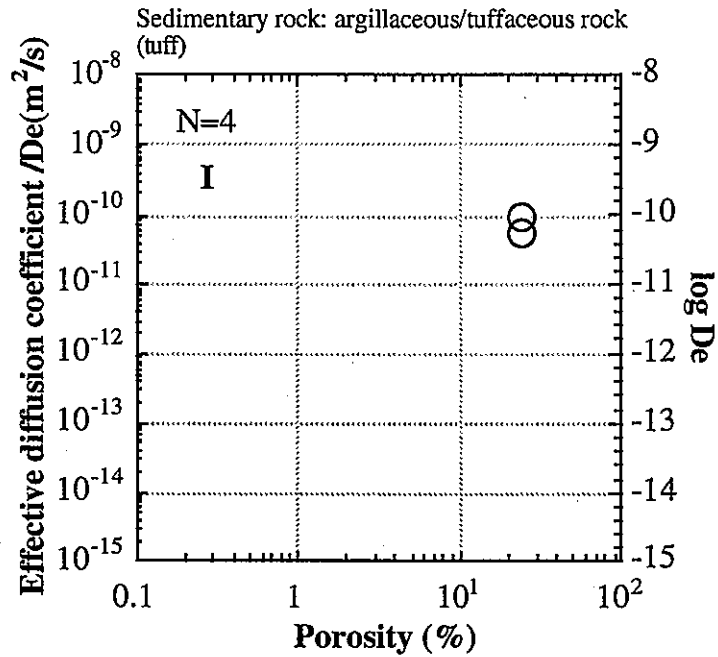
The  $D_0$  of  $I^-$  is estimated to be  $2.0 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C by the Nernst-Einstein equation (it is also called the Nernst expression)[Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991; Marcus, 1997]. Formation factor (FF) and geometric factor (Gf) were also calculated based on  $D_0$ ,  $D_e$  and porosity in the same way as Cl and Cs. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.1.10.



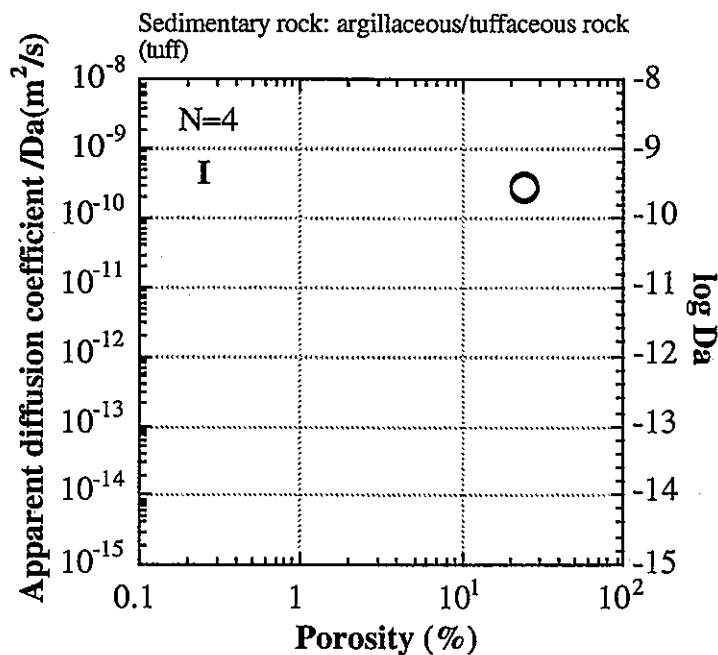
**Figure 2.1.3-1** Histogram for  $D_e$  values of  $I^-$  ion in sedimentary rock (tuff)  
The average  $D_e$  of  $I^-$  in sedimentary rock was estimated to be  $7.01 \times 10^{-11} \text{ m}^2/\text{s}$ .



**Figure 2.1.3-2** Histogram for  $D_a$  values of  $I^-$  ion in sedimentary rock (tuff)  
The average  $D_a$  of  $I^-$  in sedimentary rock was estimated to be  $2.73 \times 10^{-10} \text{ m}^2/\text{s}$ .



**Figure 2.1.3-3**  $D_e$  values of  $I^-$  ion as a function of rock porosity for sedimentary rock (tuff) The correlations were plotted only for data reported both porosity and  $D_e$ . Totally, 4 plots have been reported for  $I^-$ . Since data enough to be able to evaluate porosity dependency for  $D_e$  are not reported, it can not be discussed.



**Figure 2.1.3-4**  $D_a$  values of  $I^-$  ion as a function of rock porosity for sedimentary rock (tuff) The correlations were plotted only for data reported both porosity and  $D_a$ . Totally, 4 plots have been reported for  $I^-$ . Data enough to be able to discuss porosity dependency for  $D_a$  are not reported.

## References

- Brookins, D. G. (1988): Eh-pH Diagrams for Geochemistry, Springer-Verlag, Berlin, Germany, pp.14~15.
- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Kita, H., Iwai, T. and Nakashima, S. (1989): Diffusion Coefficient Measurement of an Ion in Pore Water of Granite and Tuff, Journal of the Japan Society of Engineering Geology, 30-2, pp.26~32 [In Japanese].
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.169.
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Nishiyama, K., Nakashima, S., Tada, R. and Uchida, T. (1990): Diffusion of an Ion in Rock Pore Water and Its Relation to Pore Characteristics, Mining Geology, 40 (5), pp.323~336 [In Japanese].
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.

## 2.1.4 Potassium (K)

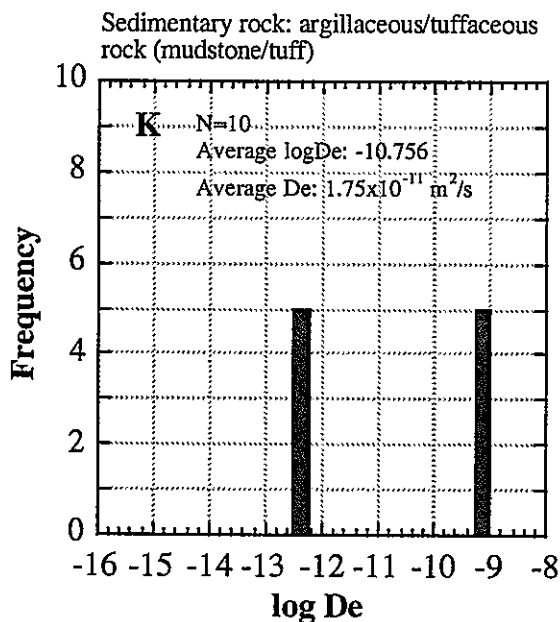
The  $D_e$  values for K ( $K^+$ ) in sedimentary rock have been reported for both mudstone and tuff [Conca et al., 1993; Conca and Wright, 1992] (In these papers,  $D_e$  is called a simple diffusion coefficient). Since K is not redox sensitive, all measurements have been carried out under aerobic conditions. Potassium is not especially regarded as an important element in performance assessment of the geological disposal of HLW, but it is presumed that K shows similar behaviour to that of Cs in solution. The chemical behaviour is not affected by redox condition, so it does not also change in reducing conditions.

All  $D_e$  data have been obtained by electrical conductivity method. Apparent diffusion coefficient can not be obtained by this method. Therefore, no  $D_a$  has been reported. Rock samples were initially saturated with a 0.1 M KCl solution, and then electrical conductivities were measured. The  $D_e$  values were determined by the Nernst-Einstein equation [Robinson and Stokes, 1959]. The principle of these measurements is as described in more detail in section 1.3.6. The porewater and tracer are therefore the same. All measurements have been performed at room temperature. The pH of the porewater is not reported.

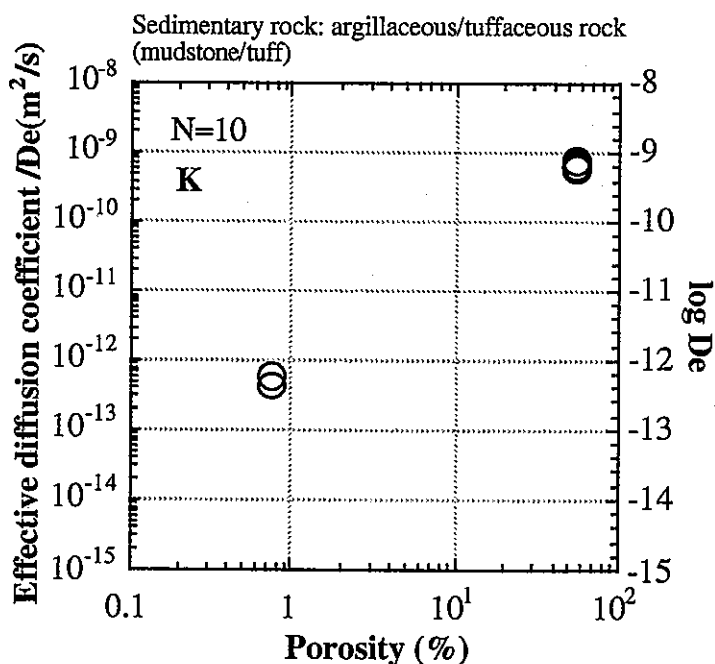
**Figure 2.1.4-1** shows a histogram for  $D_e$  values of  $K^+$  in sedimentary rock reported up to the present. Ten  $D_e$  data have been totally reported for mudstone and tuff. No  $D_a$  has been reported. The  $D_e$  was in the order of tuff and mudstone from highest to lowest. This is presumed to be due to the difference of porosity. The porosity of tuff is about 56 % and that of mudstone is 0.74 %.

**Figure 2.1.4-2** shows a plot of  $D_e$  values of  $K^+$  as a function of rock porosity for sedimentary rock. Totally, 10 plots have been reported together with porosity for  $D_e$ . The  $D_e$  clearly shows a tendency to increase with increasing rock porosity.

The  $D_0$  of  $K^+$  is estimated to be  $2.0 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C by the Nernst-Einstein equation (it is also called the Nernst expression) [Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991; Marcus, 1997]. Formation factor (FF) and geometric factor (Gf) were also calculated based on  $D_0$ ,  $D_e$  and porosity. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.1.10.



**Figure 2.1.4-1** Histogram for De values of K<sup>+</sup> ion in sedimentary rock (mudstone, tuff) The average De of K<sup>+</sup> in sedimentary rock was estimated to be  $1.75 \times 10^{-11} \text{ m}^2/\text{s}$ . In this histogram, the difference of porosity is not considered. Actually, the porosity of tuff (56 %) is quite larger than that of mudstone (0.74 %).



**Figure 2.1.4-2** De values of K<sup>+</sup> ion as a function of rock porosity for sedimentary rock (mudstone, tuff)

The correlations were plotted only for data reported both porosity and De. Totally, 10 plots have been reported for K<sup>+</sup>. The De clearly shows a tendency to increase with increasing rock porosity.



## References

- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Conca, J. L., Apted, M. J. and Arthur, R. C. (1993): Direct Determinations of Transport Parameters in Repository Materials, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVI (Interrante, C. G. and Pabalan, R. T. eds.), Vol.294, pp.839~844, Pittsburgh, Pennsylvania, U.S.A.
- Conca, J. L. and Wright, J. (1992): Direct Determinations of Unsaturated Flow and Transport, Proceedings of the Twelfth Annual American Geophysical Union Hydrology Days Meeting, Fort Collins, March 1992, pp.103~116.
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, *Geochimica et Cosmochimica Acta*, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, *Journal of Geotechnical Engineering*, Vol.117, No.3, American Society of Civil Engineering.

### 2.1.5 Sodium (Na)

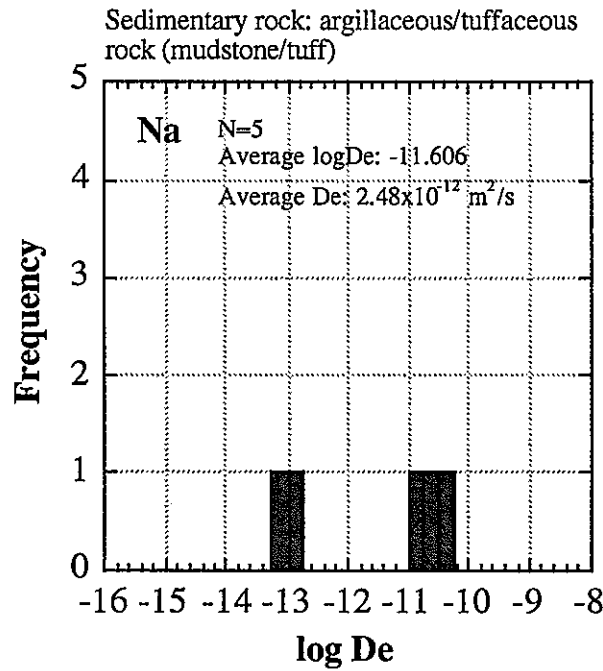
The  $D_e$  values for Na ( $\text{Na}^+$ ) in sedimentary rock have been reported for mudstone [Sato et al., 1997] and tuff [Sato, 1996]. Since Na is not redox sensitive, all measurements have been carried out under aerobic conditions. Sodium is not included in important elements for performance assessment of the geological disposal of HLW, but it is considered that Na shows similar chemical behaviour to those of Cs and K in solution. The chemical behaviour doesn't also change in reducing conditions.

The  $D_e$  data for Na have been obtained by through-diffusion method and  $D_a$  data have been derived based on the time-lag method [Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor. The porewater initially saturated is distilled water for all measurements. All measurements have been performed at 25 °C. The reported porewater pH was in a range of 5.5 to 9.54. Sodium chloride (NaCl) was used as a tracer solution for the measurements.

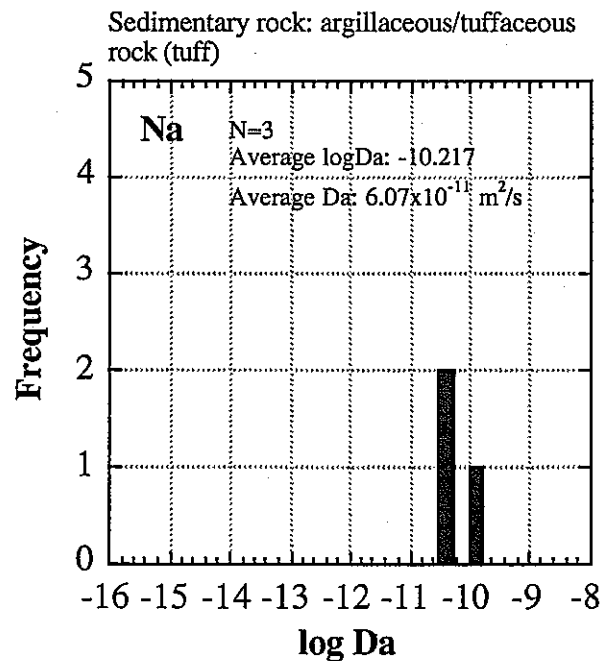
**Figures 2.1.5-1 and 2.1.5-2** show histograms for  $D_e$  and  $D_a$  values of  $\text{Na}^+$  in sedimentary rock reported up to the present, respectively. The  $D_e$  values of  $\text{Na}^+$  in sedimentary rock have been reported for mudstone and tuff, and  $D_a$  values have been reported only for tuff. Totally, 5  $D_e$  and 3  $D_a$  data have been reported for Na. The  $D_e$  was in the order of tuff and mudstone from highest to lowest. Relatively a wide range of variation is found in  $D_e$ . This is presumed to be due to the difference of porosity, because the porosity of tuff is much larger than that of mudstone. Those of tuff and mudstone are 1.8 % and 37.4 %, respectively, being quite different.

**Figures 2.1.5-3 and 2.1.5-4** show plots of  $D_e$  and  $D_a$  values of  $\text{Na}^+$  as a function of rock porosity for sedimentary rock, respectively. Totally, 5 and 3 plots have been reported for  $D_e$  and  $D_a$ , respectively. The  $D_e$  shows a tendency to increase with increasing rock porosity. Data enough to be able to evaluate porosity dependency are not reported for  $D_a$ .

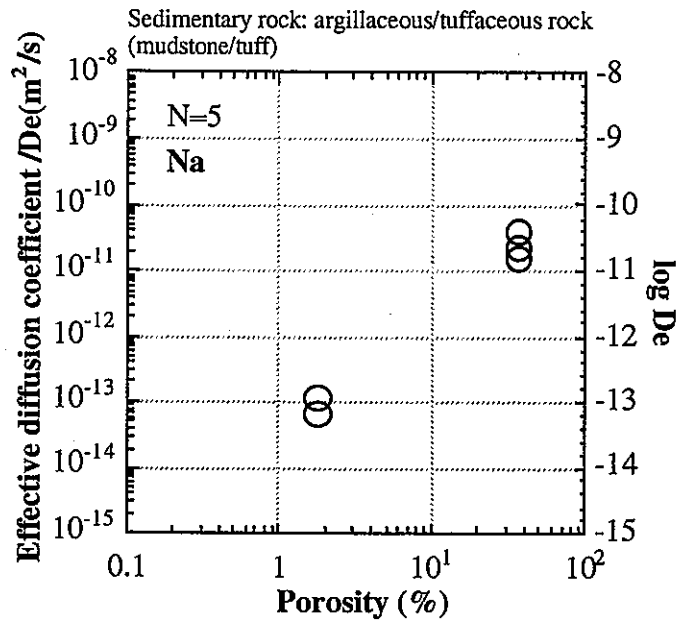
The  $D_0$  of  $\text{Na}^+$  is estimated to be  $1.3 \times 10^{-9} \text{ m}^2/\text{s}$  at 25 °C by the Nernst-Einstein equation (it is also called the Nernst expression)[Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991; Marcus, 1997]. Formation factor (FF) and geometric factor (Gf) were also calculated based on  $D_0$ ,  $D_e$  and porosity. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.1.10.



**Figure 2.1.5-1** Histogram for De values of Na<sup>+</sup> ion in sedimentary rock (mudstone, tuff) The average De of Na<sup>+</sup> in sedimentary rock was estimated to be  $2.48 \times 10^{-12} \text{ m}^2/\text{s}$ . In this histogram, the difference of porosity is not considered. However, the porosity is quite different between tuff and mudstone.

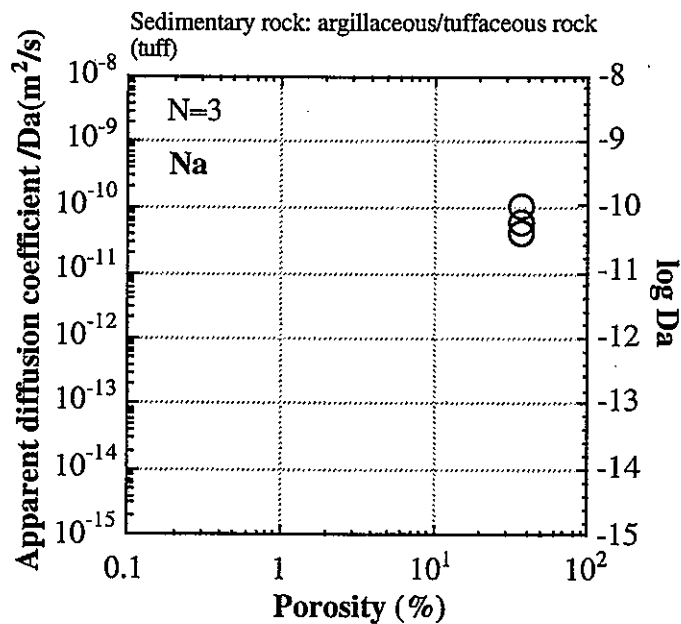


**Figure 2.1.5-2** Histogram for Da values of Na<sup>+</sup> ion in sedimentary rock (tuff) The average Da of Na<sup>+</sup> in sedimentary rock was estimated to be  $6.07 \times 10^{-11} \text{ m}^2/\text{s}$ .



**Figure 2.1.5-3**  $D_e$  values of  $Na^+$  ion as a function of rock porosity for sedimentary rock (mudstone, tuff)

The correlations were plotted only for data reported both porosity and  $D_e$ . Totally, 5 plots have been reported for  $Na^+$ . The  $D_e$  clearly shows a tendency to increase with increasing porosity.



**Figure 2.1.5-4**  $D_a$  values of  $Na^+$  ion as a function of rock porosity for sedimentary rock (tuff)

Totally, 3 plots have been reported for  $Na^+$ . Data enough to be able to discuss porosity dependency for  $D_a$  are not reported.

## References

- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Sato, H. (1996): Through-Diffusion Experiments on NaCl through Tuff -Measurement of Tortuosity-, PNC Technical Report, PNC TN8410 96-111.
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119~133, Elsevier Science B. V.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.

## 2.1.6 Nickel (Ni)

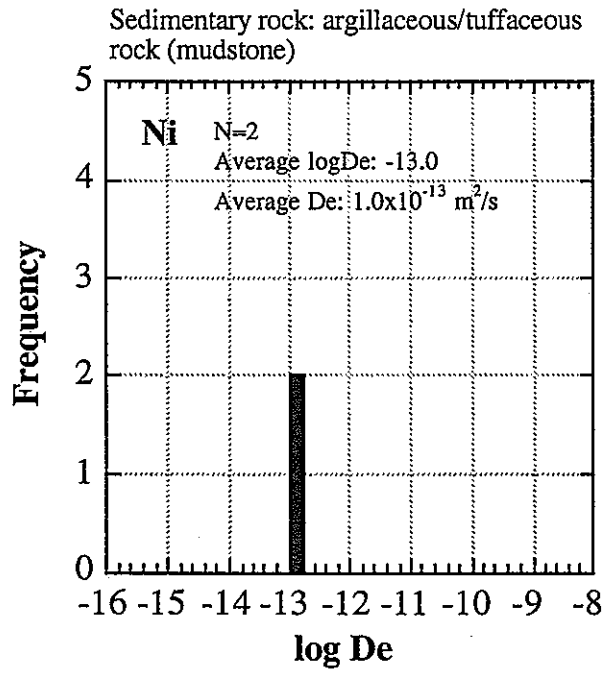
The  $D_e$  values for Ni ( $Ni^{2+}$ ) in sedimentary rock have been reported for mudstone [PNC, 1996; Sato et al., 1997]. Since Ni is not redox sensitive, all measurements have been carried out under aerobic conditions. Nickel is accounted as one of the important elements for performance assessment of the geological disposal of HLW. Nickel-59 has been especially regarded as an important nuclide ( $\beta^+$  decay property, half-life:  $7.5 \times 10^4$  y) because of its long half-life [PNC, 1991; Sato and Yui, 1997]. This is produced by activation of the corrosion products in a power reactor and is included in waste liquid in the process of reprocessing. Nickel is soluble in solution and it is presumed that the chemical behaviour does not also change in reducing conditions. Nickel chemically takes Ni(II), independent of redox condition, and  $Ni^{2+}$  is predominant in a pH range of  $< 9$ ,  $Ni(OH)_2$  predominant between 9 and 11, and  $HNiO_3^-$  predominant in a pH range of  $> 11$  in the system of Ni-O-H-S (activities:  $10^{-6}$  and  $10^{-3}$  for Ni and S, respectively)[Brookins, 1988].

The  $D_e$  data have been obtained by through-diffusion method and  $D_a$  values have been derived based on the time-lag method [e.g. Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor from breakthrough curves in the through-diffusion experiments. All measurements have been carried out in a distilled water system. All experiments have been performed at 25 °C. The reported porewater pH was 6 for all measurements. The species of Ni in the porewater in this condition is considered to be  $Ni^{2+}$  from the pH of the porewater and redox condition.

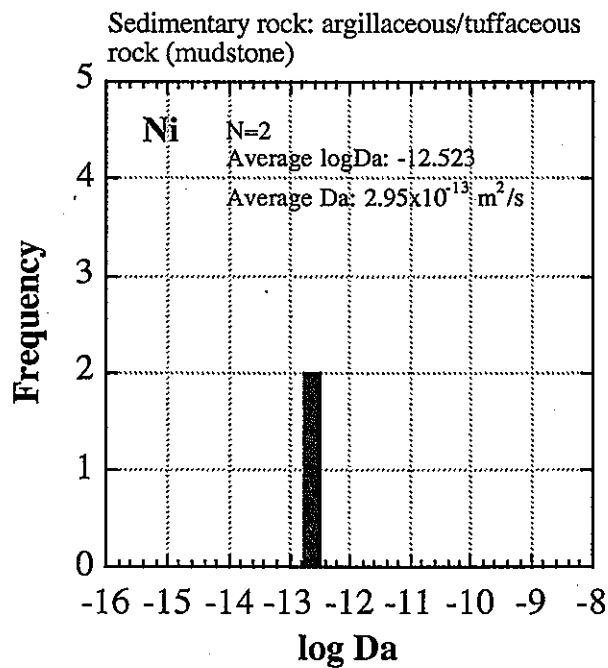
**Figures 2.1.6-1 and 2.1.6-2** show histograms for  $D_e$  and  $D_a$  values of  $Ni^{2+}$  in sedimentary rock reported up to the present, respectively. The  $D_e$  values of  $Ni^{2+}$  in sedimentary rock have been reported only for mudstone. Two  $D_e$  data have been reported in total. Also for  $D_a$ , 2 data have been totally reported for mudstone.

**Figures 2.1.6-3 and 2.1.6-4** show plots of  $D_e$  and  $D_a$  values of  $Ni^{2+}$  as a function of rock porosity for sedimentary rock, respectively. Since the correlations were plotted only for data reported both porosity and  $D_e$  or  $D_a$ , only one plot has been totally reported for both  $D_e$  and  $D_a$ . Porosity dependency for diffusion can not be therefore discussed here.

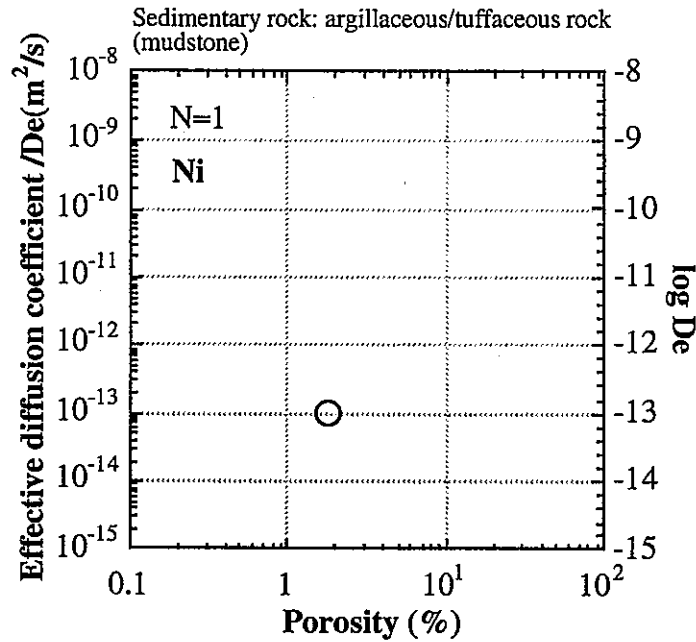
The  $D_0$  of  $Ni^{2+}$  is estimated to be  $6.6 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C by the Nernst-Einstein equation (it is also called the Nernst expression)[Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991; Marcus, 1997]. Formation factor (FF) and geometric factor (Gf) were also calculated based on  $D_0$ ,  $D_e$  and porosity. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.1.10.



**Figure 2.1.6-1** Histogram for De values of Ni<sup>2+</sup> ion in sedimentary rock (mudstone)  
The average De of Ni<sup>2+</sup> in sedimentary rock was estimated to be  $1.0 \times 10^{-13} \text{ m}^2/\text{s}$ .

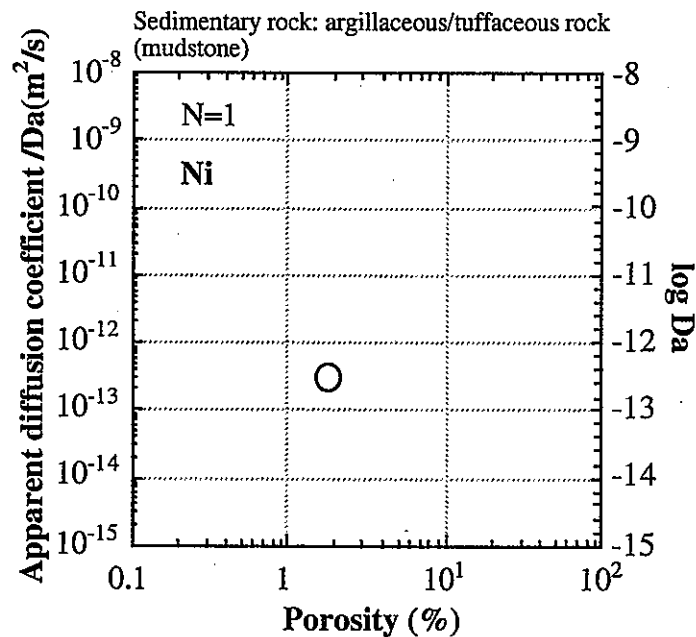


**Figure 2.1.6-2** Histogram for Da values of Ni<sup>2+</sup> ion in sedimentary rock (mudstone)  
The average Da of Ni<sup>2+</sup> in sedimentary rock was estimated to be  $2.95 \times 10^{-13} \text{ m}^2/\text{s}$ .



**Figure 2.1.6-3**  $D_e$  values of  $Ni^{2+}$  ion as a function of rock porosity for sedimentary rock (mudstone)

The correlation was plotted only for data reported both porosity and  $D_e$ . Totally, only one plot has been reported for  $Ni^{2+}$ . Therefore, porosity dependency for  $D_e$  cannot be evaluated.



**Figure 2.1.6-4**  $D_a$  values of  $Ni^{2+}$  ion as a function of rock porosity for sedimentary rock (mudstone)

The correlation was plotted for data reported both porosity and  $D_a$ . One plot has been reported for  $Ni^{2+}$ . Data enough to be able to discuss porosity dependency for  $D_a$  are not reported.



## References

- Brookins, D. G. (1988): Eh-pH Diagrams for Geochemistry, Springer-Verlag, Berlin, Germany, pp.68~70.
- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1991): Current Status for Research and Development of Geological Disposal -FY1989-, PNC Technical Report, PNC TN1410 91-009 [In Japanese].
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Sato, H. and Yui, M. (1997): Diffusion of Ni in Compacted Sodium Bentonite, Journal of Nuclear Science and Technology, Vol.34, No.3, pp.334~336.
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119~133, Elsevier Science B. V.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.

### 2.1.7 Selenium (Se)

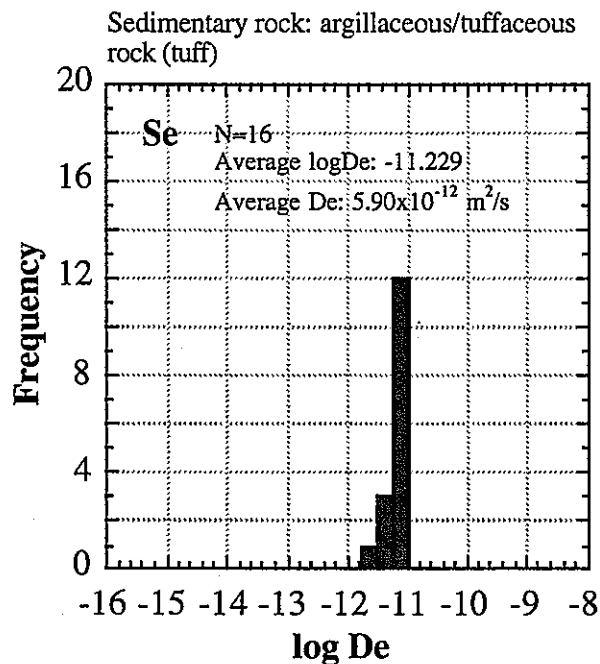
The De values for Se in sedimentary rock have been reported for tuff [Sato and Shibutani, 1996; PNC, 1996; Tachi et al., 1988]. Totally, 16 De data have been reported for Se. Also for Da, 16 data have been totally reported for the same kind of rock. Selenium is redox sensitive and it is well known that the valence changes depending on redox condition. Therefore, all experiments have been carried out in an N<sub>2</sub>-atmosphere glove-box, in which the concentration of oxygen was kept less than 1ppm. Selenium can chemically take Se(-II), (0), (II), (IV) and (VI), but since Se(II) is unstable, it doesn't normally exist. It is presumed that SeO<sub>3</sub><sup>2-</sup> species (selenite) is predominant in solution under anaerobic conditions [Ticknor et al. 1988; Brookins, 1988]. It is also well known that HSe<sup>-</sup> species (hydroselenide) is predominant in solution under reducing conditions [Ticknor et al. 1988; Brookins, 1988]. Moreover, Se exists as anionic species when it forms an ion in solution. It is therefore known that Se is generally weak sorptive onto bentonite and rocks, but Se relatively sorbs onto iron minerals such as pyrite, goethite, biotite and chlorite in the pH range less than their ZPC values (zero point of charge)[Shibutani et al., 1994a; 1994b]. Selenium is accounted as one of the important elements for performance assessment of the geological disposal of HLW. Selenium-79 has been especially regarded as an important nuclide ( $\beta^-$  decay property, half-life:  $6.5 \times 10^4$  y) because of its long half-life [PNC, 1992; Sato, 1997]. This is produced as one of the fission products in a power reactor and is included in HLW liquid.

All De data have been obtained by through-diffusion method and Da values have been derived based on the time-lag method [Muurinen et al., 1987] or based on De and rock capacity factor from breakthrough curves in the through-diffusion experiments. All measurements have been carried out in a 0.1 M NaCl system to make the ionic strength of porewater in both sides of cells the same. The experiments have been performed at 22 °C or temperature in glove-box. The reported porewater pH was in a range of 7.7 to 11.

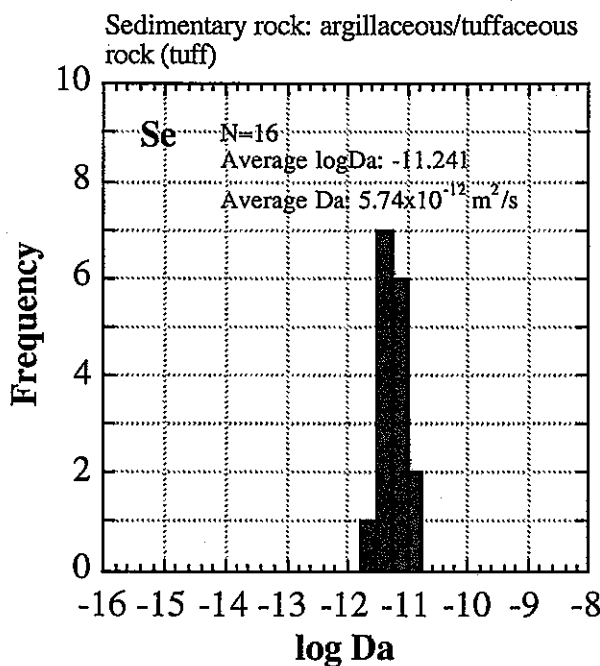
Figures 2.4.7-1 and 2.1.7-2 show histograms for De and Da values of SeO<sub>3</sub><sup>2-</sup> in sedimentary rock reported up to the present, respectively. The De values of SeO<sub>3</sub><sup>2-</sup> in sedimentary rock have been reported only for tuff. Also for Da, data for tuff have been reported. Sixteen data have been reported in total for both De and Da in tuff. The width of variation in De and Da is also small. This is presumed to be due to the width of variation in rock porosity was also small.

Figures 2.1.7-3 and 2.1.7-4 show plots of De and Da values of SeO<sub>3</sub><sup>2-</sup> as a function of rock porosity for sedimentary rock. Totally, 16 data have been plotted for both De and Da, but porosity dependency for diffusion could not be obtained, because the variation in porosity was too small as described above, the sorptivity of SeO<sub>3</sub><sup>2-</sup> onto rocks is generally small. Since the porosity of tuff was relatively high (about 37 %), it is presumed that both De and Da values were also relatively high.

The Do of SeO<sub>3</sub><sup>2-</sup> is estimated to be  $8.1 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C by the Nernst-Einstein equation (it is also called the Nernst expression)[Marcus, 1997]. Formation factor (FF) and geometric factor (Gf) were also calculated based on Do, De and porosity. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.1.10.



**Figure 2.1.7-1** Histogram for  $D_e$  values of  $\text{SeO}_3^{2-}$  ion in sedimentary rock (tuff). The average  $D_e$  of  $\text{SeO}_3^{2-}$  in sedimentary rock was estimated to be  $5.90 \times 10^{-12} \text{ m}^2/\text{s}$ . Almost all measured values are in good producibility within one order of magnitude in a deviation.



**Figure 2.1.7-2** Histogram for  $D_a$  values of  $\text{SeO}_3^{2-}$  ion in sedimentary rock (tuff). The average  $D_a$  of  $\text{SeO}_3^{2-}$  in sedimentary rock was estimated to be  $5.74 \times 10^{-12} \text{ m}^2/\text{s}$ . Also for  $D_a$ , relatively good producibility is found in measured values.

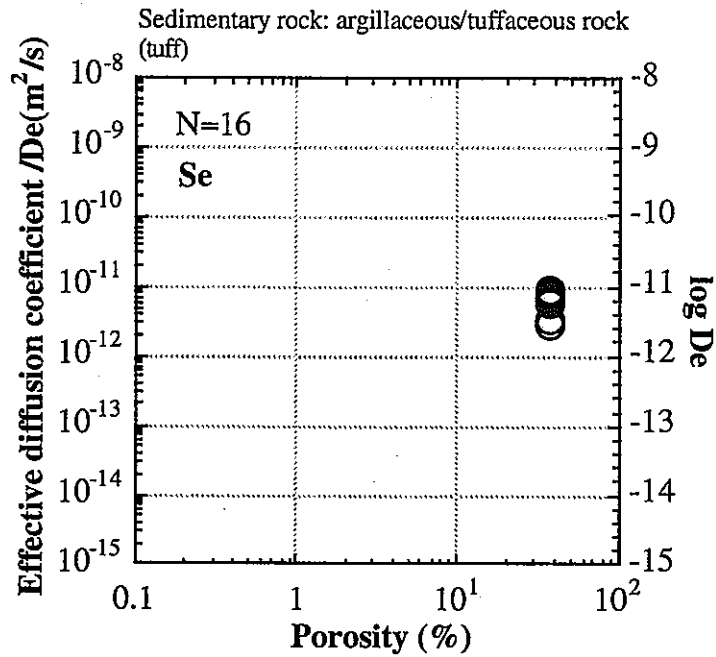


Figure 2.1.7-3  $D_e$  values of  $SeO_3^{2-}$  ion as a function of rock porosity for sedimentary rock (tuff)

Totally, 16  $D_e$  plots have been reported for  $SeO_3^{2-}$ . However, since the width of variation in porosity was too small, porosity dependency for  $D_e$  could not be evaluated.

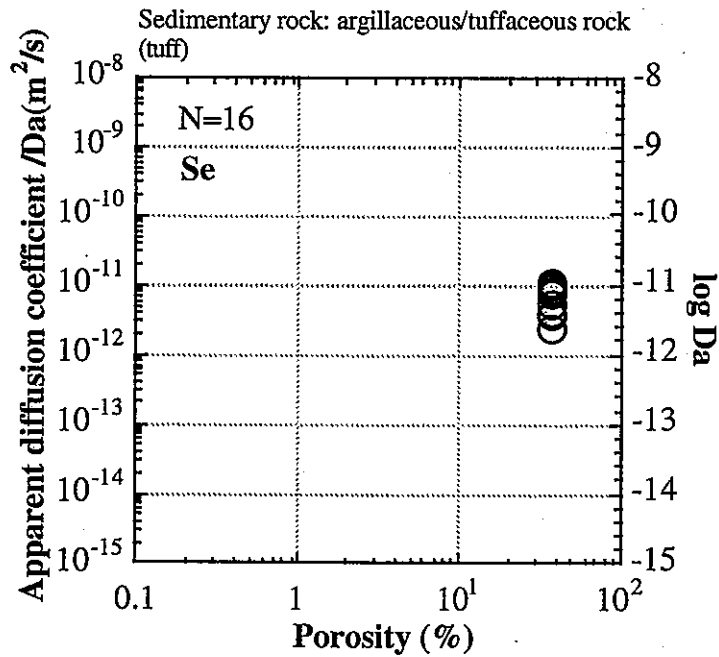


Figure 2.1.7-4  $D_a$  values of  $SeO_3^{2-}$  ion as a function of rock porosity for sedimentary rock (tuff)

Totally, 16  $D_a$  plots have been reported for  $SeO_3^{2-}$ . However, since the width of variation in porosity was also too small, porosity dependency for  $D_a$  could not be evaluated.

## References

- Brookins, D. G. (1988): Eh-pH Diagrams for Geochemistry, Springer-Verlag Berlin, Germany, pp.18~19.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.170.
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1992): Research and Development on Geological Disposal of High-Level Radioactive Waste -First Progress Report-, PNC Technical Report, PNC TN1410 93-059.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.208~210 [In Japanese].
- Sato, H. and Shibutani, T. (1996): Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste, PNC Technical Report, PNC TN1100 96-010, pp.150~155 [In Japanese].
- Sato, H. (1997): Diffusion Behaviour of Se in Compacted Sodium Bentonite under Reducing Conditions, PNC Technical Report, PNC TN8410 97-075.
- Shibutani, T., Nishikawa, Y., Inui, S., Uchidate, N., Yui, M. and Mano, T. (1994a): A Study on Sorption Behaviour for Se onto Rocks and Minerals, PNC Technical Report, PNC TN8410 94-395 [In Japanese].
- Shibutani, T., Yui, M. and Yoshikawa, H. (1994b): Sorption Mechanism of Pu, Am and Se on Sodium-Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVII (Barkatt, A. and Van Konynenburg, R. A. eds.), Vol.333, pp.725~730, Pittsburgh, Pennsylvania, U.S.A.
- Tachi, Y., Shibutani, T., Sato, H. and Yui, M. (1998): Sorption and Diffusion Behavior of Selenium in Tuff, Journal of Contaminant Hydrology, **35**, pp.77~89, Elsevier Science B. V.
- Ticknor, K. V. Harris, D. R. and Vandergraaf, T. T. (1988): Sorption/Desorption Studies of Selenium on Fracture-Filling Minerals under Aerobic and Anaerobic Conditions, Atomic Energy of Canada Limited Research Company Technical Report, TR-453.

## 2.1.8 Samarium (Sm)

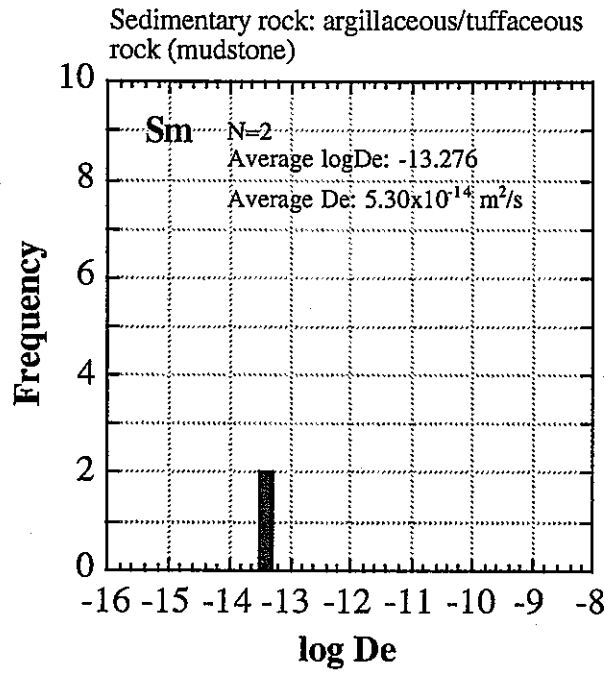
The De values for Sm in sedimentary rock have been reported for tuff [PNC, 1996; Sato et al., 1997]. Totally, 2 data have been reported for both De and Da. Samarium is accounted as one of the important elements for performance assessment of the geological disposal of HLW. Samarium-151 has been especially regarded as an important nuclide ( $\beta^-$  decay property, half-life: 93 y). Though Sm-151 relatively has a short half-life, it has a high activity in the initial condition after the closure of repository. This is produced as one of the fission products in a power reactor. Samarium is not redox sensitive. Therefore, all measurements have been carried out under aerobic conditions. However, the chemical behaviour of Sm is sensitive to the concentration of carbonate in solution. Since carbonate is supplied by the dissolution of calcite contained as one of the impurities or constituent minerals in bentonite or rock, it is considered that carbonate constantly exists in porewater. Samarium can chemically take Sm(II) and (III), but it takes Sm(III) in solution in a wide pH range [Brookins, 1988]. It has been clarified by Shibutani et al. [Shibutani et al., 1995a, 1995b; Shibutani, 1996] that the species of Sm changes as a function of pH and that three dominant species are predicted in a pH range lower than 6, around 8 and in a pH range higher than 10 under aerobic conditions. In this case, the dominant species are predicted to be  $\text{Sm}^{3+}$ ,  $\text{SmCO}_3^+$  and  $\text{Sm}(\text{CO}_3)_2^-$  in a pH range lower than 6, around 8 and in a pH range higher than 10, respectively. If no carbonate exists in solution, the dominant species of Sm in a high pH range is different from samarium carbonates, samarium hydroxides such as  $\text{SmOH}^{2+}$ ,  $\text{Sm}(\text{OH})_2^+$  and  $\text{Sm}(\text{OH})_3(\text{aq})$  become predominant [Shibutani et al. 1995c]. Although Sm is one of the lanthanides, it is generally considered that the chemical behaviour of Sm is similar to those of elements, which take trivalent elements such as Ac, Am and Cm, which are actinides [e.g. Sato, 1997; Sato, 1998a, 1998b]. Therefore, Sm was used as a representative of trivalent elements.

The De data have been obtained by through-diffusion method and Da values have been derived based on the time-lag method [Muurinen et al., 1987] or based on De and rock capacity factor. All measurements have been carried out in a distilled water system. In addition, all experiments have been performed at 25 °C. The reported porewater pH was 6 for all measurements. The species of Sm in the porewater in this condition is considered to be  $\text{Sm}^{3+}$ .

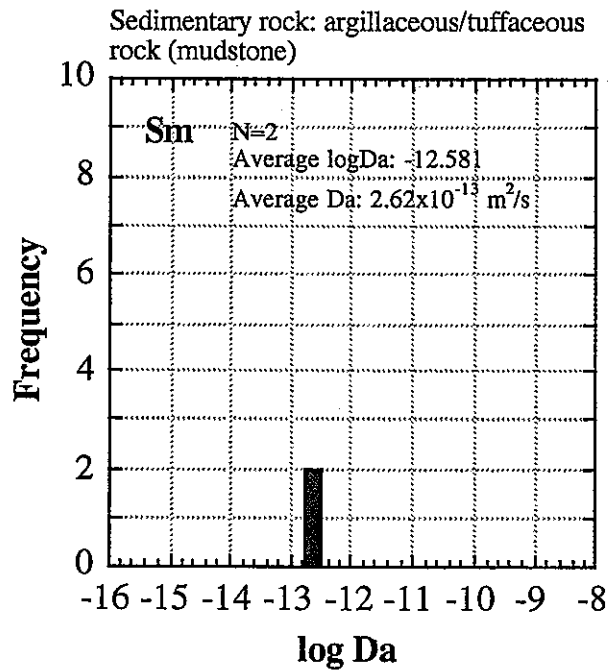
Figures 2.1.8-1 and 2.1.8-2 show histograms for De and Da values of  $\text{Sm}^{3+}$  in sedimentary rock reported up to the present, respectively. The De and Da values of  $\text{Sm}^{3+}$  have been reported only for mudstone. Two De data have been reported in total. Also for Da, 2 data have been reported.

Figures 2.1.8-3 and 2.1.8-4 show plots of De and Da values of  $\text{Sm}^{3+}$  as a function of rock porosity for sedimentary rock, respectively. Since the correlations were plotted only for data reported both porosity and De or Da, only one plot has been totally reported for both De and Da. Therefore, porosity dependency for diffusion cannot be discussed here.

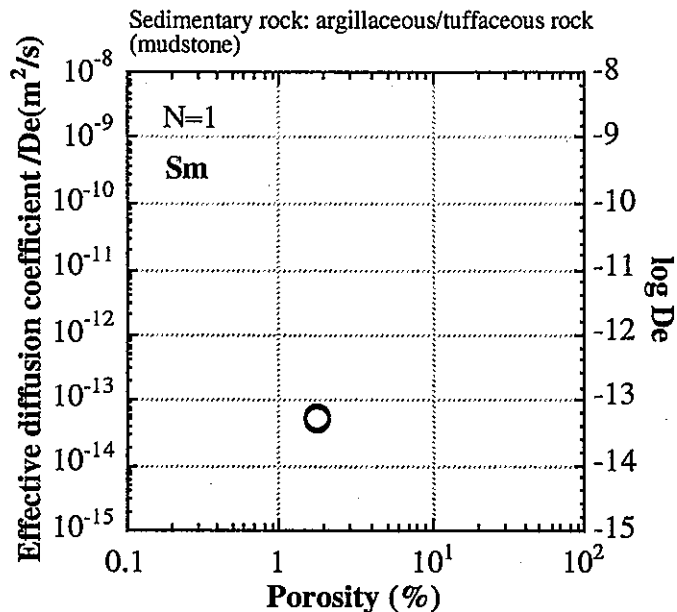
The Do of  $\text{Sm}^{3+}$  is estimated to be  $6.1 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C by the Nernst-Einstein equation (it is also called the Nernst expression)[Marcus, 1997]. Formation factor (FF) and geometric factor (Gf) were also calculated based on Do, De and porosity. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.1.10.



**Figure 2.1.8-1** Histogram for De values of  $\text{Sm}^{3+}$  ion in sedimentary rock (mudstone). The average De of  $\text{Sm}^{3+}$  in sedimentary rock was estimated to be  $5.30 \times 10^{-14} \text{ m}^2/\text{s}$ .

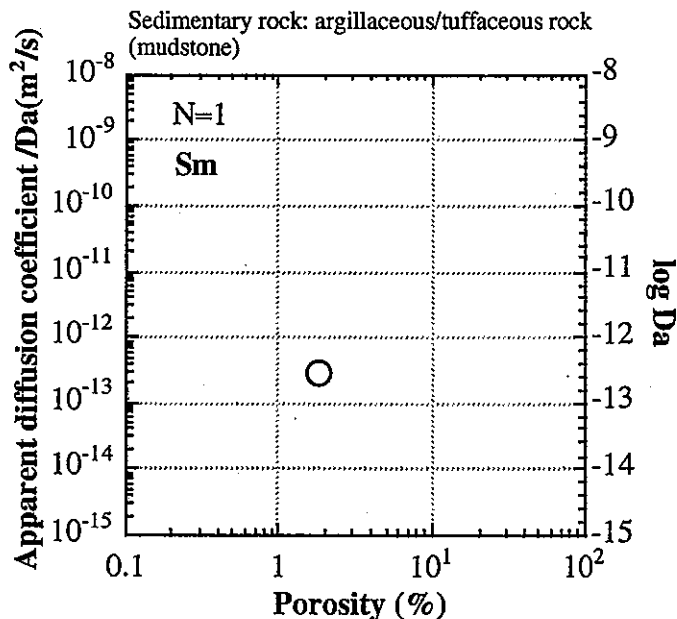


**Figure 2.1.8-2** Histogram for Da values of  $\text{Sm}^{3+}$  ion in sedimentary rock (mudstone). The average Da of  $\text{Sm}^{3+}$  in sedimentary rock was estimated to be  $2.62 \times 10^{-13} \text{ m}^2/\text{s}$ .



**Figure 2.1.8-3**  $D_e$  values of  $Sm^{3+}$  ion as a function of rock porosity for sedimentary rock (mudstone)

The correlation was plotted only for data reported both porosity and  $D_e$ . Totally, only one plot has been reported for  $Sm^{3+}$ . Therefore, porosity dependency for  $D_e$  cannot be evaluated here.



**Figure 2.1.8-4**  $D_a$  values of  $Sm^{3+}$  ion as a function of rock porosity for sedimentary rock (mudstone)

The correlation was plotted only for data reported both porosity and  $D_a$ . Totally, only one plot has been reported for  $Sm^{3+}$ . Therefore, porosity dependency for  $D_a$  cannot be evaluated here.



## References

- Brookins, D. G. (1988): Eh-pH Diagrams for Geochemistry, Springer-Verlag, Berlin, Germany, p.136.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.169.
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119~133, Elsevier Science B. V.
- Sato, H. (1997): Effect of Dry Density and pH on Diffusion for Sm in Compacted Sodium Bentonite, PNC Technical Report, PNC TN8410 97-007.
- Sato, H. (1998a): Diffusion Behaviour of Se(-II) and Sm(III) in Compacted Sodium Bentonite, JNC Technical Review, No.1, pp.51~62, Japan Nuclear Cycle Development Institute [In Japanese].
- Sato, H. (1998b): Diffusion Behaviour of Se(-II) and Sm(III) in Compacted Sodium Bentonite, Radiochimica Acta, 82, pp.173~178.
- Shibutani, S., Makino, H., Yoshikawa, H. and Yui, M. (1995a): Solubility and Formation Constants of  $\text{SmOHCO}_3$ , PNC Technical Report, PNC TN8410 95-031 [In Japanese].
- Shibutani, S., Makino, H., Yoshikawa, H. and Yui, M. (1995b): Solubility and Formation Constants of  $\text{SmOHCO}_3$ , 1995 Annual Meeting of the Atomic Energy Society of Japan, J9 [In Japanese].
- Shibutani, S., Shibutani, T., Yoshokawa, H. and Yui, M. (1995c): Solubility Measurements of Samarium Hydroxide under Low  $\text{CO}_2$  Concentration System, PNC Technical Report, PNC TN8410 95-204 [In Japanese].
- Shibutani, S. (1996): Solubility Measurement of Trivalent Lanthanide for Performance Assessment, PNC Technical Review, No.97, PNC TN8410 96-011, pp.67~75 [In Japanese].

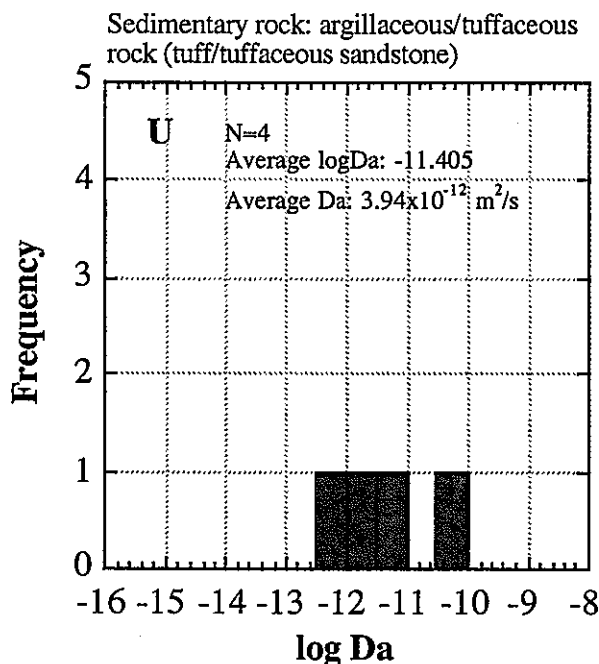
## 2.1.9 Uranium (U)

The Da values for U(VI) in sedimentary rock have been reported for tuff [Kosaka et al., 1984] and tuffaceous sandstone [Ota and Yoshida, 1994], but no De has been reported. Totally, 4 Da data have been reported for U. Uranium is accounted as one of the important elements for performance assessment of the geological disposal of HLW. Uranium-233, 234, 235, 236 and 238 have been particularly regarded as important nuclides ( $^{233}\text{U}$ :  $\alpha$  decay property, half-life  $1.592 \times 10^5$  y;  $^{234}\text{U}$ :  $\alpha$  decay property, half-life  $2.446 \times 10^5$  y;  $^{235}\text{U}$ :  $\alpha$  decay property, half-life  $7.038 \times 10^8$  y;  $^{236}\text{U}$ :  $\alpha$  decay property, half-life  $2.342 \times 10^7$  y;  $^{238}\text{U}$ :  $\alpha$  decay property, half-life  $4.468 \times 10^9$  y) because of their long half-lives [PNC, 1991; Sato, 1997, 1998]. These nuclides are produced as decay chain nuclides. Uranium is redox sensitive and it is known that the valence changes depending on redox condition. Uranium can chemically take U(III), (IV), (V) and (VI) in solution, but U(VI) and U(IV) are predominant in solution under aerobic and reducing conditions, respectively. The chemical species also depends on the porewater pH and the concentration of carbonate dissolving in solution.

All Da data have been obtained by in-diffusion method (this method is also called back-to-back method. See sections 1.3.1 and 1.3.3 for the difference between in-diffusion and back-to-back method.). The measurements for tuff and tuffaceous sandstone have been carried out in a deionized water and a groundwater system, respectively. The experiments for tuff and tuffaceous sandstone have been performed in a range of 20 to 50 °C and at room temperature, respectively. However, no activation energy for diffusion is reported for tuff. The porewater pH is not reported for all measurements. All measurements have been carried out under aerobic conditions. In this case, the valence of U is considered to be hexavalent. The dominant chemical species of U in the porewater in this case is considered to be  $\text{UO}_2(\text{CO}_3)_3^{4-}$  from the chemical and redox conditions, but the speciation of U is not reported.

Figure 2.1.9-1 shows a histogram for Da values of  $\text{UO}_2(\text{CO}_3)_3^{4-}$  in sedimentary rock reported up to the present. Since no datum for rock porosity is reported, porosity dependency for Da cannot be discussed. The width of variation in Da is over two orders of magnitude even though almost all measurements have been carried out in similar chemical conditions. It is considered that temperature conditions and pore structural factors such as porosity affected diffusion behaviour.

The Do of  $\text{UO}_2(\text{CO}_3)_3^{4-}$  has not been directly measured and no datum is existing, but it has been estimated to be  $7.2 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C based on the Stokes equation [Robinson and Stokes, 1959] by the ionic radius calculated from the molecular structure [Yamaguchi, 1996]. Since neither De nor porosity is reported, porosity dependencies for formation factor (FF) and geometric factor (Gf) cannot be estimated.



**Figure 2.1.9-1** Histogram for Da values of  $\text{UO}_2(\text{CO}_3)_3^{4-}$  ion in sedimentary rock (tuff, tuffaceous sandstone)

The average Da of  $\text{UO}_2(\text{CO}_3)_3^{4-}$  in sedimentary rock was estimated to be  $3.94 \times 10^{-12} \text{ m}^2/\text{s}$ . Since no datum was reported for both porosity and Da, porosity dependency for Da cannot be discussed here.

## References

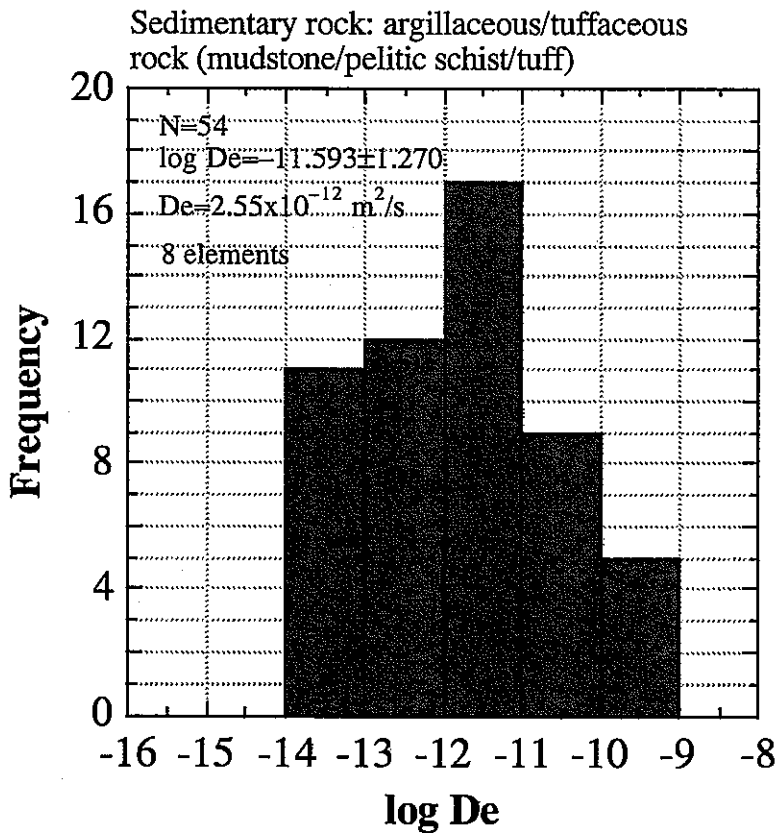
- Kosaka, T., Sato, S., Furuya, H., Araya, S. and Matsuo, K. (1984): Diffusion Behavior of Uranium in a Rock Block, 1984 Fall Meeting of the Atomic Energy Society of Japan, G82 [In Japanese].
- Ota, K. and Yoshida, H. (1994): Influence Microscopic Heterogeneity on Diffusion for Sedimentary Rocks, Proceedings of an NEA/SKI Symposium, Paris, France, 11-14, October 1994, pp.237~243.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1991): Current Status for Research and Development of Geological Disposal -FY1989-, PNC Technical Report, PNC TN1410 91-009 [In Japanese].
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Sato, H. (1997): A Study on Nuclide Migration in Buffer Materials and Rocks for Geological Disposal of Radioactive Waste, Doctoral Thesis of Akita University [In Japanese].

Sato, H. (1998): A Study on Nuclide Migration in Buffer Materials and Rocks for Geological Disposal of Radioactive Waste, PNC Technical Report, PNC TN8410 97-202 [In Japanese].

Yamaguchi, T. (1996): Diffusivity of Actinides in a Granite from Inada, Ibaraki, Japan, PNC Technical Report, PNC TN1100 96-010, pp.156~160 [In Japanese].

### 2.1.10 Summary for diffusivity data in argillaceous and tuffaceous rock

The  $D_e$  and  $D_a$  values for totally 9 elements were discussed in sections 2.1.1~2.1.9. Figure 2.1.10-1 shows a histogram of  $D_e$  values in argillaceous and tuffaceous rock of sedimentary rock for 8 elements<sup>3</sup> reported in each section. Totally, 9 elements were discussed for sedimentary rock, but no  $D_e$  only for U has been reported. Though the average  $D_e$  of all elements in sedimentary rock was estimated to be  $2.55 \times 10^{-12} \text{ m}^2/\text{s}$ , relatively a wide range of variation is found in  $D_e$ . The width of variation in  $D_e$  is approximately 5 orders of magnitude. This attributes to the variation in rock porosity. In these rocks, the porosity of tuff was between 25 and 56 %, and that of mudstone was between 0.74 and 1.8 %. That of pelitic schist was 1.0%. As is clear from this, rock porosity is quite different depending on the kind of rock. Therefore,  $D_e$  should be discussed together with rock porosity.



**Figure 2.1.10-1** Histogram for  $D_e$  values in sedimentary rock for 8 elements (mudstone, pelitic schist, tuff)

The average  $D_e$  of all elements in sedimentary rock was estimated to be  $2.55 \times 10^{-12} \text{ m}^2/\text{s}$ . Since the variation in rock porosity is not considered in the histogram, relatively a wide range of variation is found in  $D_e$ .

<sup>3</sup> 8 elements: Cl, Cs, I, K, Na, Ni, Se and Sm.

Figure 2.1.10-2 shows a histogram of Da values in sedimentary rock for 8 elements (no Da has been reported only for K) discussed in sections 2.1.1~2.1.9. The average Da of all elements in sedimentary rock was estimated to be  $7.28 \times 10^{-12} \text{ m}^2/\text{s}$  and a variation of approximately three orders of magnitude is found in Da. The Da includes sorption onto minerals composing rock, so the difference in Da can not be simply explained only by porosity and pore structure. The Da is affected by both sorption and De. Since De depends on the pore structure of rock, and sorption also depends on the kind of rock and chemical conditions, it is presumed that the variation in Da was mainly caused by the differences of constituent minerals and pore structure by the difference of the kind of rock.

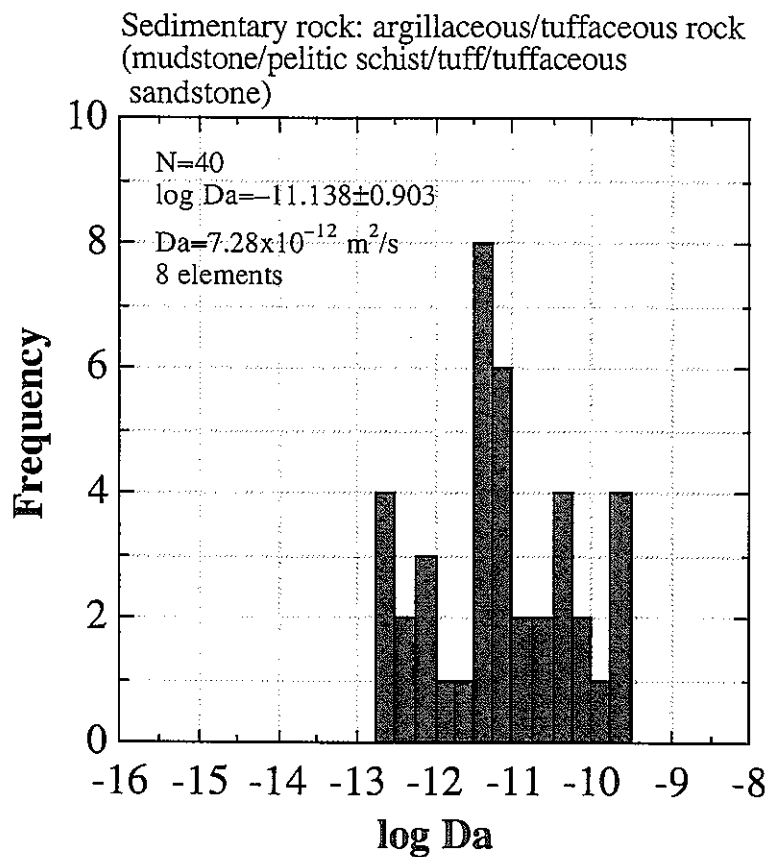


Figure 2.1.10-2 Histogram for Da values in sedimentary rock for 8 elements (mudstone, pelitic schist, tuff, tuffaceous sandstone)

The average value of Da in sedimentary rock (argillaceous/tuffaceous rock) was estimated to be  $7.28 \times 10^{-12} \text{ m}^2/\text{s}$ . The width of variation in Da is approximately three orders of magnitude. This is mainly considered to attribute to the differences and sorptivity onto rock.

Figure 2.1.10-3 shows a porosity dependency of De values for 8 elements discussed in sections 2.1.1-2.1.9 (no De has been reported only for U). Totally, 50 plots have been reported for De together with porosity. The De clearly shows a tendency to increase with increasing rock porosity, although some variation is found.

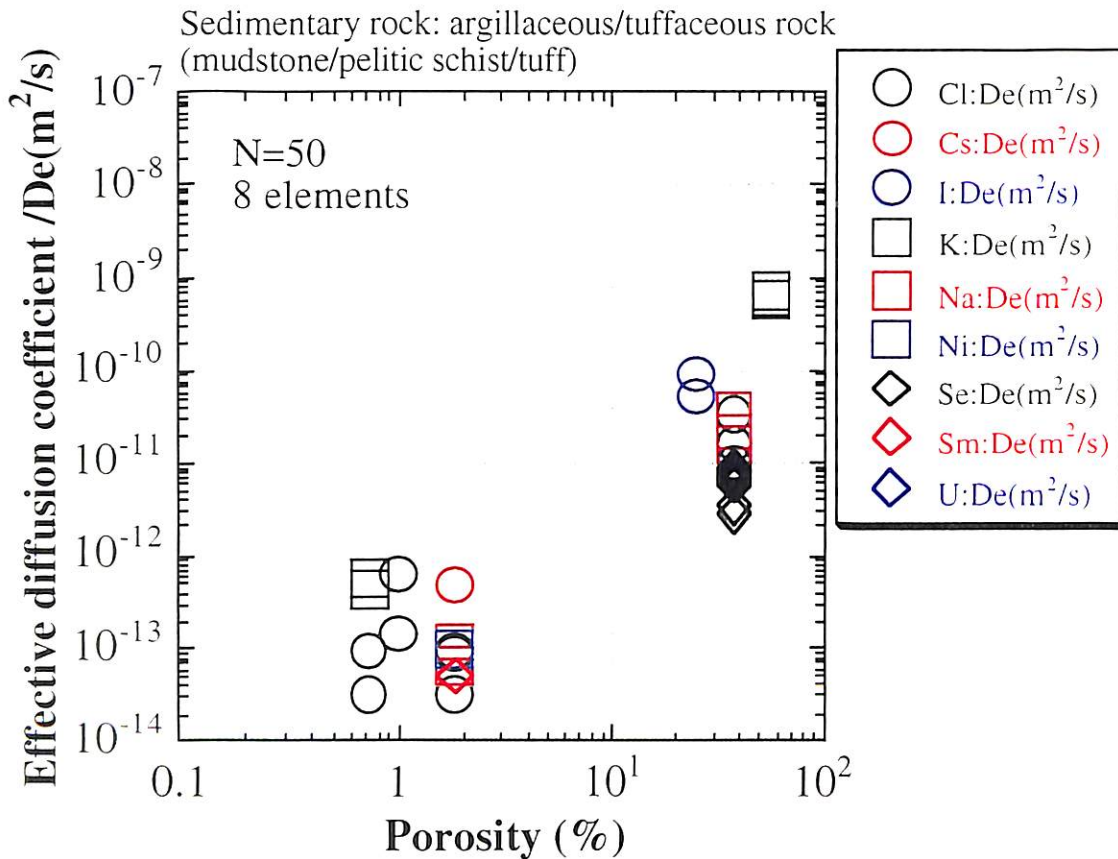


Figure 2.1.10-3 De values for 8 elements as a function of porosity for sedimentary rock (mudstone, pelitic schist, tuff)

No De only for U in sedimentary rock (argillaceous and tufaceous rock) has been reported. Though some variation is found, De clearly shows a tendency to increase with increasing rock porosity.

Figure 2.1.10-4 shows a least squares fitting curve for  $D_e$  values as a function of rock porosity. In the derivation of empirical equation,  $D_e$  data for all elements (8 elements except U) were taken into account without distinguishing the kind of element. Consequently, the empirical equation:  $D_e = (10^{-12.87 \pm 0.159}) \cdot \phi^{1.39 \pm 0.127}$  was derived, where  $\phi$  is the porosity in % and the errors stand for the standard deviations.

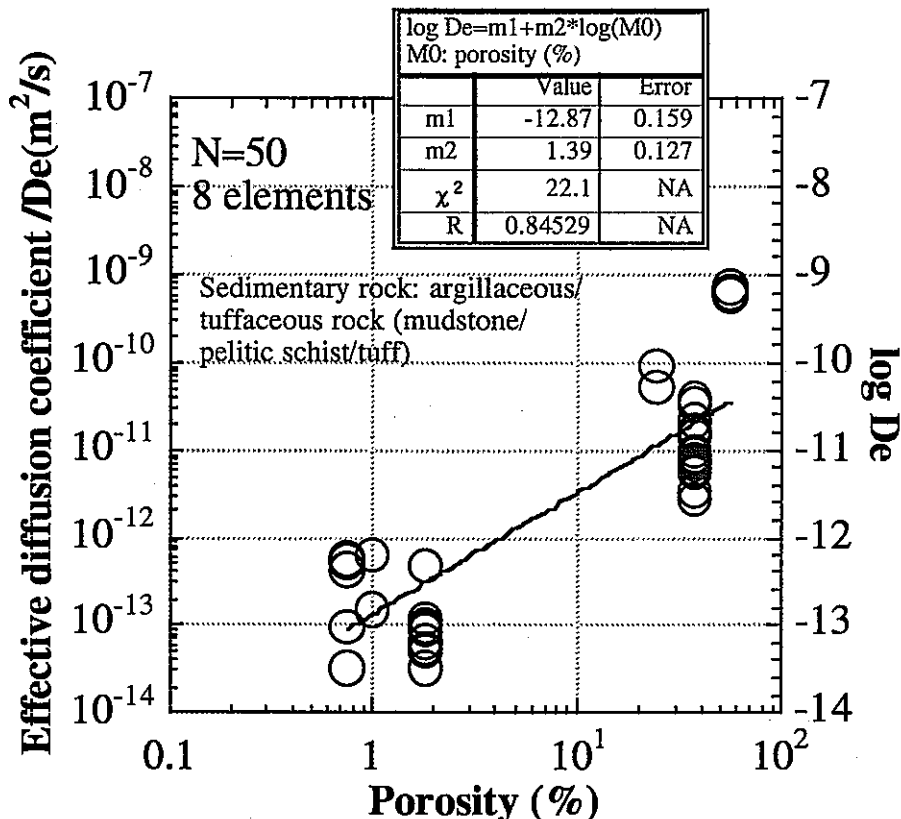


Figure 2.1.10-4 Empirical equation for  $D_e$  values as a function of rock porosity for sedimentary rock (mudstone, pelitic schist, tuff)



Figure 2.1.10-5 shows a width of variation in De values as a function of rock porosity. Two lines were drawn to be parallel to the empirical equation of De values as a function of rock porosity, which is shown in Figure 2.1.10-4 and to pass on the maximum and minimum values of plotted De values. This width of variation does not have statistical meaning, merely showing measured maximum and minimum values.

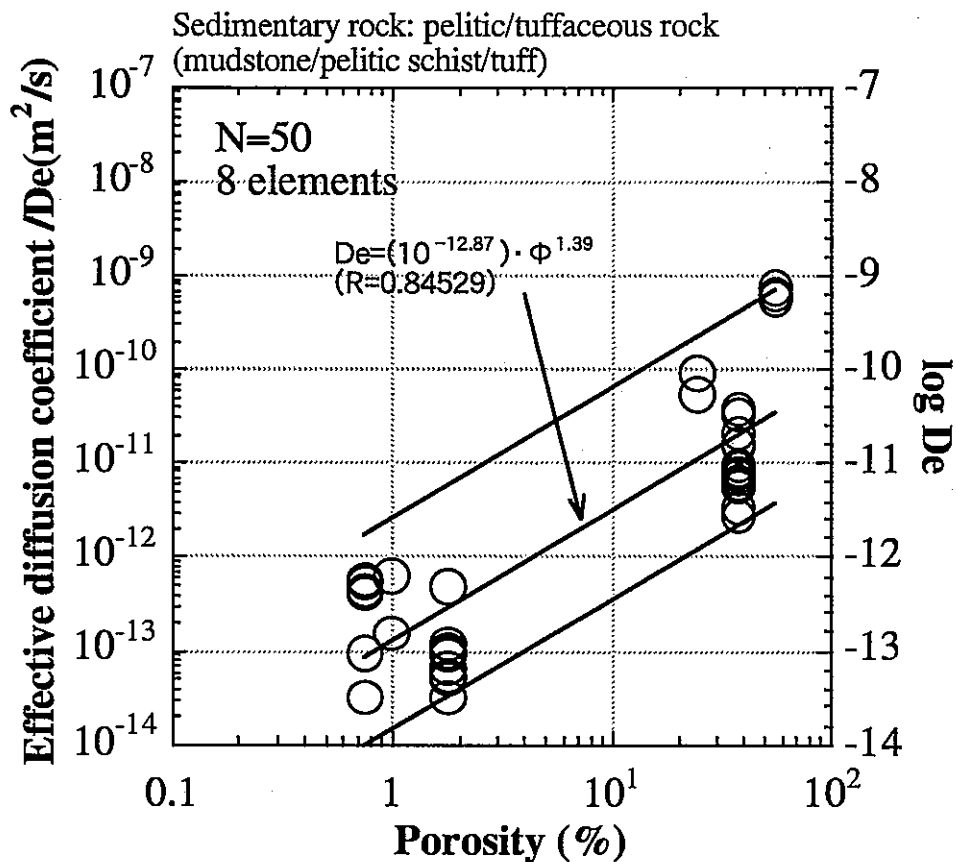


Figure 2.1.10-5 A width of variation in De values as a function of porosity for sedimentary rock (mudstone, pelitic schist, tuff)

Figure 2.1.10-6 shows a least squares fitting curve for Da values as a function of rock porosity. Though Da shows a slight tendency to increase with increasing rock porosity, clear trend is not found. A wide range of variation is found in Da, even similar porosity.

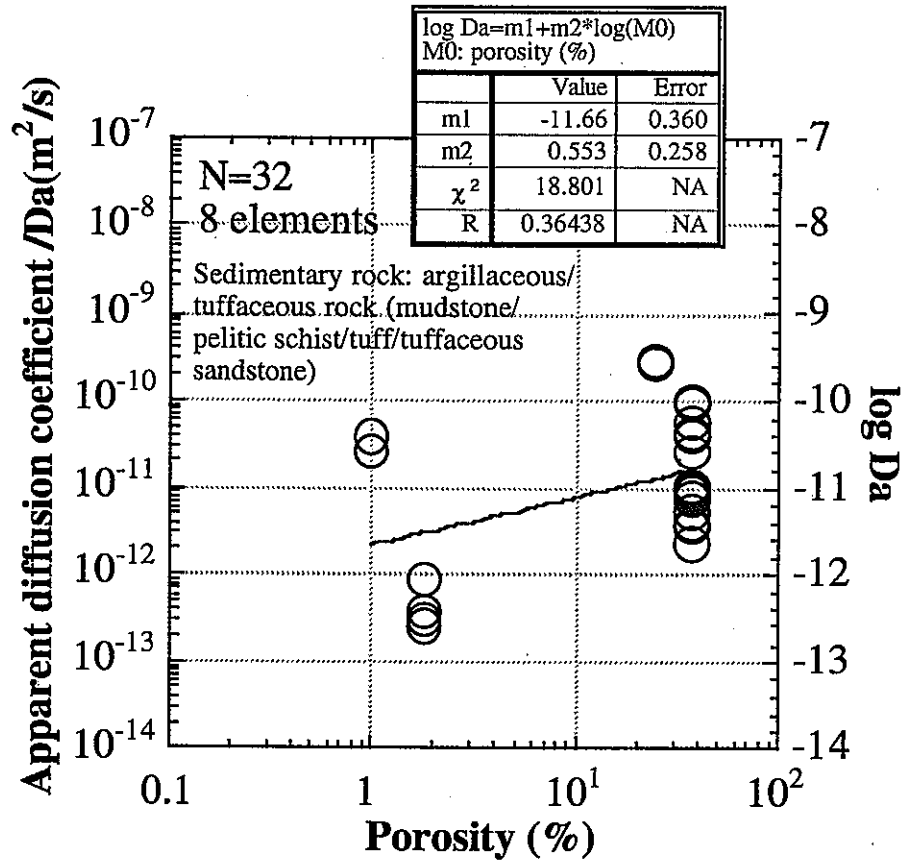


Figure 2.1.10-6 Da values as a function of rock porosity for sedimentary rock (mudstone, pelitic schist, tuff, tuffaceous sandstone)  
A wide range of variation is found in Da, even similar rock porosity.

Figure 2.1.10-7 shows a correlation of De values as a function of charge number of species. No systematic trend is particularly found. Since porosity actually depends on the kind of rock, it is presumed that such a wide range of variation in De was caused by the difference of rock porosity.

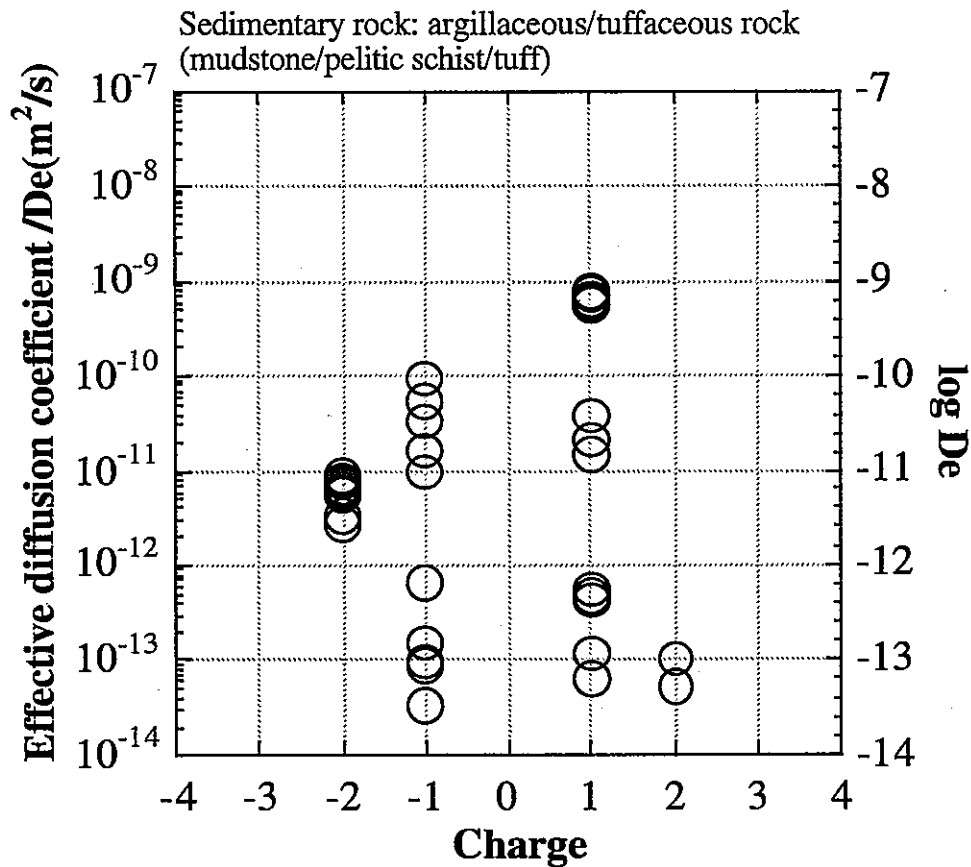


Figure 2.1.10-7 De values as a function of charge number of species  
A wide range of De values is found even though the charge number is the same.

Figure 2.1.10-8 shows a correlation of formation factors (FF) as a function of rock porosity. Similarly to De values, FF clearly shows a tendency to increase with increasing rock porosity.

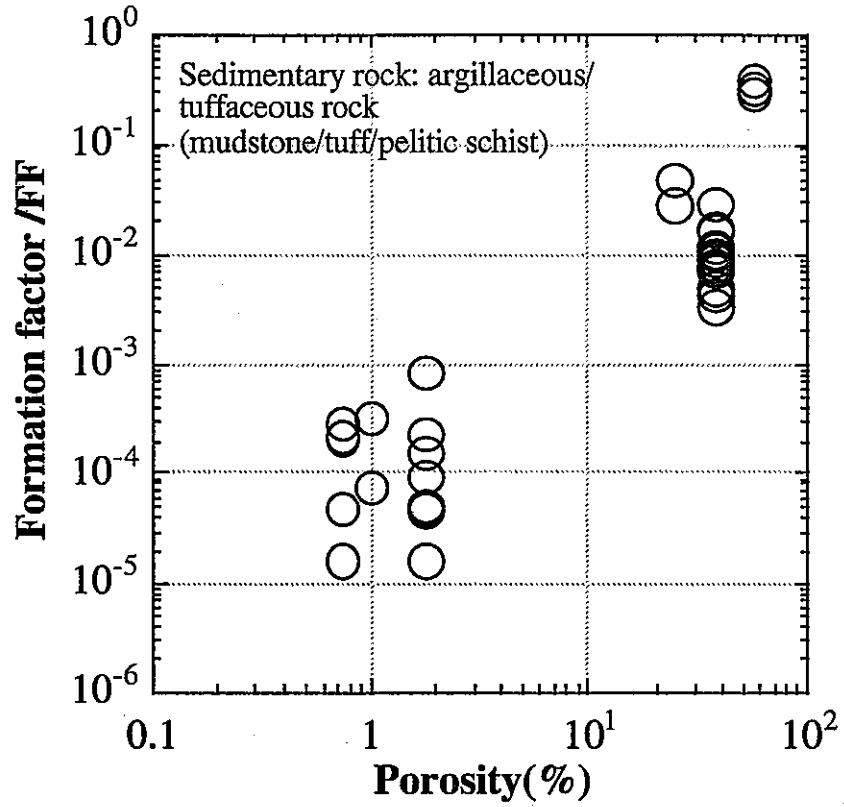


Figure 2.1.10-8 FF values as a function of rock porosity (mudstone, tuff, pelitic schist)  
The FF clearly shows a tendency to increase with increasing rock porosity.

Figure 2.1.10-9 shows a correlation of geometric factors (Gf) as a function of rock porosity. As shown in Figure 2.1.10-9, Gf shows a slight tendency to increase with increasing rock porosity. Since FF clearly increased with increasing rock porosity, that Gf depended on rock porosity indicates that FF strongly depends on both Gf and porosity.

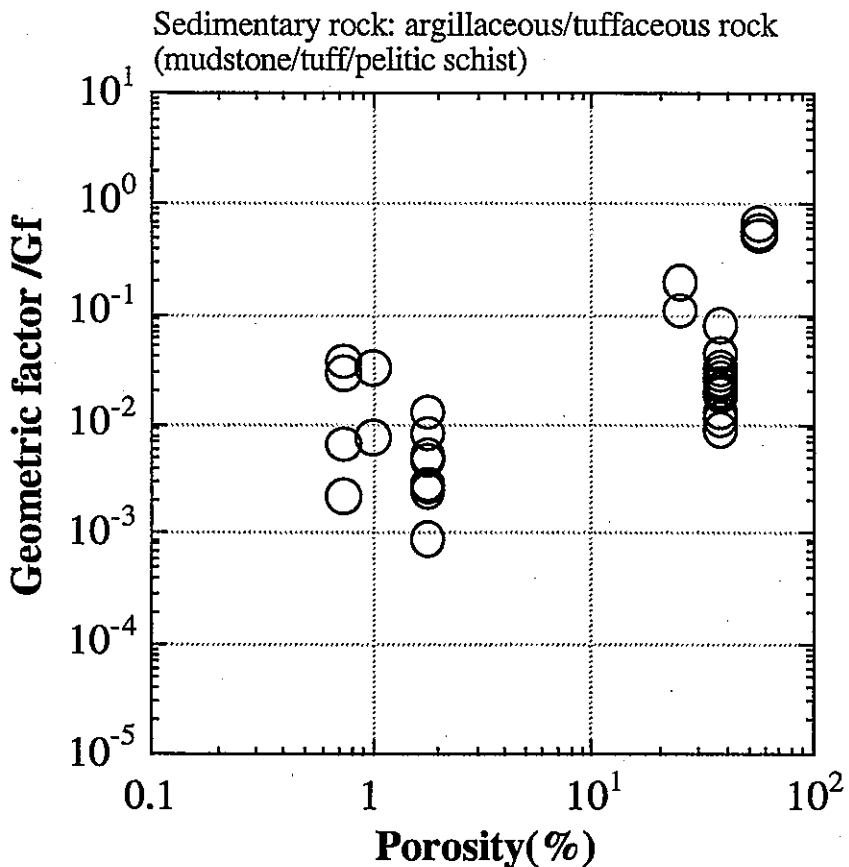


Figure 2.1.10-9 Gf values as a function of rock porosity (mudstone, tuff, pelitic schist) The Gf shows a slight tendency to increase with increasing rock porosity.

Figure 2.1.10-10 shows a plot of  $D_e$  values for individual elements. No particular tendency between  $D_e$  values and elements is found. Even the same element has a wide range of  $D_e$  values (for example, Cl, K, Na). However, the variation in rock porosity is also included in the variation in  $D_e$ . As described in each section,  $D_e$  values for Cl, K and Na increased with increasing rock porosity. Therefore, such wide variation in  $D_e$  is considered to be due to the variation of porosity.

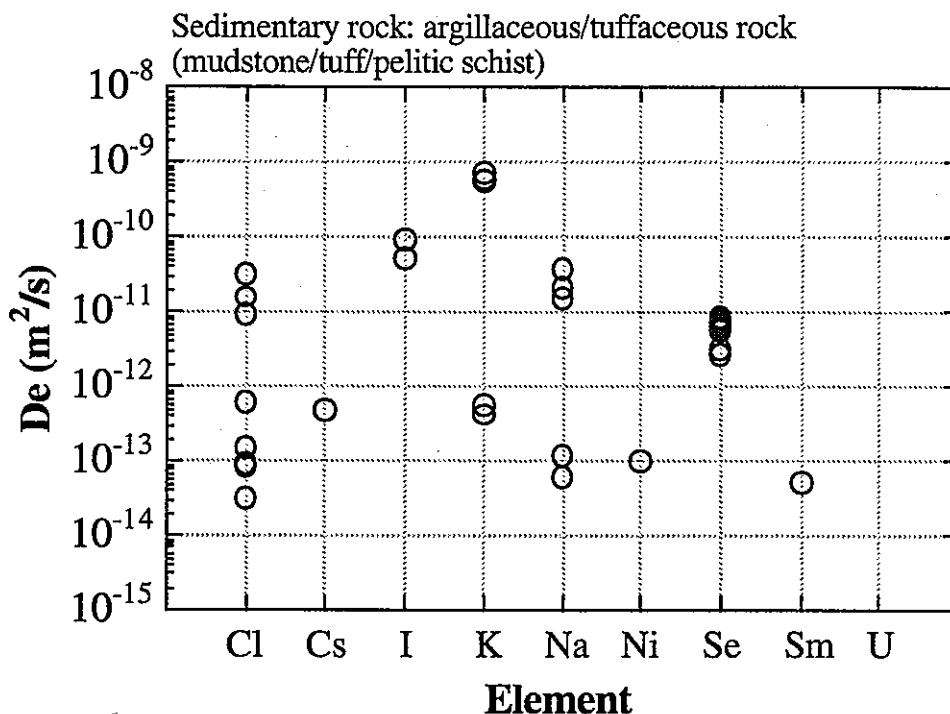


Figure 2.1.10-10 A plot of  $D_e$  values for individual elements in sedimentary rock (mudstone, tuff, pelitic schist)  
No tendency between  $D_e$  values and elements is particularly found.

Figure 2.1.10-11 shows a correlation of  $D_e$  values to  $D_o$  values for sedimentary rock (argillaceous/tuffaceous rock). A wide range of variation in  $D_e$  is found at the same  $D_o$  and no particular correlation seems not to be found between  $D_e$  and  $D_o$ . This reason may be because rock porosity is variable.

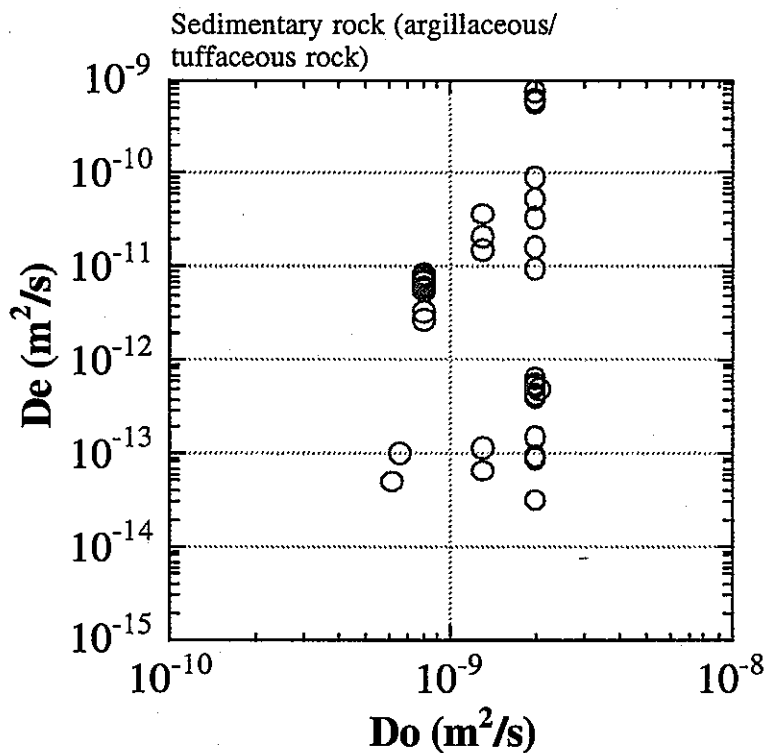


Figure 2.1.10-11 A correlation of  $D_e$  values to  $D_o$  values for sedimentary rock (mudstone, tuff, pelitic schist)

## 2.2 Sedimentary Rock: psammitic rock/sandy stone

Eleven  $D_e$  and 11  $D_a$  values have been totally reported for psammitic rock or sandy stone among sedimentary rock to date based on a review of literatures. Only one element has been discussed for sedimentary rock (psammitic rock/sandy stone). In this section, data regarding matrix diffusion obtained to date (in this case, I (iodine)) are introduced, as well as are discussed.

### 2.2.1 Iodine (I)

The  $D_e$  values for I ( $I^-$ ) in sedimentary rock (psammitic rock/sandy stone) have been reported for siliceous sedimentary rock [Nishiyama et al., 1990]. All experiments for acquisition have been carried out under aerobic conditions. The chemical behaviour and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.3(I).

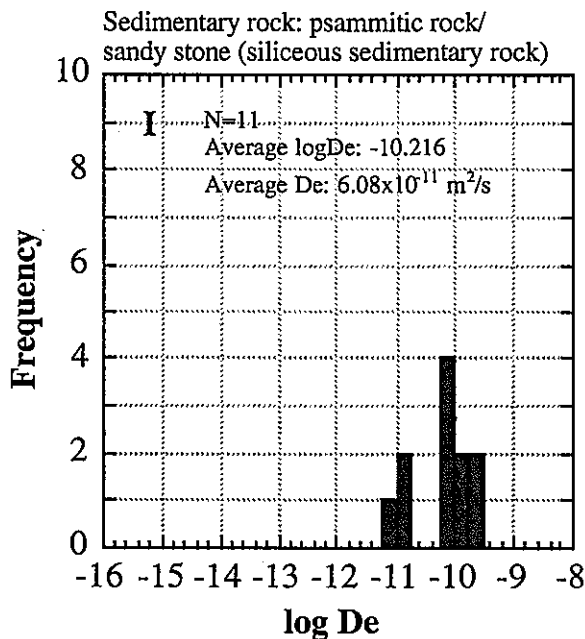
The  $D_e$  data have been obtained by through-diffusion method and  $D_a$  data have been calculated from  $D_e$  values and rock capacity factors reported in literature. The porewater initially saturated is all deionized water. All measurements have been performed at room temperature and no information for temperature is particularly described. The pH of the porewater is also not reported.

Figures 2.2.1-1 and 2.2.1-2 show histograms for  $D_e$  and  $D_a$  values of  $I^-$  in sedimentary rock (psammitic rock/sandy stone) reported up to the present, respectively. Both diffusive parameters ( $D_e$  and  $D_a$ ) of  $I^-$  in sedimentary rock (psammitic rock/sandy stone) have been reported only for siliceous sedimentary rock. Totally, 11  $D_e$  and 11  $D_a$  data have been reported.

Figures 2.2.1-3 and 2.2.1-4 show plots of  $D_e$  and  $D_a$  values of  $I^-$  as a function of rock porosity, respectively. The  $D_e$  clearly shows a tendency to increase with increasing rock porosity. Whilst, significant porosity dependency is not found for  $D_a$ . However, it seems that the lower porosity is, the lower  $D_a$  is.

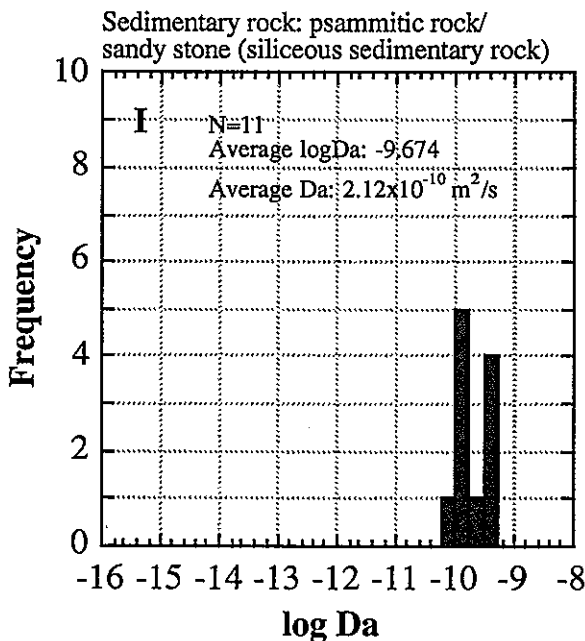
The  $D_0$  of  $I^-$  is estimated to be  $2.0 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C by the Nernst-Einstein equation [Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991; Marcus, 1997]. Formation factor (FF) and geometric factor (Gf) were also calculated based on  $D_0$ ,  $D_e$  and porosity in the same way as section 2.1. Porosity dependencies for FF and Gf will be also discussed in more detail in next section.





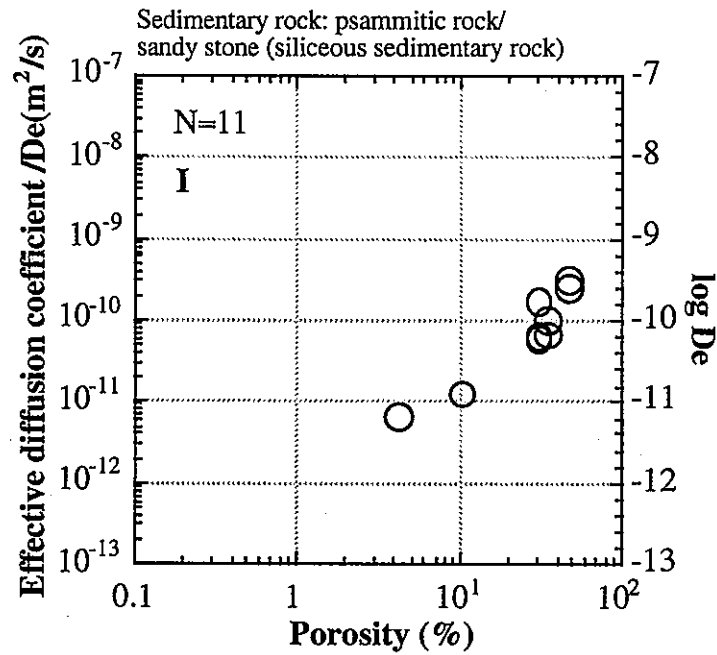
**Figure 2.2.1-1** Histogram for  $D_e$  values of  $I^-$  ion in sedimentary rock (siliceous sedimentary rock)

The average  $D_e$  of  $I^-$  in sedimentary rock (psammitic rock/sandy stone) was estimated to be  $6.08 \times 10^{-11} \text{ m}^2/\text{s}$ .



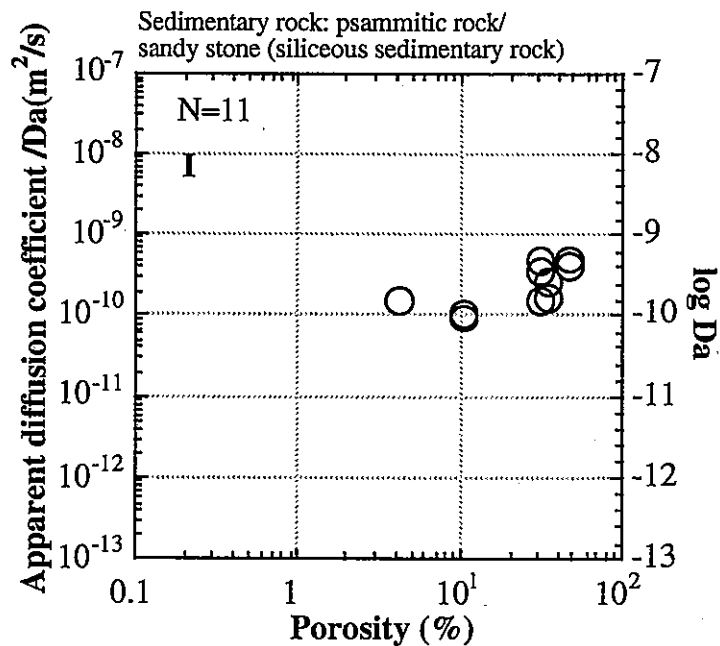
**Figure 2.2.1-2** Histogram for  $D_a$  values of  $I^-$  ion in sedimentary rock (siliceous sedimentary rock)

The average value of  $D_a$  in sedimentary rock (psammitic rock/sandy stone) was estimated to be  $2.12 \times 10^{-10} \text{ m}^2/\text{s}$ .



**Figure 2.2.1-3**  $D_e$  values of  $\Gamma^-$  ion as a function of rock porosity for sedimentary rock (siliceous sedimentary rock)

Totally, 11 plots have been reported for  $\Gamma^-$ . The  $D_e$  clearly shows a tendency to increase with increasing rock porosity.



**Figure 2.2.1-4**  $D_a$  values of  $\Gamma^-$  ion as a function of rock porosity for sedimentary rock (siliceous sedimentary rock)

Totally, 11 plots have been reported for  $\Gamma^-$ . No significant porosity dependency is found for  $D_a$ , but it seems that the lower porosity is, the lower  $D_a$  is.

## References

- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo, Japan (1975) [In Japanese].
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.169.
- Nishiyama, K., Nakashima, S., Tada, R. and Uchida, T. (1990): Diffusion of an Ion in Rock Pore Water and Its Relation to Pore Characteristics, Mining Geology, 40 (5), pp.323~336 [In Japanese].
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.

## 2.2.2 Summary for diffusivity data in psammitic rock/sandy stone

Totally, 1 element was discussed in section 2.2.1. Figure 2.2.2-1 shows a histogram of  $De$  values in psammitic rock or sandy stone of sedimentary rock for 1 element (I) reported in section 2.2.1(I). Though the average  $De$  of all data in sedimentary rock (psammitic rock/sandy stone) was estimated to be  $6.08 \times 10^{-11} \text{ m}^2/\text{s}$ , not so wide range of variation is found in  $De$ . The width of variation in  $De$  is within two orders of magnitude. This attributes to the variation of rock porosity. In these rocks, the porosity of siliceous sedimentary rock had a range from 4.3 to 47.5 %, showing a wide range of porosity. Therefore,  $De$  should be discussed together with rock porosity.

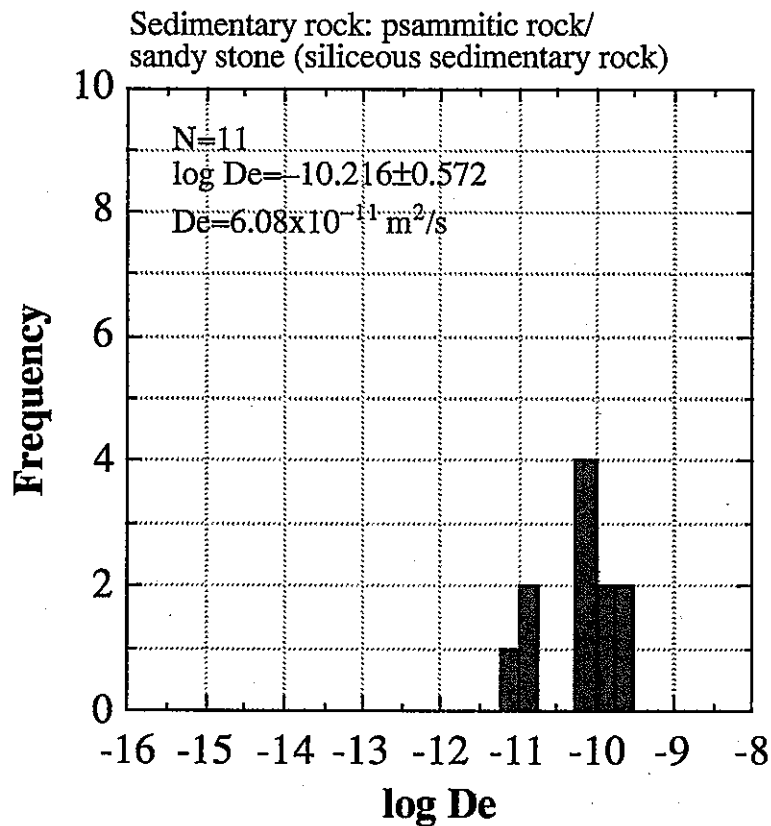


Figure 2.2.2-1 Histogram for  $De$  values in sedimentary rock for one element (for I) (siliceous sedimentary rock)

Figure 2.2.2-2 shows a histogram of Da values in sedimentary rock (psammitic rock/sandy stone) for 1 element discussed in section 2.2.1(I). The average value of Da for siliceous sedimentary rock in sedimentary rock was estimated to be  $2.12 \times 10^{-10} \text{ m}^2/\text{s}$  and the width of variation in Da is within one order of magnitude. The Da also includes sorption onto minerals composing rock, so the difference in Da can not be simply explained only by porosity and pore structure. The Da is mainly affected by both sorption and De. Since De depends on the pore structure of rock, and sorption also depends on the kind of rock and chemical conditions, it is presumed that the variation in Da was caused by the differences of constituent minerals and pore structure by the difference of the kind of rock.

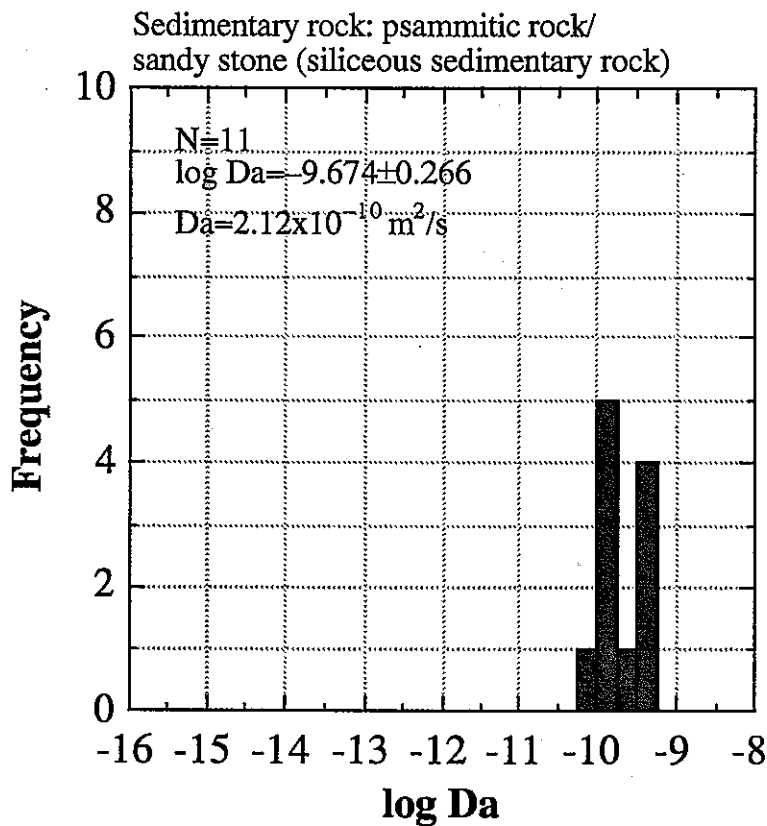


Figure 2.2.2-2 Histogram for Da values in sedimentary rock for one element (for I) (siliceous sedimentary rock)

The average value of Da in sedimentary rock (psammitic rock/sandy stone) was estimated to be  $2.12 \times 10^{-10} \text{ m}^2/\text{s}$ . The width of variation in Da is within one order of magnitude.

Figure 2.2.2-3 shows a porosity dependence of  $D_e$  values discussed in section 2.2.1(I). Totally, 11 data have been reported for  $D_e$  together with porosity. The  $D_e$  clearly shows a tendency to increase with increasing rock porosity.

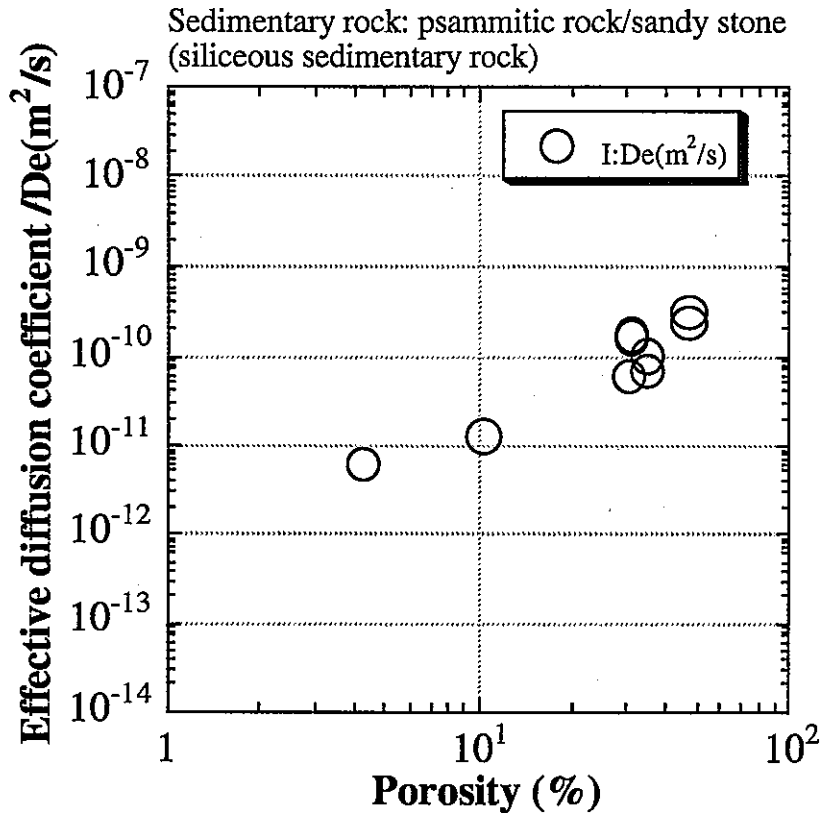


Figure 2.2.2-3  $D_e$  values of  $\Gamma$  ion as a function of porosity for sedimentary rock (siliceous sedimentary rock)

The  $D_e$  data for siliceous sedimentary rock in sedimentary rock have been reported only for I.  $D_e$  clearly shows a tendency to increase with increasing rock porosity.

Figure 2.2.2-4 shows a least squares fitting curve for  $D_e$  values as a function of porosity. The  $D_e$  is calculated to be  $6.5 \times 10^{-10} \text{ m}^2/\text{s}$  at a porosity of 100 % (in this  $D_e$  is equivalent to  $D_0$ ), being near  $2.0 \times 10^{-9} \text{ m}^2/\text{s}$  at 25 °C which is the  $D_0$  of  $\Gamma$ . From this fitting, the experimental equation:  $(10^{-12.43 \pm 0.278}) \cdot \phi^{1.62 \pm 0.198}$  was derived, where  $\phi$  is the porosity in % and the errors stand for the standard deviations.

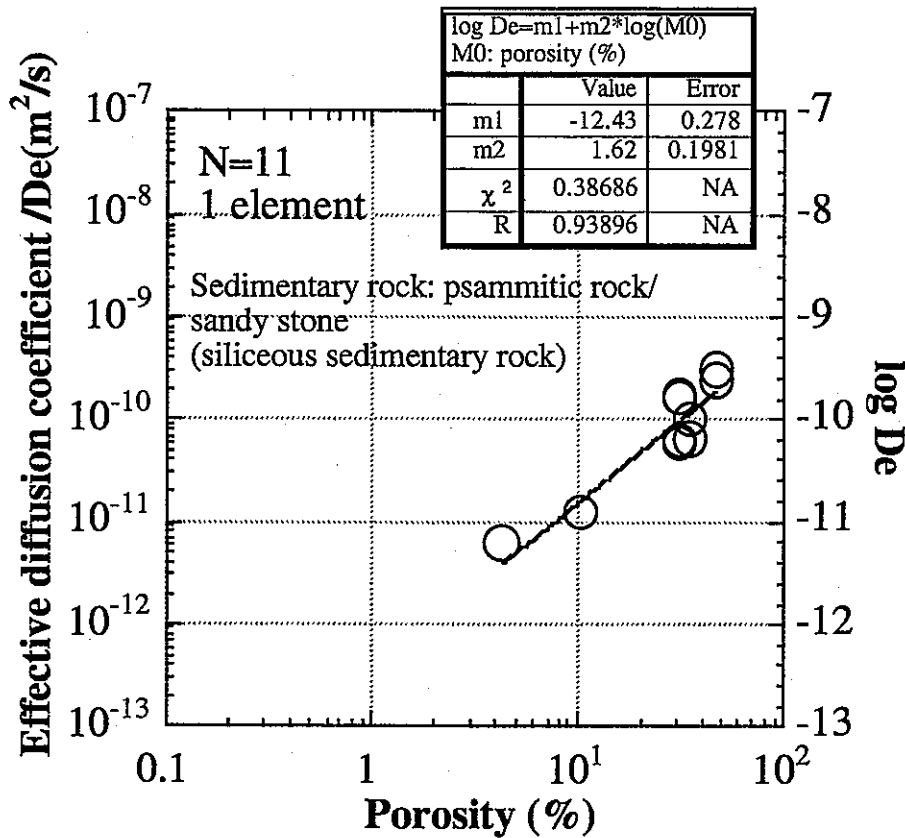


Figure 2.2.2-4 Empirical equation for  $D_e$  values as a function of rock porosity for sedimentary rock (siliceous sedimentary rock)  
 Good correlativity is found between  $D_e$  and porosity.

Figure 2.2.2-5 shows a width of variation in De values as a function of rock porosity. Two lines were drawn to be parallel to the empirical equation of De values as a function of porosity and to pass on the maximum and minimum values of plotted De values. Since good correlativity is originally found between De and porosity, the width of variation in De is small, as shown in Figure 2.2.2-5.

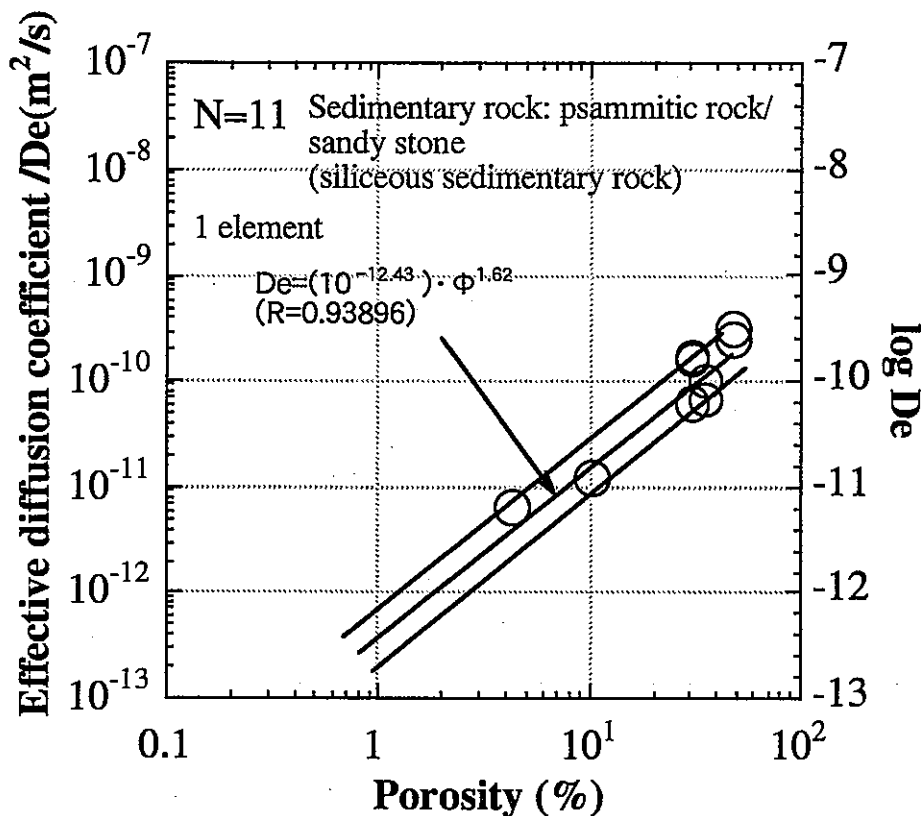


Figure 2.2.2-5 A width of variation in De values as a function of porosity for sedimentary rock (siliceous sedimentary rock)  
 Since good correlativity is found between De and porosity, the width of variation in De is small.



Figure 2.2.2-6 shows a least squares fitting curve for Da values as a function of rock porosity. Though Da shows a slight tendency to increase with increasing rock porosity, clear trend is not found than the case of De. Not so wide range of variation is found in Da.

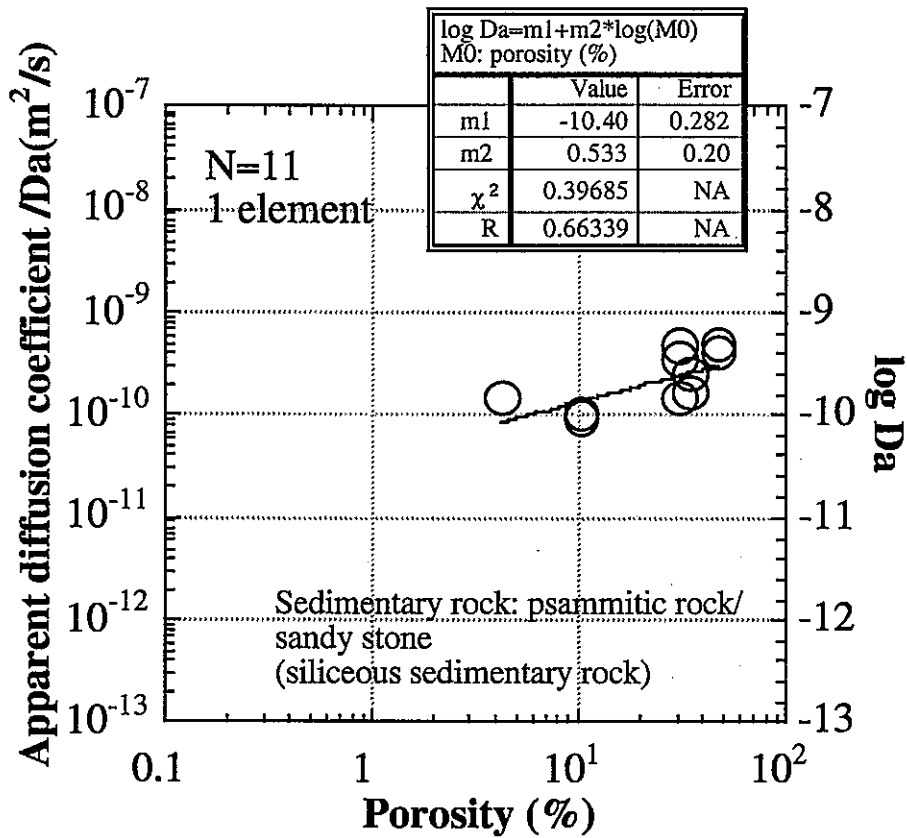


Figure 2.2.2-6 Da values as a function of rock porosity for sedimentary rock (siliceous sedimentary rock)

Though Da shows a slight tendency to increase with increasing rock porosity, it is not clear than the case of De.

Figure 2.2.2-7 shows a correlation of  $D_e$  values as a function of charge number of species. In this case, since all data are for  $I^-$ , it can not be discussed for the effect of ionic charge of species here. Though some variation is found in  $D_e$ , this is considered to be due to the difference in rock porosity.

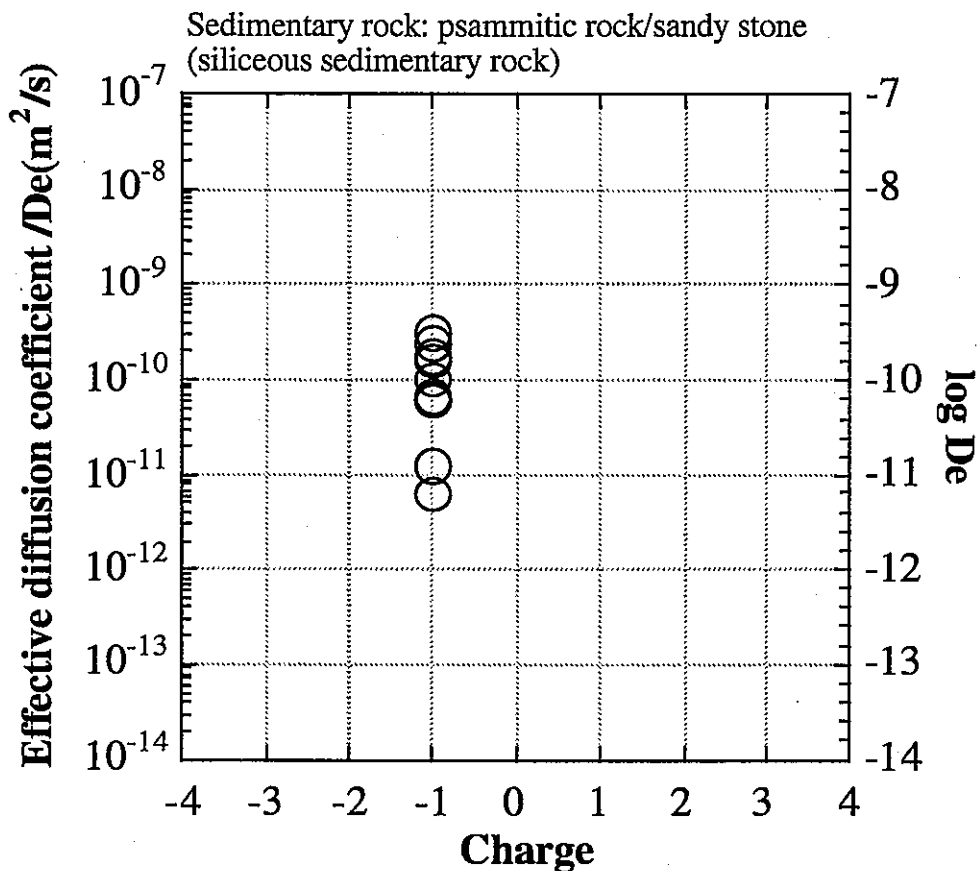


Figure 2.2.2-7  $D_e$  values as a function of charge number of species  
Though some variation is found in  $D_e$ , this may be due to the variation of rock porosity.

Figure 2.2.2-8 shows a correlation of formation factors (FF) as a function of rock porosity. Similarly to De values, FF clearly shows a tendency to increase with increasing rock porosity.

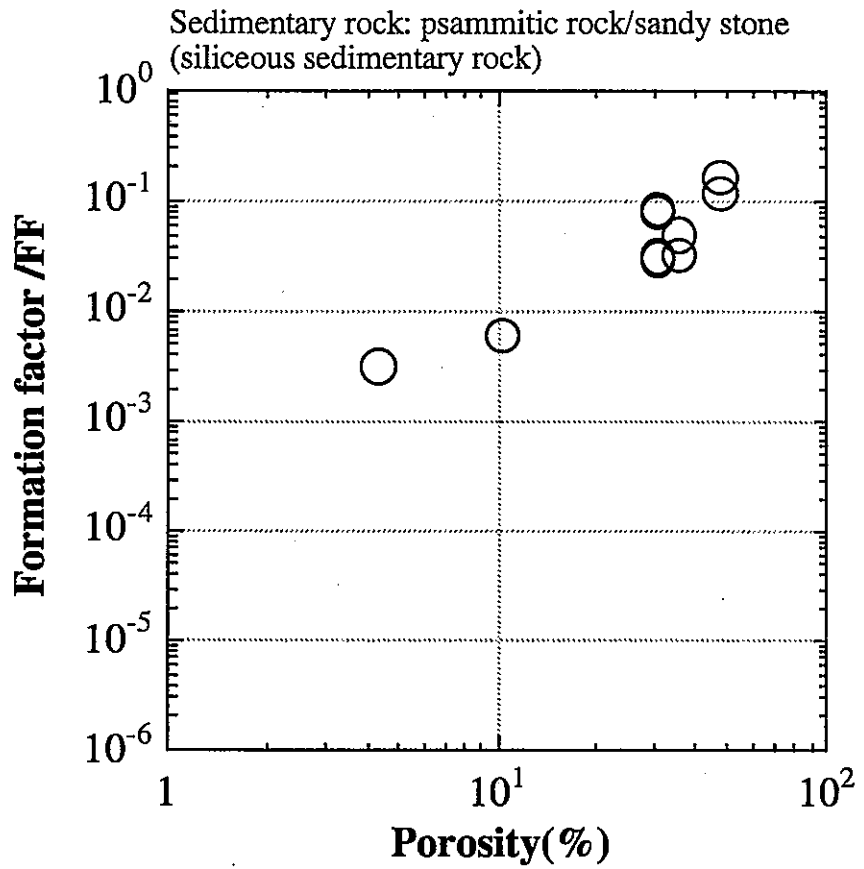


Figure 2.2.2-8 FF values as a function of rock porosity for sedimentary rock (siliceous sedimentary rock)

The FF also clearly shows a tendency to increase with increasing rock porosity, similarly to De.

Figure 2.2.2-9 shows a correlation of geometric factors (Gf) as a function of rock porosity. The Gf shows a slight tendency to increase with increasing rock porosity. Since FF clearly increased with increasing rock porosity, that Gf depended on rock porosity indicates that FF strongly depends on both Gf and porosity.

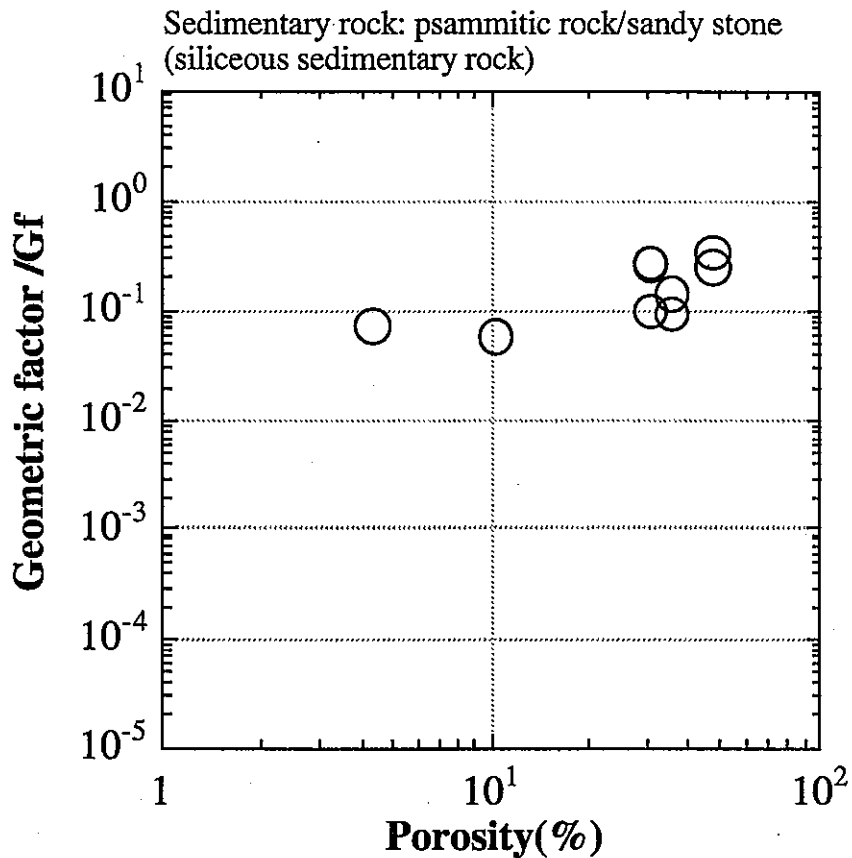


Figure 2.2.2-9 Gf values as a function of rock porosity for sedimentary rock (siliceous sedimentary rock)

The Gf shows a slight tendency to increase with increasing rock porosity. Even though the variation in porosity is relatively wide to be from 4.3 to 47.5 %, the range of Gf is within one order of magnitude.

Figure 2.2.2-10 shows a plot of  $De$  values for individual elements. In this case, since reported  $De$  data are only for  $I^-$ , no systematic tendency can not be discussed here. As described in Figure 2.2.2-10, the variation in  $De$  is about 2 orders of magnitude. This may be due to the variation of porosity.

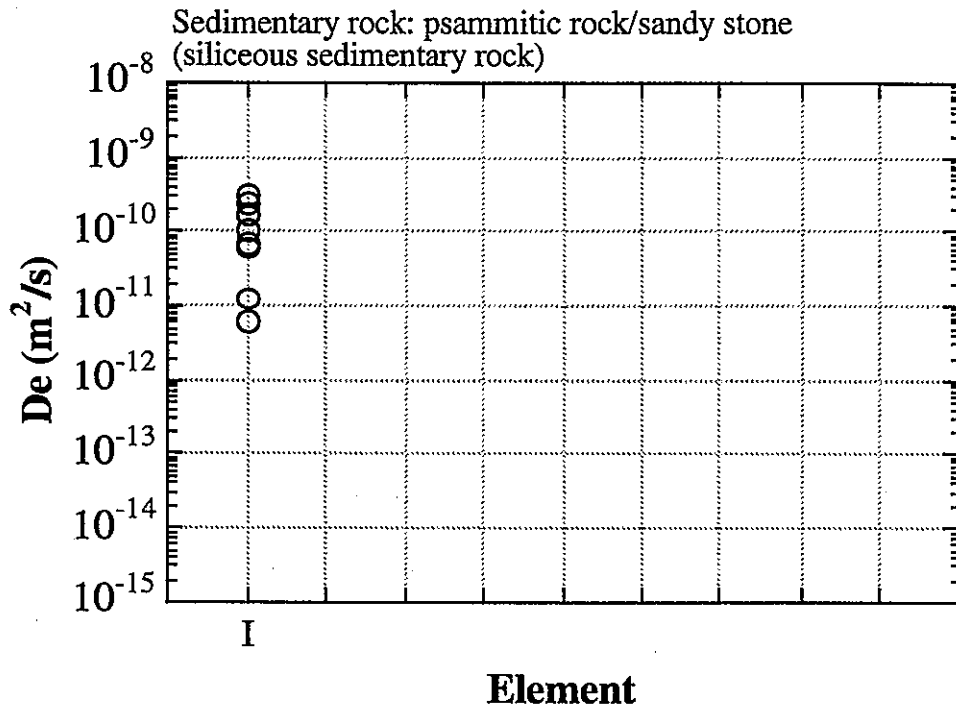


Figure 2.2.2-10 A plot of  $De$  values for individual elements in sedimentary rock (siliceous sedimentary rock)

Figure 2.2.2-11 shows a correlation of  $D_e$  values to  $D_o$  values for sedimentary rock (psammitic rock/sandy stone). For this kind of rock, since data only for I (iodine) have been reported, no variation in  $D_o$  exists. However, some variation in  $D_e$  is found. This variation is considered to be due to the variety of rock porosity, because this kind of rock has a porosity range of 4.3 to 47.5 %. Therefore, correlation between  $D_e$  and  $D_o$  can not be simply discussed here.

In comparison between sedimentary rock (argillaceous/tuffaceous rock) and sedimentary rock (psammitic rock/sandy stone), similar  $D_e$  values are found for similar porosity. From this matter, it is considered that diffusion behaviour to determine  $D_e$  is similar for both sedimentary rocks.

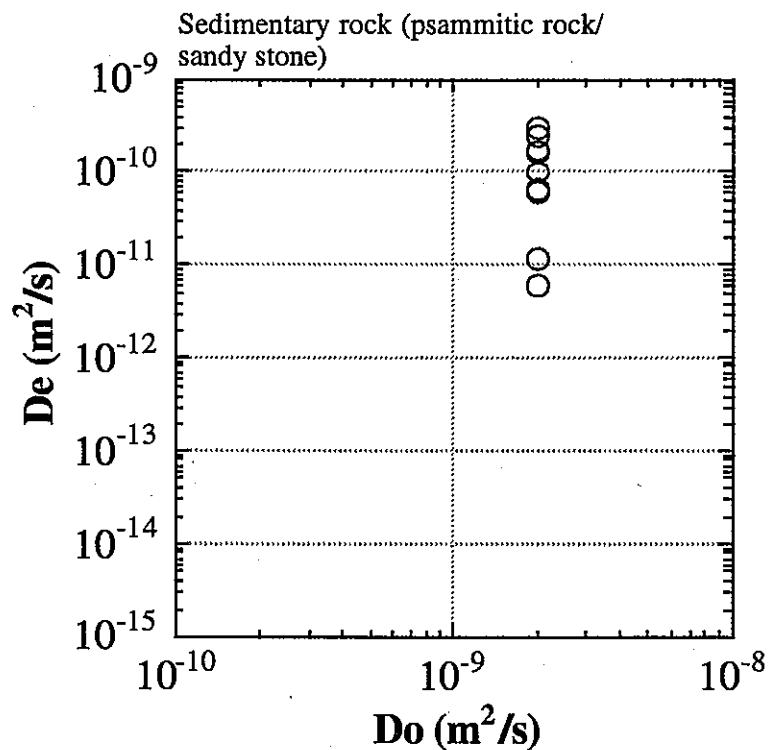


Figure 2.2.2-11 A correlation of  $D_e$  values to  $D_o$  values for sedimentary rock (siliceous sedimentary rock)

Though some variation is found in  $D_e$  even though  $D_o$  is the same, this may be due to the variation of rock porosity.

## 2.3 Alkaline Crystalline Rock

Totally, 32 De and 14 Da data have been reported for alkaline crystalline rock based on a review of literatures. Six elements<sup>1</sup> have been totally discussed for alkaline crystalline rock. In this section, data regarding matrix diffusion obtained for individual elements are introduced, as well as are discussed.

### 2.3.1 Chlorine (Cl)

The De values for Cl ( $\text{Cl}^-$ ) in alkaline crystalline rock have been reported for basalt [Sato et al., 1992, 1997; Sato and Yui, 1992; PNC, 1996]. The chemical behaviour and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.1(Cl).

For basalt, totally, 11 De values, obtained by through-diffusion method, have been reported up to the present. All measurements have been carried out under aerobic conditions. The porewater initially saturated is all distilled water. All measurements have been performed at room temperature or 25 °C. The reported porewater pH was in a range of 5.5 to 7.1.

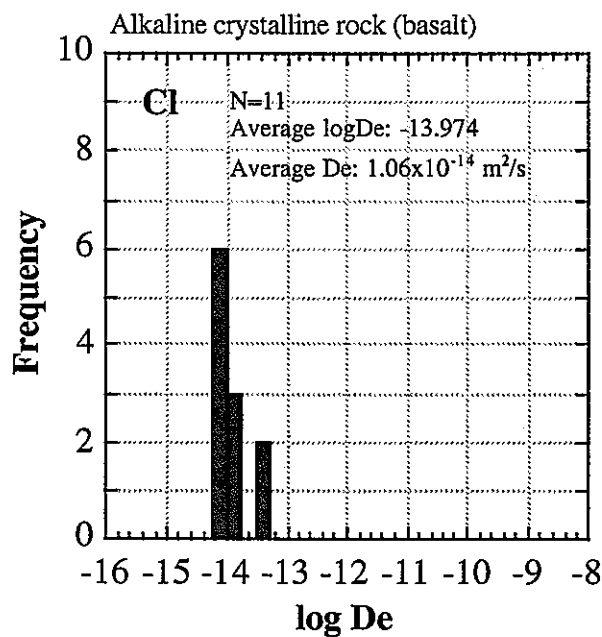
Figures 2.3.1-1 and 2.3.1-2 show histograms for De and Da values of  $\text{Cl}^-$  in alkaline crystalline rock reported to date, respectively. Totally, 11 De data have been reported for basalt. The width of variation in De is within one order of magnitude, but the degree of data is lower than that in acid crystalline rock. For Da, 2 data have been reported in total.

Figures 2.3.1-3 and 2.3.1-4 show plots of De and Da values of  $\text{Cl}^-$  as a function of rock porosity, respectively. The correlations were plotted only for data reported both porosity and De or Da. For De, 10 plots have been totally reported here and 1 plot has been totally reported for Da. Since all De values were obtained for similar porosities, porosity dependency for De can not be discussed here. However, it seems that the higher porosity is, the higher De is. Since Da is only 1 plot, porosity dependency for Da can not be discussed here.

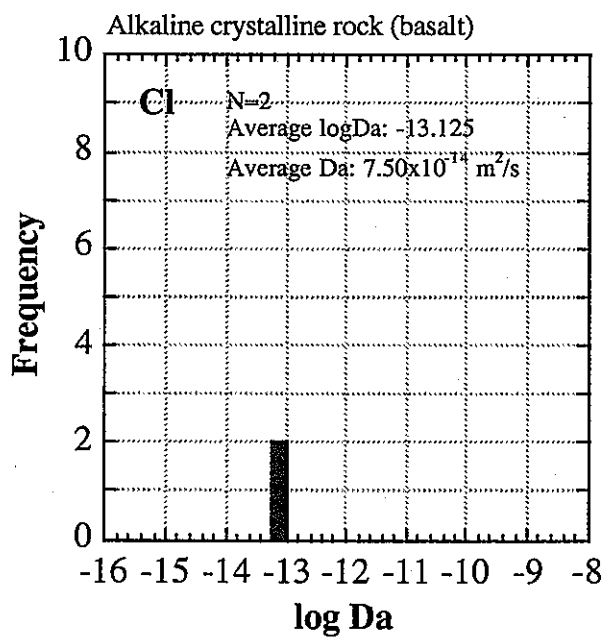
The Do of  $\text{Cl}^-$ , FF and Gf were estimated in the same way as section 2.1.1(Cl).

---

<sup>1</sup> 6 elements to be introduced: Cl, Cs, I, K, Na and Ni.

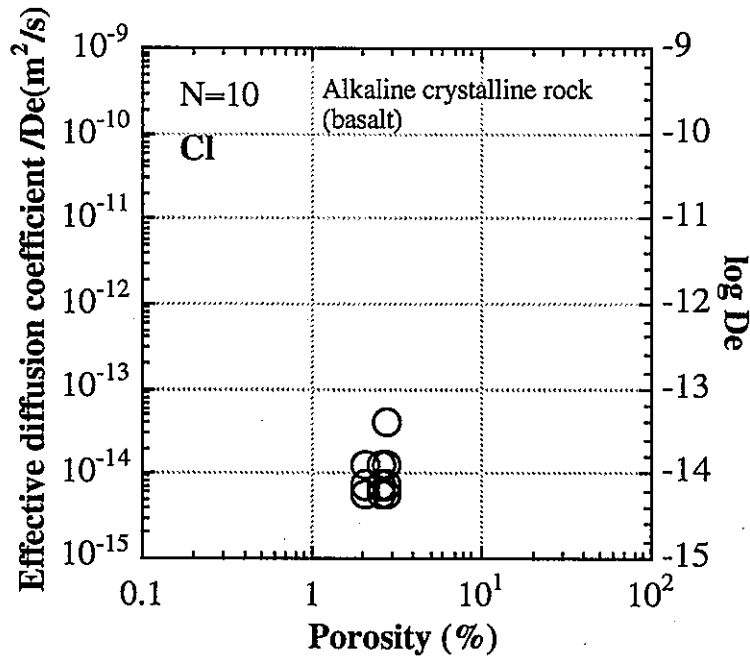


**Figure 2.3.1-1** Histogram for De values of Cl<sup>-</sup> ion in alkaline crystalline rock (basalt)  
The average value of De of Cl<sup>-</sup> in alkaline crystalline rock was estimated to be  $1.06 \times 10^{-14}$  m<sup>2</sup>/s.



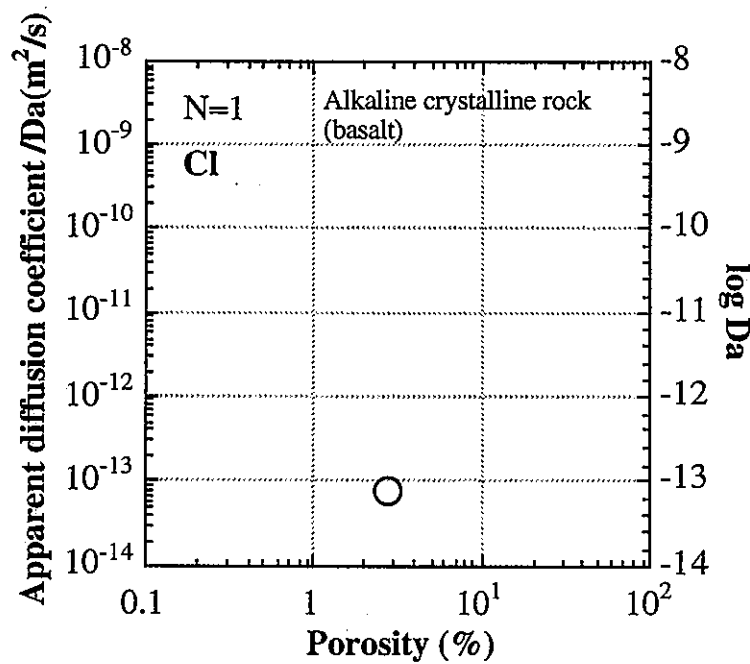
**Figure 2.3.1-2** Histogram for Da values of Cl<sup>-</sup> ion in alkaline crystalline rock (basalt)  
The average value of Da of Cl<sup>-</sup> in alkaline crystalline rock was estimated to be  $7.5 \times 10^{-14}$  m<sup>2</sup>/s.





**Figure 2.3.1-3** De values of Cl<sup>-</sup> ion as a function of rock porosity for alkaline crystalline rock (basalt)

Since all De values are plotted on similar porosities, porosity dependency for De can not be discussed here.



**Figure 2.3.1-4** Da values of Cl<sup>-</sup> ion as a function of rock porosity for alkaline crystalline rock (basalt)

## References

- Sato, H. and Yui, M. (1992): Measurement of Ionic Diffusion Coefficients in Rocks, 1992 Fall Meeting of the Atomic Energy Society of Japan, F15, p.323 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119~133, Elsevier Science B. V.
- Sato, H., Yui, M. and Ishikawa, H. (1992): A Study on Pore Structure of Rocks, PNC Technical Report, PNC TN8410 92-222, Power Reactor and Nuclear Fuel Development Corporation [In Japanese].
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].

### 2.3.2 Cesium (Cs)

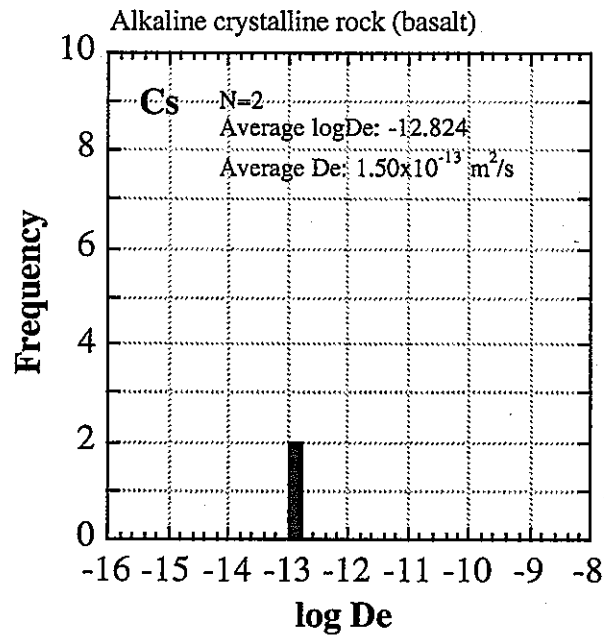
The  $D_e$  values for Cs ( $Cs^+$ ) in alkaline crystalline rock have been reported for basalt [PNC, 1996; Sato, 1997]. Since Cs is not redox sensitive, all measurements have been carried out under aerobic conditions. Cesium is one of the important elements for performance assessment of the geological disposal of HLW. The chemical behaviour and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.2(Cs).

The  $D_e$  data have been obtained by through-diffusion method and  $D_a$  values have been derived based on the time-lag method [e.g. Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor. The porewater initially saturated is all distilled water. All measurements have been performed at 25 °C and no measurement for temperature dependence on  $D_e$  has been especially carried out. The reported porewater pH was 7.1 for all measurements.

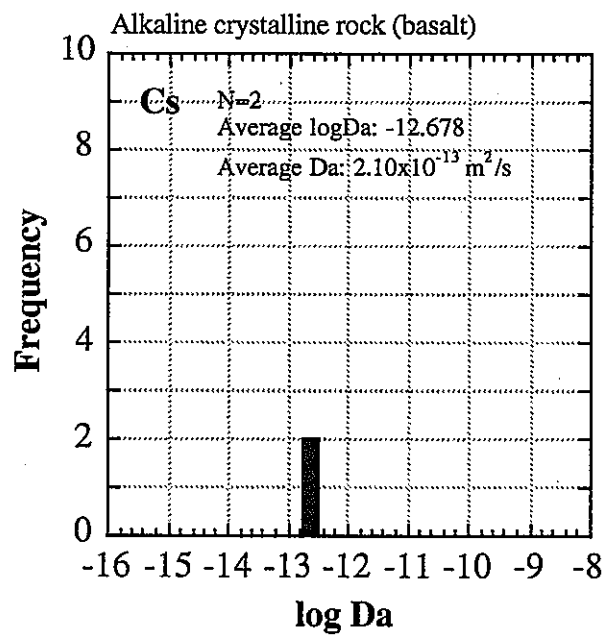
Figures 2.3.2-1 and 2.3.2-2 show histograms for  $D_e$  and  $D_a$  values of  $Cs^+$  in alkaline crystalline rock reported to date, respectively. Totally, 2  $D_e$  data have been reported only for basalt. Also for  $D_a$ , totally, 2 data have been reported for basalt.

Figures 2.3.2-3 and 2.3.2-4 show plots of  $D_e$  and  $D_a$  values as a function of rock porosity for alkaline crystalline rock, respectively. The correlations were plotted only for data reported both porosity and  $D_e$  or  $D_a$ . Only one plot has been reported for both  $D_e$  and  $D_a$ . Porosity dependency for diffusion can not be therefore discussed here.

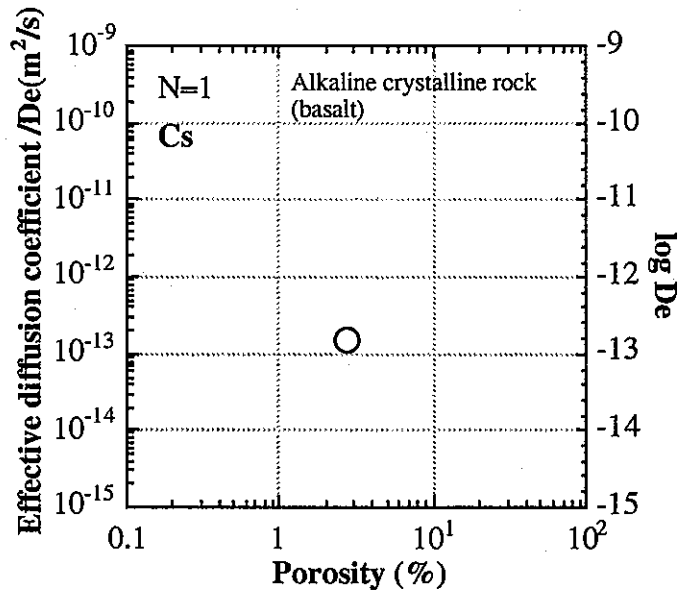
The  $D_o$  of  $Cs^+$ , FF and Gf were estimated in the same way as section 2.1.2(Cs). Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.3.7.



**Figure 2.3.2-1** Histogram for De values of Cs<sup>+</sup> ion in alkaline crystalline rock (basalt)  
 The average value of De of Cs<sup>+</sup> in basalt was estimated to be  $1.50 \times 10^{-13} \text{ m}^2/\text{s}$ .

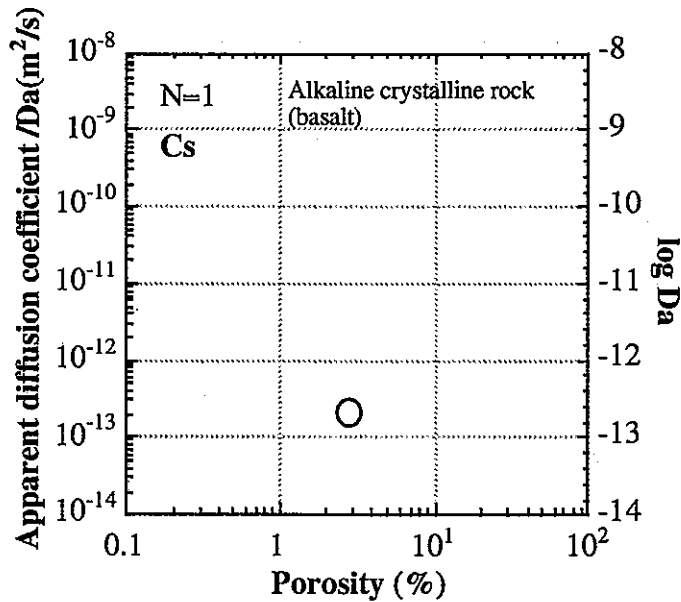


**Figure 2.3.2-2** Histogram for Da values of Cs<sup>+</sup> ion in alkaline crystalline rock (basalt)



**Figure 2.3.2-3**  $D_e$  values of  $Cs^+$  ion as a function of rock porosity for alkaline crystalline rock (basalt)

The correlation was plotted only for data reported both porosity and  $D_e$ . Totally, only one plot has been reported for  $Cs^+$ . Porosity dependency for  $D_e$  can not be therefore discussed here.



**Figure 2.3.2-4**  $D_a$  values of  $Cs^+$  ion as a function of rock porosity for alkaline crystalline rock (basalt)

The correlation was plotted only for data reported both porosity and  $D_a$ . Totally, only one plot has been reported for  $Cs^+$ . Porosity dependency for  $D_a$  can not be therefore discussed here.

## References

- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, **26**, pp.119~133, Elsevier Science B. V.

### 2.3.3 Iodine (I)

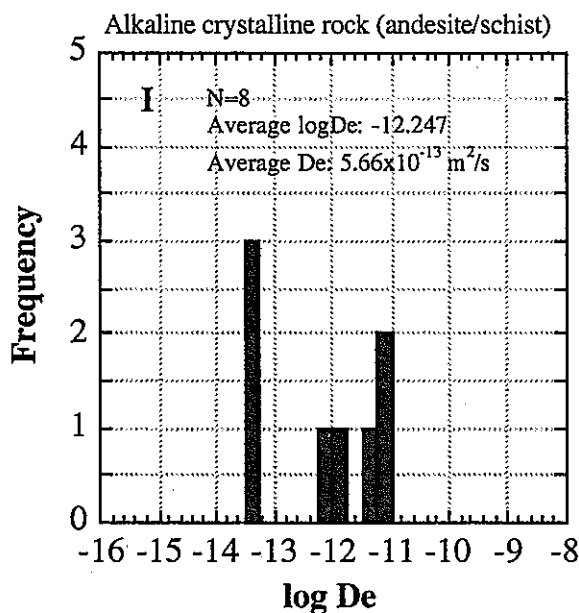
The  $D_e$  values for I ( $I^-$ ) in alkaline crystalline rock have been reported for andesite and schist [Nishiyama et al., 1990]. Since I is not redox sensitive, all measurements have been carried out under aerobic conditions. Iodine is not directly included in important elements for performance assessment of the geological disposal of HLW. The chemical behaviour and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.3(I).

The  $D_e$  data have been obtained by through-diffusion method and  $D_a$  data have been derived based on the time-lag method [e.g. Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor. The porewater initially saturated is all distilled water. All measurements have been performed at room temperature. The pH of the porewater is not reported.

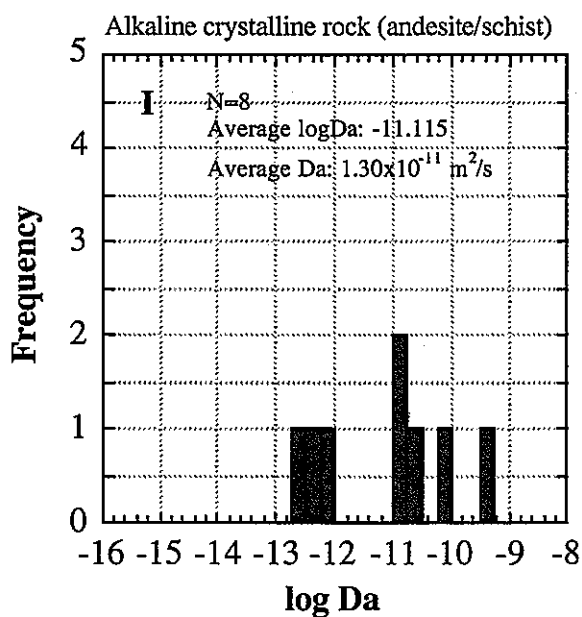
Figures 2.3.3-1 and 2.3.3-2 show histograms for  $D_e$  and  $D_a$  values of  $I^-$  in alkaline crystalline rock reported up to the present, respectively. The  $D_e$  values of  $I^-$  in alkaline crystalline rock have been totally reported for andesite and schist. Eight  $D_e$  data have been totally reported for both rocks (6 data for andesite and 2 data for schist). Also for  $D_a$ , 8 data have been reported in total for both rocks.

Figures 2.3.3-3 and 2.3.3-4 show plots of  $D_e$  and  $D_a$  values of  $I^-$  as a function of rock porosity for alkaline crystalline rock, respectively. The correlations were plotted only for data reported both porosity and  $D_e$  or  $D_a$ . Eight plots have been totally reported for both  $D_e$  and  $D_a$ . Both  $D_e$  and  $D_a$  show a tendency to increase with increasing rock porosity. However, the width of variation in both diffusive parameters is large, as shown in figures.

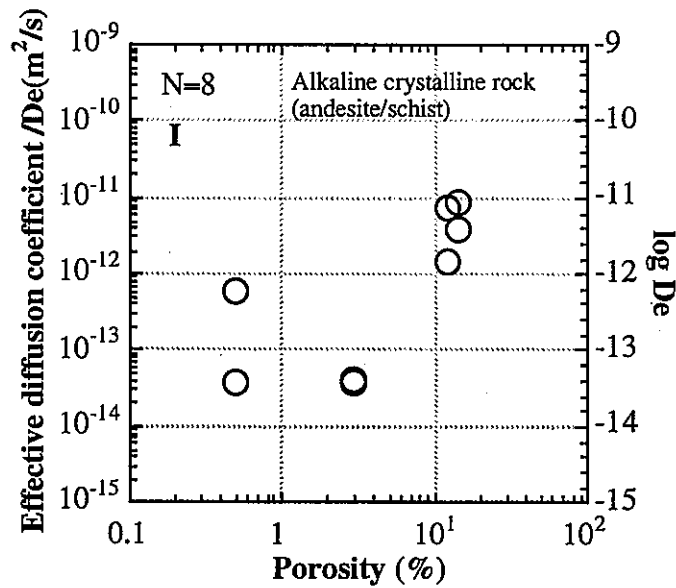
The  $D_o$  of  $I^-$ , FF and Gf were estimated in the same way as section 2.1.3. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.3.7.



**Figure 2.3.3-1** Histogram for De values of I<sup>-</sup> ion in alkaline crystalline rock (andesite, schist) The average value of De of I<sup>-</sup> in alkaline crystalline rock (andesite and schist) was estimated to be  $1.50 \times 10^{-13} \text{ m}^2/\text{s}$ .

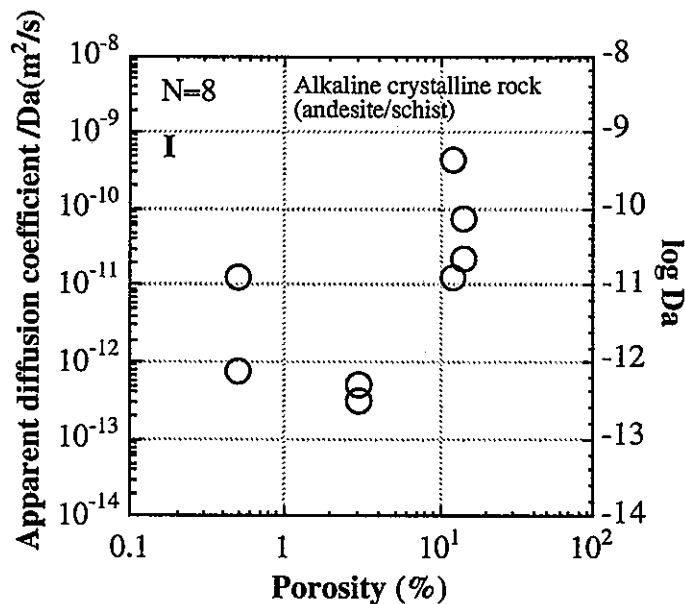


**Figure 2.3.3-2** Histogram for Da values of I<sup>-</sup> ion in alkaline crystalline rock (andesite, schist) The average value of Da of I<sup>-</sup> in alkaline crystalline (andesite and schist) rock was estimated to be  $2.73 \times 10^{-10} \text{ m}^2/\text{s}$ . The range of Da, wide, is over 3 orders of magnitude.



**Figure 2.3.3-3** De values of  $\Gamma^-$  ion as a function of rock porosity for alkaline crystalline rock (andesite, schist)

The correlations were plotted only for data reported both porosity and De. Totally, 8 plots have been reported for  $\Gamma^-$ . The De shows a tendency to increase with increasing rock porosity, but it is not clear, showing a wide range of variation in De.



**Figure 2.3.3-4** Da values of  $\Gamma^-$  ion as a function of rock porosity for alkaline crystalline rock (andesite, schist)

The correlations were plotted only for data reported both porosity and Da. Totally, 8 plots have been reported for  $\Gamma^-$ . The Da shows a tendency to increase with increasing rock porosity, but it is also unclear, showing a wide range of variation in Da.

## References

- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Nishiyama, K., Nakashima, S., Tada, R. and Uchida, T. (1990): Diffusion of an Ion in Rock Pore Water and Its Relation to Pore Characteristics, Mining Geology, **40** (5), pp.323~336 [In Japanese].

### 2.3.4 Potassium (K)

The  $D_e$  values for K ( $K^+$ ) in alkaline crystalline rock have been reported for basalt [Conca et al., 1993; Conca and Wright, 1992]. Since K is not redox sensitive, all measurements have been carried out under aerobic conditions. Potassium is not especially regarded as an important element in performance assessment of the geological disposal of HLW, but it is presumed that K shows similar behaviour to Cs in solution. The chemical behaviour is not affected by redox condition, so its behaviour does not also change in reducing conditions.

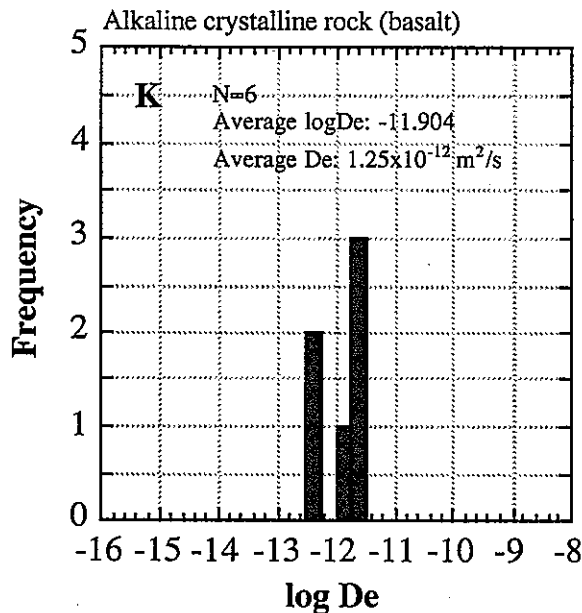
The  $D_e$  data have been obtained by electrical conductivity method. The  $D_a$  can not be obtained by this method. Rock samples were initially saturated with a 0.1 M KCl solution and then electrical conductivities were measured. The  $D_e$  values were determined by the Nernst-Einstein equation [Robinson and Stokes, 1959]. The principle of this method is as described in section 1.3.6. The porewater and tracer are therefore the same. All measurements have been performed at room temperature. The pH of the porewater is not reported.

Figure 2.3.4-1 shows a histogram for  $D_e$  values of  $K^+$  in alkaline crystalline rock reported up to the present. Six  $D_e$  data have been totally reported for basalt. But, no  $D_a$  has been reported.

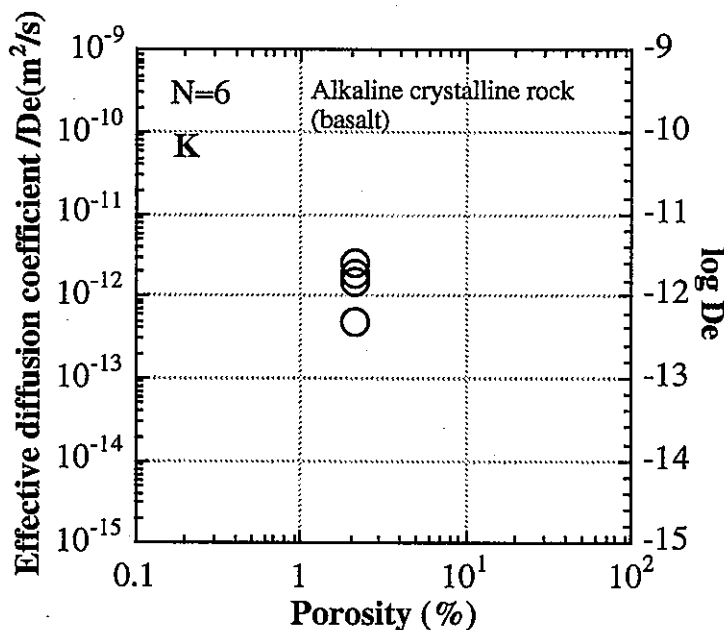
Figure 2.3.4-2 shows a plot of  $D_e$  values of  $K^+$  as a function of rock porosity for alkaline crystalline rock. Totally, 6 plots have been reported for  $D_e$ . However, since all porosities are the same, porosity dependence for  $D_e$  can not be discussed here.

The  $D_o$  of  $K^+$ , FF and Gf were estimated in the same way as section 2.1.4(K). Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.3.7.





**Figure 2.3.4-1** Histogram for De values of K<sup>+</sup> ion in alkaline crystalline rock (basalt)  
 The average value of De of K<sup>+</sup> in alkaline crystalline rock was estimated to be  $1.25 \times 10^{-12} \text{ m}^2/\text{s}$ .  
 The width of variation in De is within one order of magnitude.



**Figure 2.3.4-2** De values of K<sup>+</sup> ion as a function of rock porosity for alkaline crystalline rock (basalt)  
 The correlations were plotted only for data reported both porosity and De. Totally, 6 plots have been reported for K<sup>+</sup>. Since all porosities are the same, porosity dependency for De can not be evaluated.

## References

- Conca, J. L., Apted, M. J. and Arthur, R. C. (1993): Direct Determinations of Transport Parameters in Repository Materials, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVI (Interrante, C. G. and Pabalan, R. T. eds.), pp.839~844, Pittsburgh, Pennsylvania, U.S.A.
- Conca, J. L. and Wright, J. (1992): Direct Determinations of Unsaturated Flow and Transport, Proceedings of the Twelfth Annual American Geophysical Union Hydrology Days Meeting, Fort Collins, March 1992, pp.103~116.
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.

### 2.3.5 Sodium (Na)

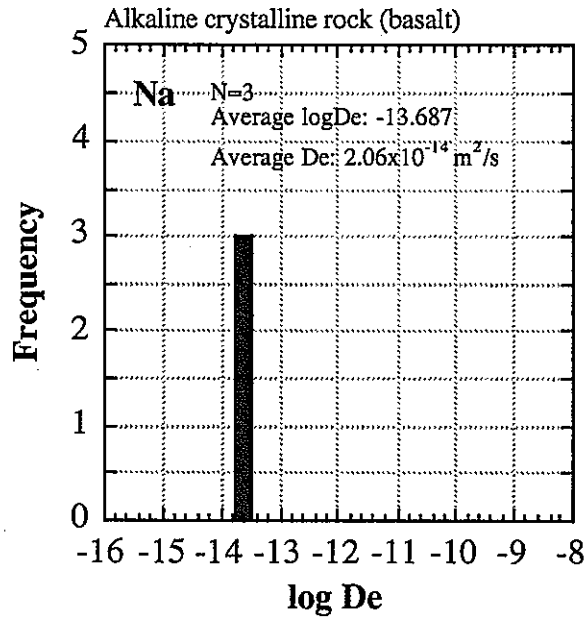
The  $D_e$  values for Na ( $\text{Na}^+$ ) in alkaline crystalline rock have been reported for basalt [Sato et al., 1997]. All measurements have been carried out under aerobic conditions. Sodium is not included in important elements for performance assessment of the geological disposal of HLW. However, Na chemically shows similar behaviour to alkaline metals such as  $\text{Cs}^+$ . The chemical behaviour and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.5(Na).

The  $D_e$  data for Na have been obtained by through-diffusion method, but no  $D_a$  datum has been reported, because all experiments were originally carried out to derive FF values from  $D_e$  values and  $D_o$ . The porewater initially saturated is distilled water for all measurements. All measurements have been performed at 25 °C. The reported porewater pH was all 5.5. Sodium chloride (10 % NaCl) was used as a tracer solution for the measurements.

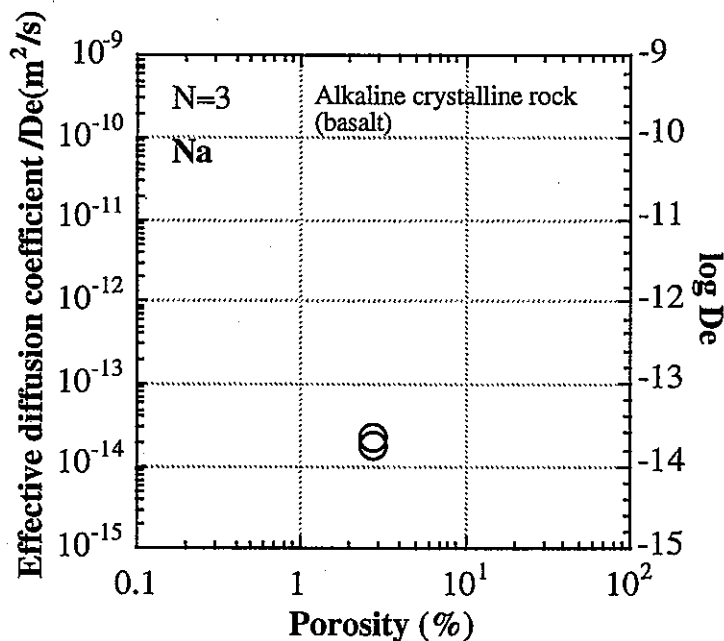
Figure 2.3.5-1 shows a histogram for  $D_e$  values of  $\text{Na}^+$  in alkaline crystalline rock reported up to the present. The  $D_e$  values of  $\text{Na}^+$  in alkaline crystalline rock have been reported for basalt. Totally, 3  $D_e$  data have been reported for Na. All  $D_e$  values were approximately the same.

Figure 2.3.5-2 shows a plot of  $D_e$  values of  $\text{Na}^+$  as a function of rock porosity for alkaline crystalline rock. Totally, 3 plots have been reported for  $D_e$ . However, no datum has been reported for  $D_a$  and  $D_a$  can not be also estimated from  $D_e$  and rock capacity factor, because no rock capacity factor has been reported. Data enough to be able to evaluate porosity dependency are not reported for  $D_e$ .

The  $D_o$  of  $\text{Na}^+$ , FF and Gf were estimated in the same way as section 2.1.5(Na). Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.3.7.



**Figure 2.3.5-1** Histogram for De values of Na<sup>+</sup> ion in alkaline crystalline rock (basalt)  
 The average value of De of Na<sup>+</sup> in alkaline crystalline rock was estimated to be  $2.06 \times 10^{-14} \text{ m}^2/\text{s}$ .  
 Three De values were approximately the same.



**Figure 2.3.5-2** De values of Na<sup>+</sup> ion as a function of rock porosity for alkaline crystalline rock (basalt)  
 The correlations were plotted only for data reported both porosity and De. Totally, 3 plots have been reported for Na<sup>+</sup>. Porosity dependency for De can not be discussed here, because all De data were obtained for similar porosities.

## References

Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, *Journal of Contaminant Hydrology*, **26**, pp.119~133, Elsevir Science B. V.

### 2.3.6 Nickel (Ni)

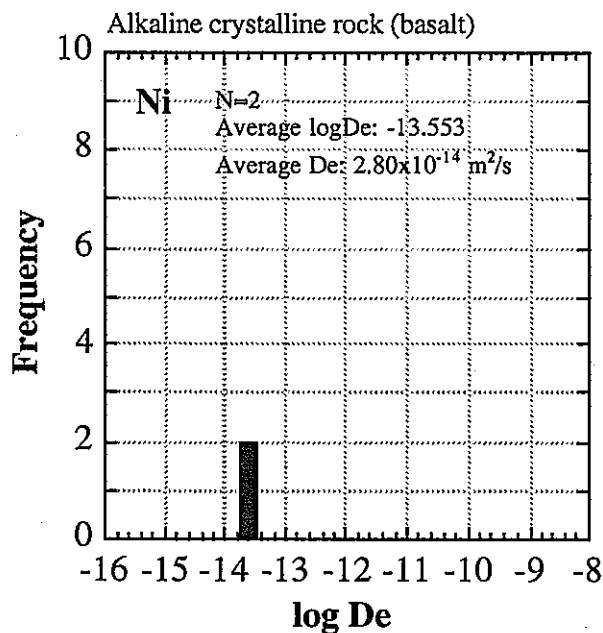
The  $D_e$  values for Ni ( $Ni^{2+}$ ) in alkaline crystalline rock have been reported for basalt [PNC, 1996; Sato et al., 1997]. All measurements have been carried out under aerobic conditions, because Ni is not sensitive to redox condition. Nickel is accounted one of the important elements for performance assessment of the geological disposal of HLW. The chemical behaviour and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.6(Ni).

The  $D_e$  data have been obtained by through-diffusion method and  $D_a$  values have been derived based on the time-lag method [e.g. Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor. All measurements have been carried out in a distilled water system. All experiments have been performed at 25 °C. The reported porewater pH was 7.1 for all measurements. The species of Ni in the porewater in this condition is considered to be  $Ni^{2+}$ . The species of Ni is rather affected by the pH of the porewater, as described in section 2.1.6.

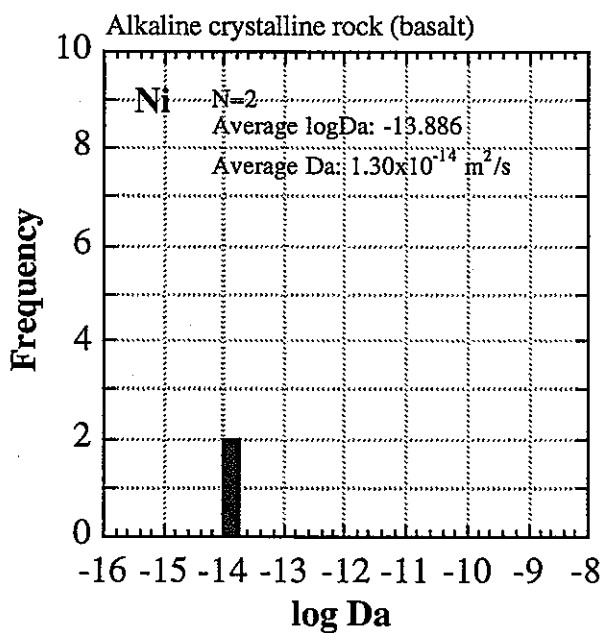
Figures 2.3.6-1 and 2.3.6-2 show histograms for  $D_e$  and  $D_a$  values of  $Ni^{2+}$  in alkaline crystalline rock reported up to the present, respectively. The  $D_e$  values of  $Ni^{2+}$  have been reported only for basalt. Two  $D_e$  data have been reported in total. Also for  $D_a$ , 2 data have been totally reported for basalt.

Figures 2.3.6-3 and 2.3.6-4 show plots of  $D_e$  and  $D_a$  values of  $Ni^{2+}$  as a function of rock porosity for alkaline crystalline rock, respectively. Since the correlations were plotted only for data reported both porosity and  $D_e$  or  $D_a$ , only one plot has been reported for both  $D_e$  and  $D_a$ . Porosity dependency for diffusion can not be therefore discussed here.

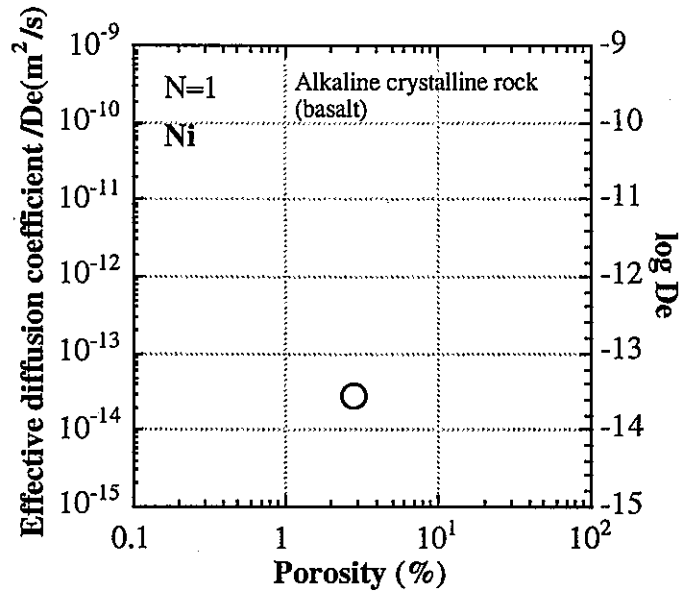
The  $D_o$  of  $Ni^{2+}$ , FF and Gf were estimated in the same way as section 2.1.6(Ni). Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.3.7.



**Figure 2.3.6-1** Histogram for De values of Ni<sup>2+</sup> ion in alkaline crystalline rock (basalt)  
The average value of De of Ni<sup>2+</sup> in alkaline crystalline rock was estimated to be  $2.8 \times 10^{-14} \text{ m}^2/\text{s}$ .

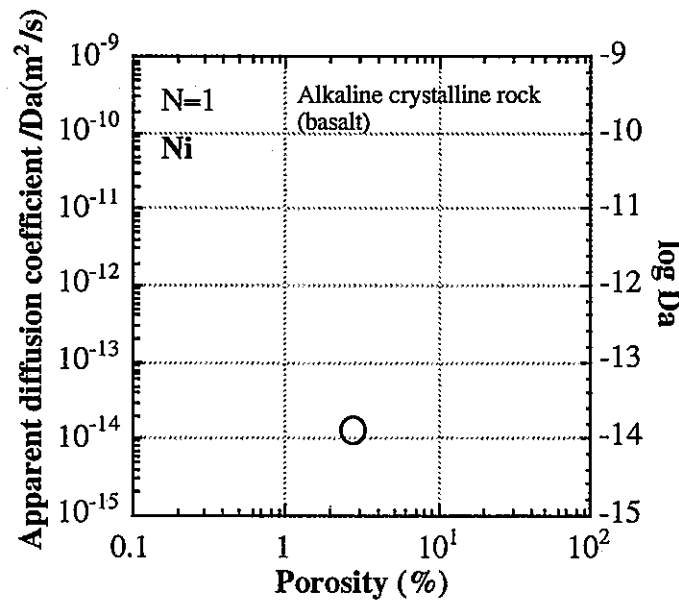


**Figure 2.3.6-2** Histogram for Da values of Ni<sup>2+</sup> ion in alkaline crystalline rock (basalt)  
The average value of Da of Ni<sup>2+</sup> in alkaline crystalline rock was estimated to be  $1.3 \times 10^{-14} \text{ m}^2/\text{s}$ .



**Figure 2.3.6-3**  $D_e$  values of  $Ni^{2+}$  ion as a function rock porosity for alkaline crystalline rock (basalt)

The correlation was plotted only for data reported both porosity and  $D_e$ . Only one plot has been reported for  $Ni^{2+}$ . Therefore, porosity dependency for  $D_e$  can not be evaluated here.



**Figure 2.3.6-4**  $D_a$  values of  $Ni^{2+}$  ion as a function of rock porosity for alkaline crystalline rock (basalt)

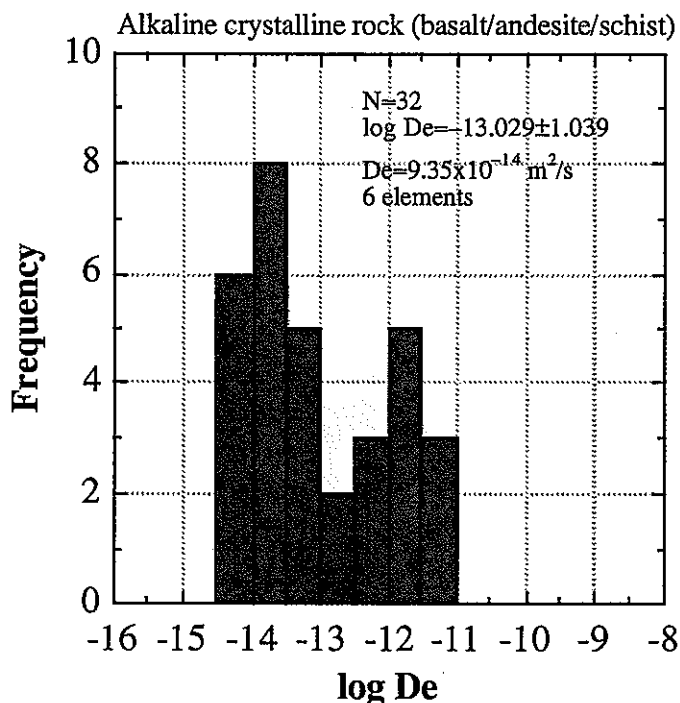
The correlation was plotted only for data reported both porosity and  $D_a$ . Only one plot has been reported for  $Ni^{2+}$ . Data enough to be able to discuss porosity dependency are not reported.

## References

- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, **26**, pp.119~133, Elsevier Science B. V.

### 2.3.7 Summary for diffusivity data in alkaline crystalline rock

Totally, 6 elements were discussed in sections 2.3.1~2.3.6. **Figure 2.3.7-1** shows a histogram of  $D_e$  values in alkaline crystalline rock for 6 elements<sup>2</sup> reported in each section. Although the average value of  $D_e$  for all elements in alkaline crystalline rock was estimated to be  $9.35 \times 10^{-14} \text{ m}^2/\text{s}$ , relatively a wide range of variation is found in  $D_e$ . The width of variation in  $D_e$  is over one order of magnitude. This is considered to attribute to the variation of porosity, but the variation in porosity is not so large, being in a range of 0.5 to 14.5 %. Besides the porosity also depends on the kind of rock. The porosity of basalt was between 2.08 and 2.8 %. That of andesite was between 3.0 and 14.5 % and that of schist was 0.5 %. As is clear, rock porosity is quite different depending on the kind of rock. Even though so, the width of variation in  $D_e$  is quite large, compared with that in sedimentary rock. This is considered to be due to porosity and difference in pore structure such as tortuosity. In these rocks,  $D_e$  values in basalt show a tendency lower than those in andesite and schist. This difference in tortuosity is considered to be caused by the differences of particle sizes and the shapes of minerals composing rock.



**Figure 2.3.7-1** Histogram for  $D_e$  values in alkaline crystalline rock for 6 elements (basalt, andesite, schist)

The average value of  $D_e$  for all elements in alkaline crystalline rock was estimated to be  $9.35 \times 10^{-14} \text{ m}^2/\text{s}$ . Since the differences in porosity and pore structure are not considered in the histogram, relatively a wide range of variation is found in  $D_e$ .

<sup>2</sup> 6 elements: Cl, Cs, I, K, Na and Ni.



Figure 2.3.7-2 shows a histogram of Da values in alkaline crystalline rock for 6 elements (no Da has been reported for K and Na) treated in sections 2.3.1-2.3.6. The average value of Da for all elements in alkaline crystalline rock was estimated to be  $9.51 \times 10^{-13} \text{ m}^2/\text{s}$  and a variation of two orders of magnitude was found in Da. The Da also includes sorption onto minerals composing rock, so the difference in Da can not be simply explained by porosity and pore structure. The Da is affected by both sorption and De. Since De depends on the pore structure of rock and sorption also depends on the kind of rock, it is presumed that the variation of Da was caused by the differences of constituent minerals and pore structure by the difference of the kind of rock.

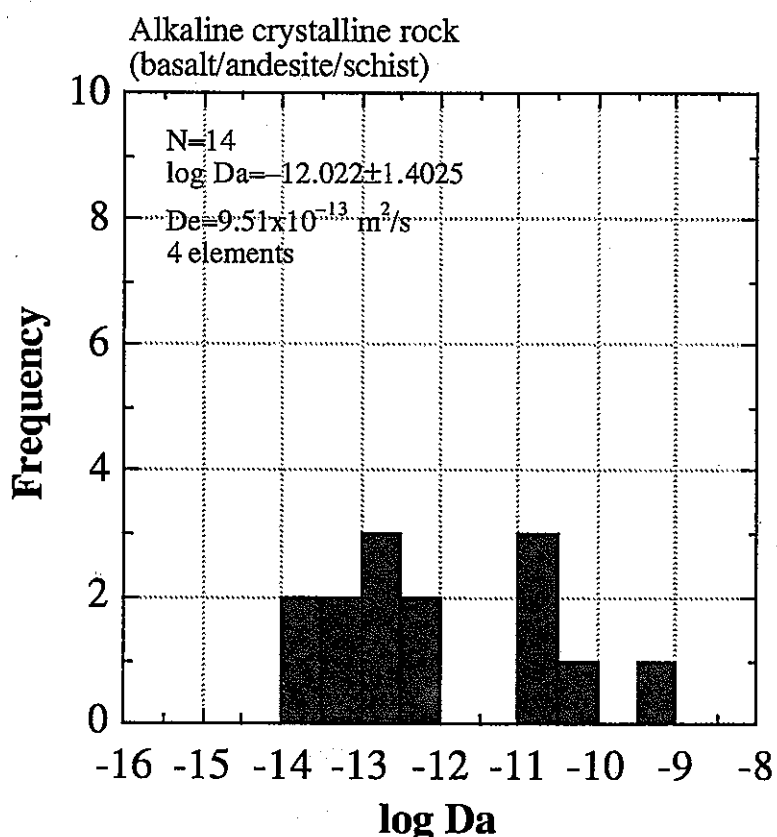


Figure 2.3.7-2 Histogram for Da values in alkaline crystalline rock (basalt, andesite, schist) The average value of Da in alkaline crystalline rock was estimated to be  $9.51 \times 10^{-13} \text{ m}^2/\text{s}$ . The width of variation in Da is quite large.

Figure 2.3.7-3 shows a porosity dependency of  $D_e$  values for 6 elements discussed in sections 2.3.1-2.3.6. Totally, 28 plots have been reported for  $D_e$  together with porosity. The  $D_e$  shows a tendency to increase with increasing rock porosity. However, the width of variation in  $D_e$  is quite larger than that of sedimentary rock. No significant tendency by the difference of the kind of element on  $D_e$  is also particularly found. It is considered that  $D_e$  is strongly affected by pore structure.

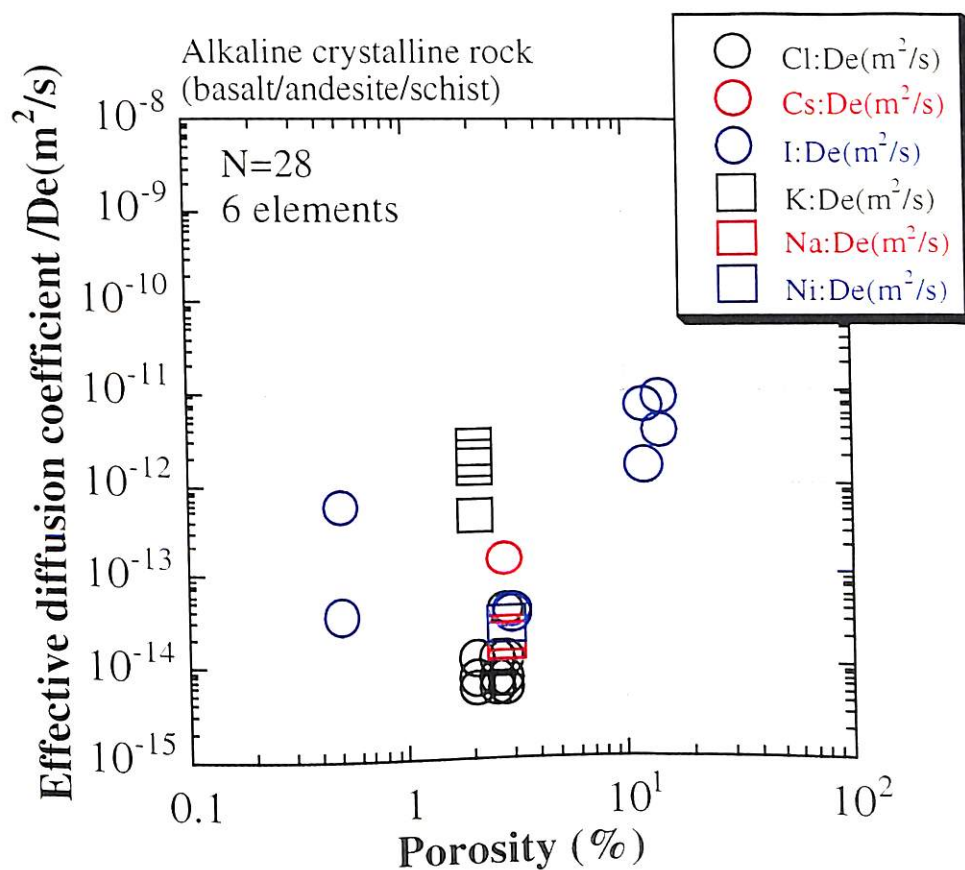


Figure 2.3.6-3  $D_e$  values for 6 elements as a function of rock porosity for alkaline crystalline rock (basalt, andesite, schist)

Figure 2.3.7-4 shows a least squares fitting curve for  $D_e$  values as a function of porosity. In the derivation of empirical equation,  $D_e$  data for all elements (6 elements) were taken into account without distinguishing the kind of element. From the fitting, the empirical equation:  $D_e = (10^{-13.56 \pm 0.321}) \cdot \phi^{1.24 \pm 0.58}$  was derived, where  $\phi$  is the porosity in % and the errors stand for the standard deviations.

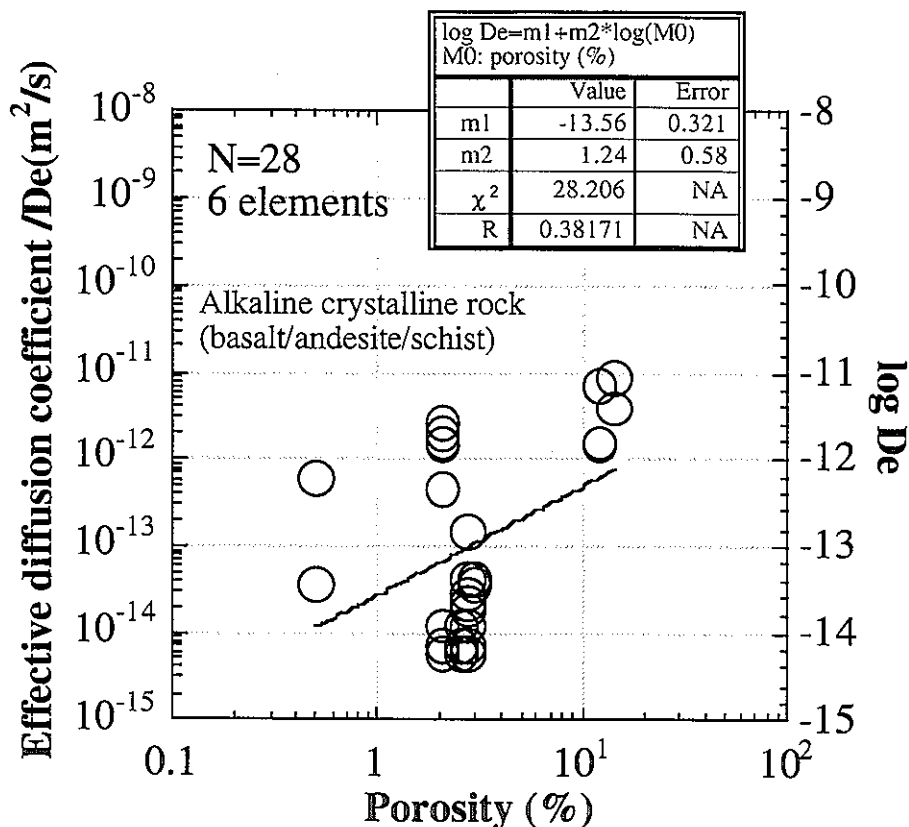


Figure 2.3.7-4 Empirical equation for  $D_e$  values as a function of rock porosity for alkaline crystalline rock (basalt, andesite, achist)

Figure 2.3.7-5 shows a width of variation in  $D_e$  values as a function of rock porosity. Two lines were drawn to be parallel to the empirical equation of  $D_e$  values as a function of rock porosity, which is shown in Figure 2.3.7-4 and to pass on the maximum and minimum values of plotted  $D_e$  values. The width of variation in  $D_e$  is approximately 3 orders of magnitude for the same porosity, being large. The upper and lower lines do not have statistical meaning, merely showing measured maximum and minimum values.

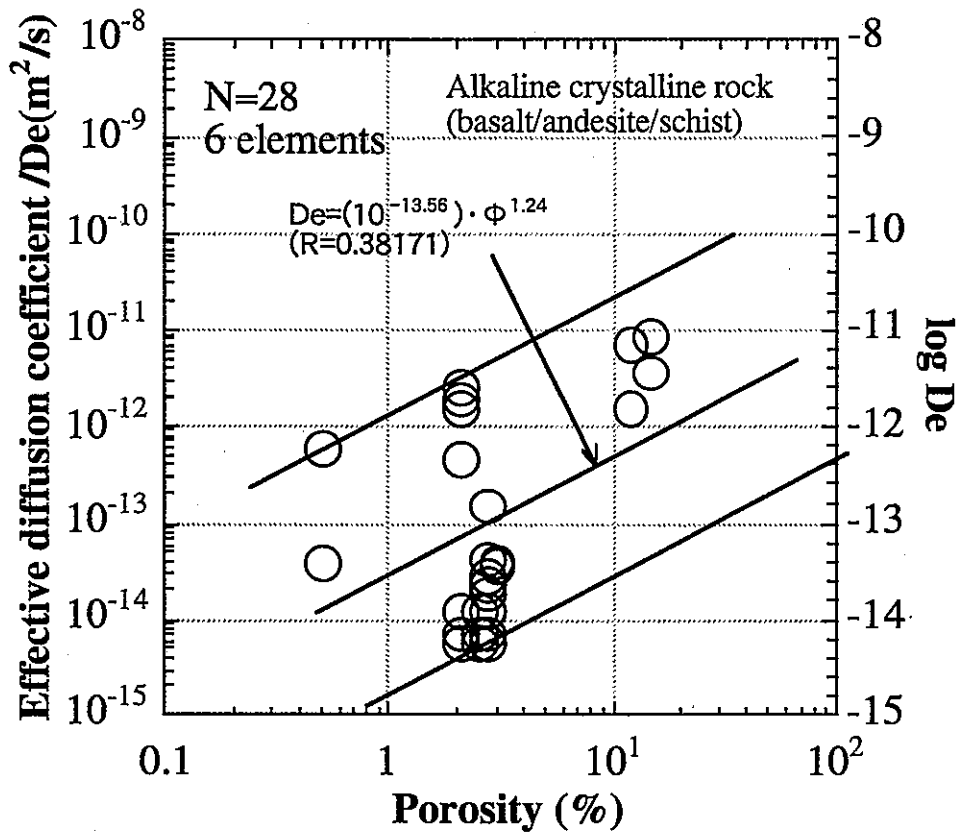


Figure 2.3.7-5 A width of variation in  $D_e$  values as a function of rock porosity for alkaline crystalline rock (basalt, andesite, schist)

Figure 2.3.7-6 shows a least squares fitting curve for  $D_a$  values as a function of rock porosity. Though  $D_a$  shows a tendency to increase with increasing rock porosity, clear trend is not found. The width of variation in  $D_a$  is quite large. This is considered to be due to the effects of rock porosity and pore structural factor by the difference of the kind of rock and the effect of sorption by the differences of the kinds of rock and element. For  $D_a$ , a width of variation of approximately 4 orders of magnitude is in maximum found for the same porosity.

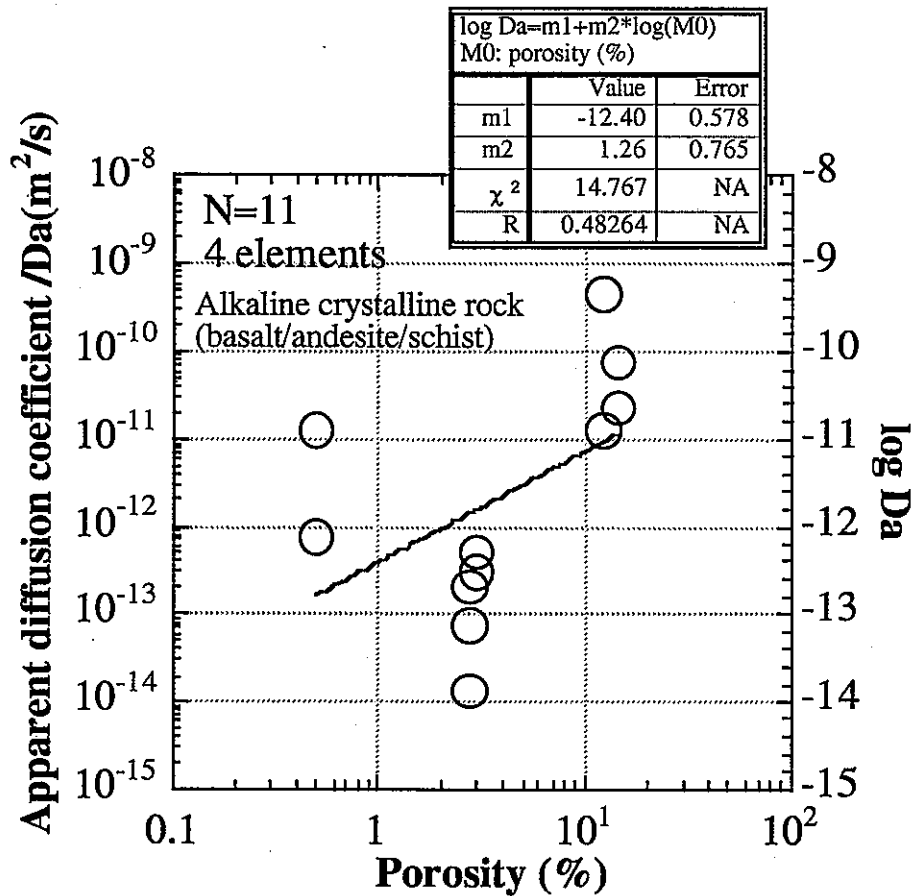


Figure 2.3.7-6  $D_a$  values as a function of rock porosity for alkaline crystalline rock (basalt, andesite, schist)

The width of variation in  $D_a$  is approximately 4 orders of magnitude in maximum for the same porosity, being very large.

Figure 2.3.7-7 shows a correlation of  $D_e$  values as a function of charge number of species. No systematic trend is particularly found. The width of variation in  $D_e$  is approximately 3 orders of magnitude, being very large, although the charge of species is the same. Since porosity and pore structural factors actually depend on the kind of rock and  $D_0$  is also different depending on  $D_0$  for each species, it is presumed that such a wide range of variation in  $D_e$  was caused by the differences of them.

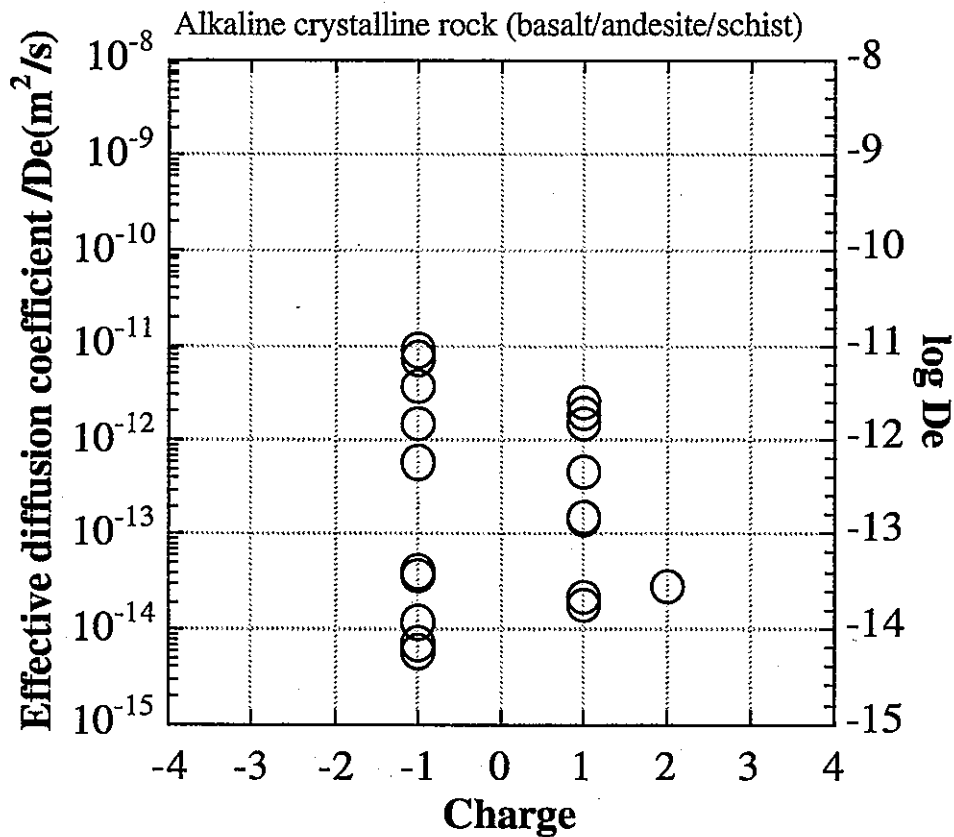
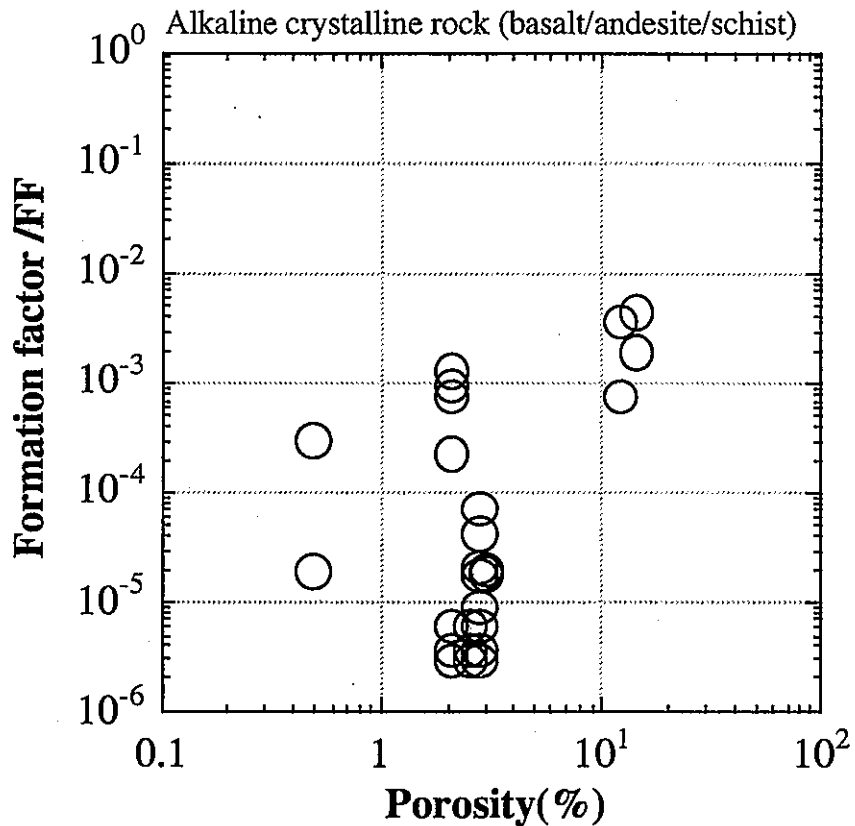


Figure 2.3.7-7  $D_e$  values as a function of charge number of species  
 The width of variation in  $D_e$  is approximately 3 orders of magnitude, being large, although the charge of species is the same. This is not only because of the variation of porosity, but also because of the difference in  $D_0$  for each species.

**Figure 2.3.7-8** shows a correlation of formation factors (FF) as a function of rock porosity. Similarly to De values, FF also shows a tendency to increase with increasing porosity. However, a wide range of variation is found in FF, although rock porosity is the same. This may be caused by the differences of tortuosity by the differences between grain sizes of individual rocks. In the porosity dependency for FF, FF values of basalt seem to be lower than those of the other rocks.



**Figure 2.3.7-8** FF values as a function of rock porosity (basalt, andesite, schist)  
 A wide range of variation in FF is found even though it is the same porosity. The FF values for basalt seem to be lower than those for the other rocks (andesite and schist).

Figure 2.3.7-9 shows a correlation of geometric factors ( $G_f$ ) as a function of rock porosity. As shown in Figure 2.3.7-9, no significant porosity dependency is found for  $G_f$ . Among rock samples used for diffusion experiments,  $G_f$  values for basalt showed a tendency lower than those for the other rocks (andesite and schist). This is considered to ascribe to grain sizes of minerals composing the rock. The grain sizes of minerals of basalt are smaller than those of the other rocks.

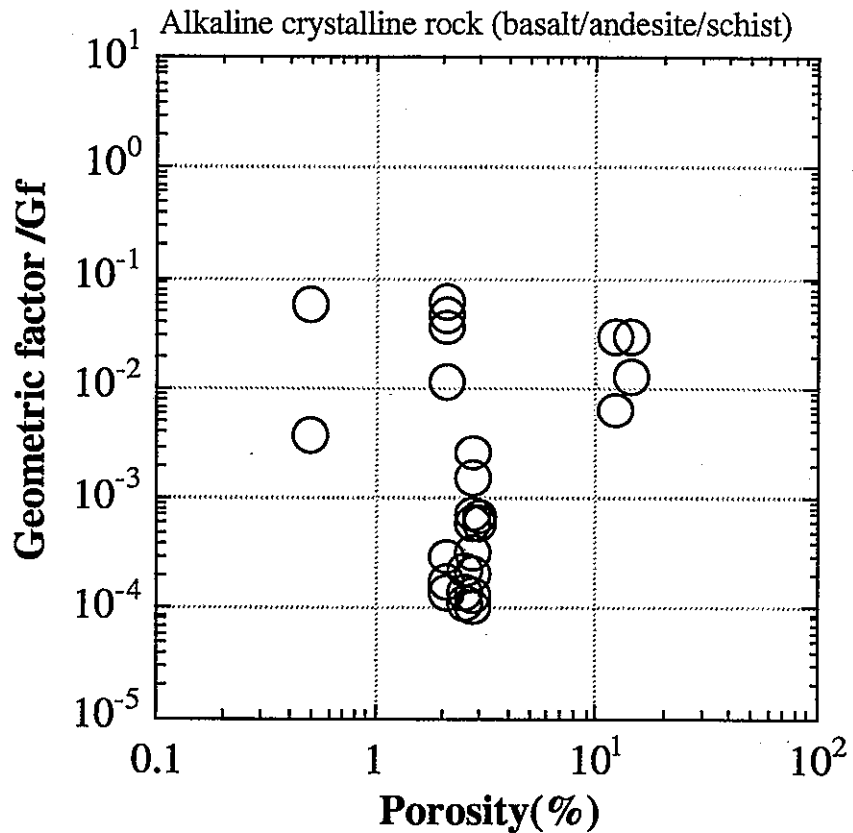


Figure 2.3.7-9  $G_f$  values as a function of rock porosity (basalt, andesite, schist)  
No porosity dependency is found for  $G_f$ . Among rock samples used for diffusion experiments,  $G_f$  values for basalt show a tendency lower than those for the other rocks (andesite and schist).



Figure 2.3.7-10 shows a plot of  $De$  values for individual elements in alkaline crystalline rock. No tendency between  $De$  values and elements is particularly found. Even the same element has a wide range of  $De$  values (for example, I). However, the variation in rock porosity is also included in the variation in  $De$ . Furthermore, the tortuosity of rock is also different depending on the kind of rock, because the grain sizes are different depending on the kind of rock. Among these elements, a wide range of variation is found for  $De$  of I. This may be due to the variation of porosity. As described in section 2.3.3,  $De$  values for I increased with increasing rock porosity.

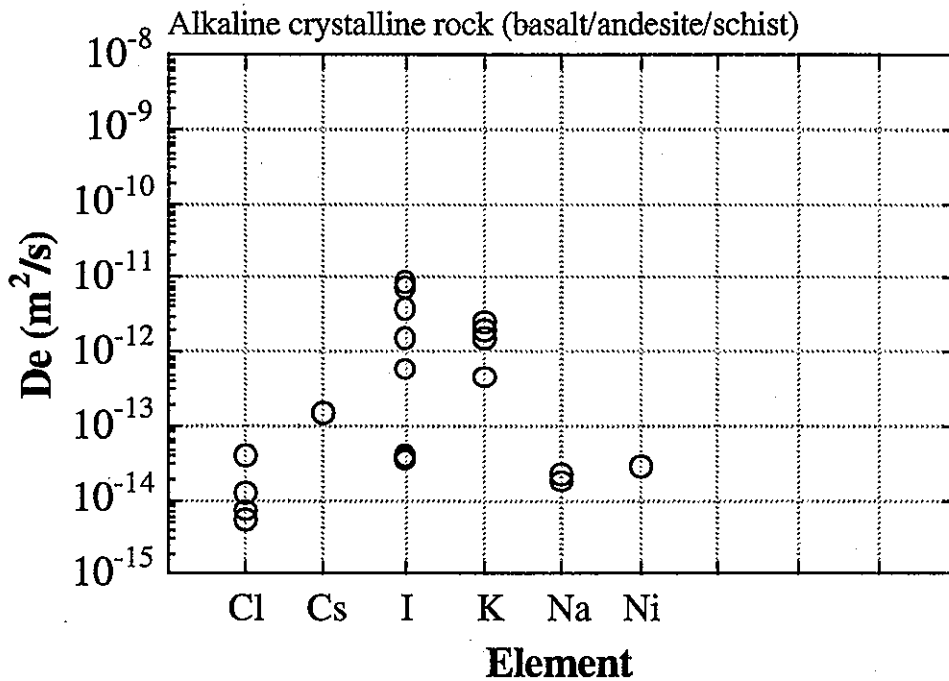


Figure 2.3.7-10 A plot of  $De$  values for individual elements in alkaline crystalline rock (basalt, andesite, schist)

No particular tendency between  $De$  values and elements is found, showing a wide range of  $De$  values even though the same element.

Figure 2.3.7-11 shows a correlation of  $De$  values to  $Do$  values for alkaline crystalline rock. No particular correlation is found between  $De$  and  $Do$ . A wide range of variation in  $De$  is found near  $Do=2 \times 10^{-9} \text{ m}^2/\text{s}$  (25 °C). This reason may be because the width of variation in rock porosity is large and pore structural factors are also different depending on the rock porosity.

The elements which take near  $Do=2 \times 10^{-9} \text{ m}^2/\text{s}$  (25 °C) are Cl, Cs and I. This wide variation in  $De$  may be due to the differences of rock porosity and of pore structural factors by the difference of the kind of rock.

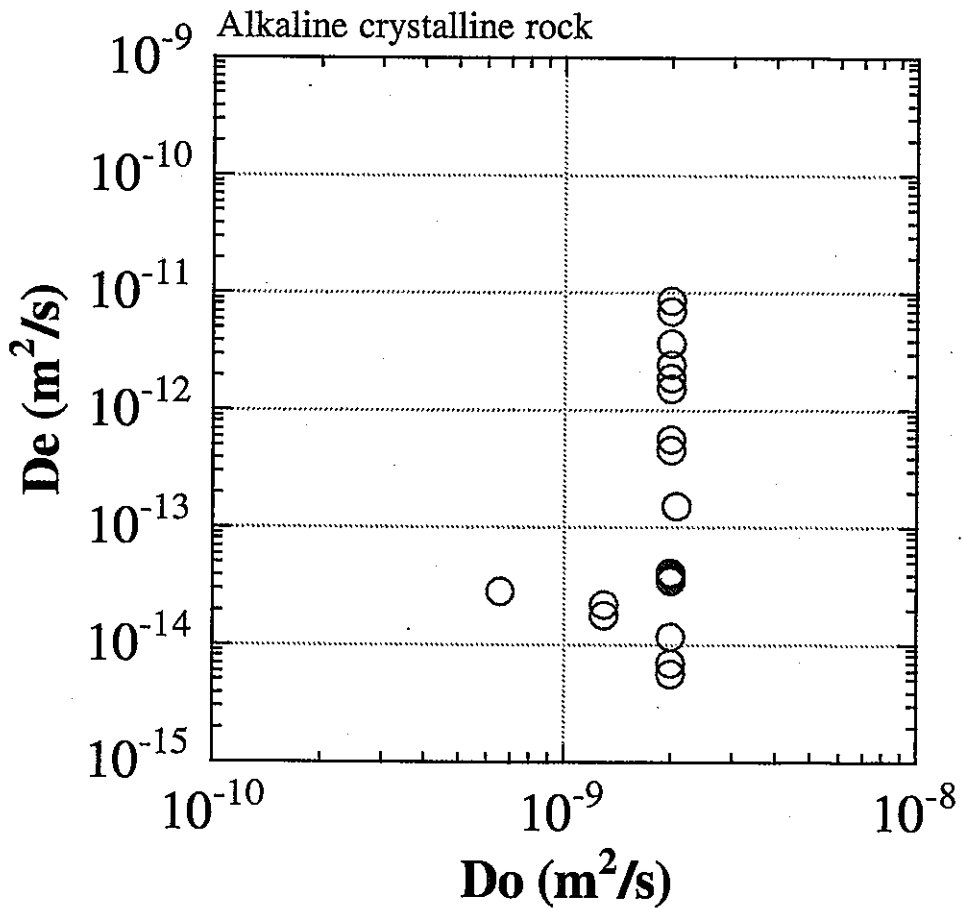


Figure 2.3.7-11 A correlation of  $De$  values to  $Do$  values for alkaline crystalline rock (basalt, andesite, schist)

No systematic tendency is found between  $De$  and  $Do$ .

## 2.4 Acid Crystalline Rock

Totally, 207 De and 159 Da data have been reported for acid crystalline rock to date based on a review of literatures. Eighteen elements and one tracer<sup>1</sup> have been totally discussed for acid crystalline rock. Almost all data were for granitic rocks. In this section, data regarding matrix diffusion obtained for individual elements for acid crystalline rock are introduced, as well as are discussed.

### 2.4.1 Barium (Ba)

The De values for Ba ( $Ba^{2+}$ ) in acid crystalline rock have been reported for granite sampled from Inada, Ibaraki, Japan [Yamaguchi et al., 1993]. Barium is not directly included in important elements for performance assessment of the geological disposal of HLW, but it is considered to chemically show similar behaviour to those of  $Ni^{2+}$ ,  $Sr^{2+}$  and  $Ra^{2+}$  in solution, being able to be treated as one of the elements which take a charge of +2. Since Ba is one of the alkaline earth metals, it is considered that elements which belong to the same group show similar behaviour in solution. Barium(II) is predominant in a wide pH range and is not sensitive to redox condition.

All measurements have been carried out under aerobic conditions, because it is not redox sensitive. The De data have been obtained by through-diffusion method for all cases. A solution of 0.1 M KCl was used as porewater for all measurements. All measurements have been performed at 25 °C. The reported porewater pH was 4 for all measurements. Besides all measurements have been carried out using a mixed solution of  $Ba(NO_3)_2$ ,  $BeSO_4$ ,  $CoCl_2$ ,  $MgCl_2$ ,  $NiCl_2$  and  $Sr(NO_3)_2$ , what is called a multi-tracer or a cocktail tracer.

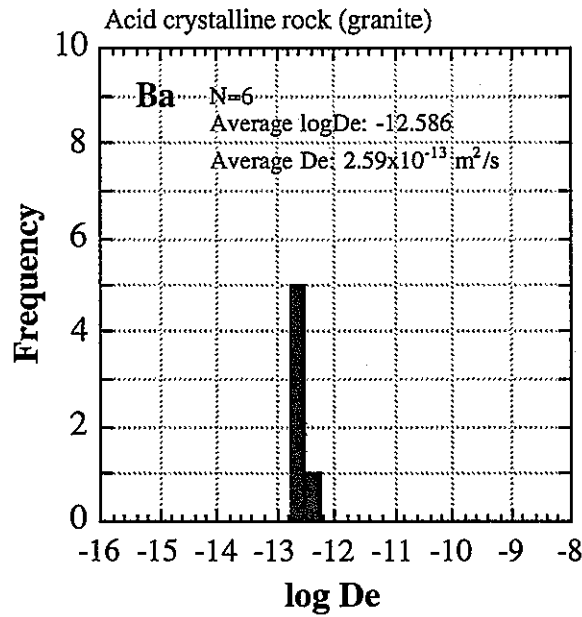
Figure 2.4.1-1 shows a histogram for De values of  $Ba^{2+}$  in acid crystalline rock reported to date. Totally, 6 De data have been reported for granite. Not so large variation in De is found. This is presumed to be due to the same porosity for granite. For Da, no datum has been reported. Since rock capacity factor has not been also reported, Da can not be estimated.

Figure 2.4.1-2 shows plots of De values of  $Ba^{2+}$  as a function of rock porosity. The correlations were plotted only for data reported both porosity and De. Totally, 6 plots have been reported for De. All plots are similar and the width of variation in De is quite small. Since all De data are plotted on the same porosity, the effect of porosity on De of  $Ba^{2+}$  can not be discussed here.

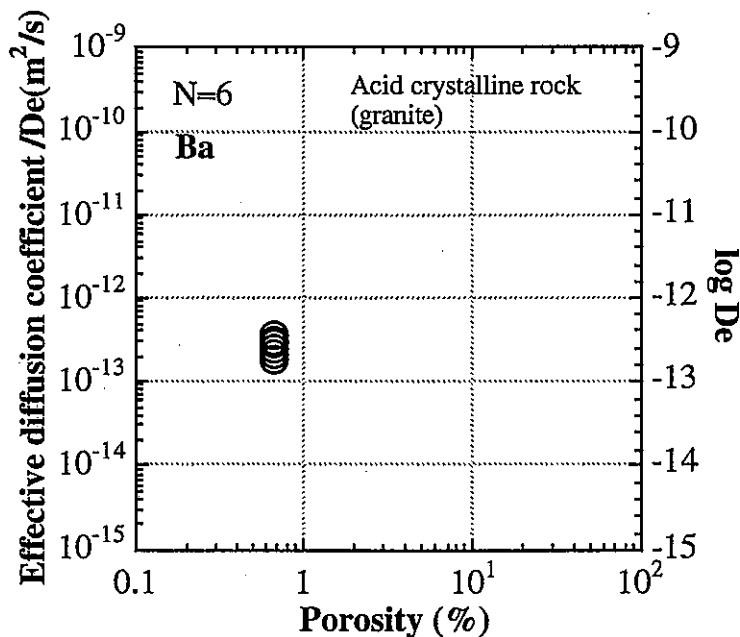
The Do of  $Ba^{2+}$  is estimated to be  $8.5 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C [Marcus, 1997] by the Nernst-Einstein equation [Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991]. The FF of granite was calculated based on the equation:  $FF=De/Do$  and Gf was also calculated based on the equation:

<sup>1</sup> 18 elements and one tracer: Ba, Be, Cl, Co, Cs, HTO, I, K, Mg, Na, Ni, Np, Pu, Se, Sm, Sr, Tc, U and Uranic.

$Gf = D_p/D_o = D_e/(\phi \cdot D_o) = FF/\phi$ . Where  $D_p$  is the porewater diffusion coefficient and  $\phi$  is the porosity. Porosity dependencies for  $FF$  and  $Gf$  will be discussed in more detail summarizing in section **2.4.20**.



**Figure 2.4.1-1** Histogram for  $De$  values of  $Ba^{2+}$  ion in acid crystalline rock (granite). The average value of  $De$  of  $Ba^{2+}$  in acid crystalline rock was estimated to be  $2.59 \times 10^{-13} \text{ m}^2/\text{s}$ . No variation in  $De$  is found.



**Figure 2.4.1-2**  $De$  values of  $Ba^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite)

The correlations were plotted only for data reported both porosity and  $De$ . All plots are similar and the width of variation in  $De$  is small. All  $De$  data are plotted on the same porosity and the effect of porosity on  $De$  can not be discussed here.

## References

- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.
- Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1993): Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796~803, the Atomic Energy Society of Japan.

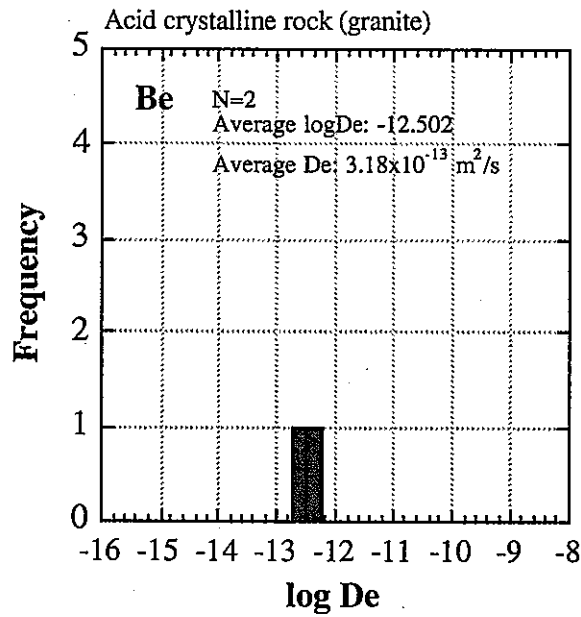
## 2.4.2 Beryllium (Be)

The  $D_e$  values for Be ( $\text{Be}^{2+}$ ) in acid crystalline rock have been reported for granite sampled from Inada, Ibaraki, Japan [Yamaguchi et al., 1993]. Beryllium is also not included in important elements for performance assessment of the geological disposal of HLW, but it is also presumed to be able to be treated in the same as  $\text{Ba}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Sr}^{2+}$  and  $\text{Ra}^{2+}$  in solution. Since Be is also one of the alkaline earth metals, it is presumed that elements which belong to the same group chemically show similar behaviour in solution. Beryllium takes Be(II) in solution [Brookins, 1988] and it is not redox sensitive. All measurements have been carried out in the same way as that of Ba (see 2.4.1(Ba)).

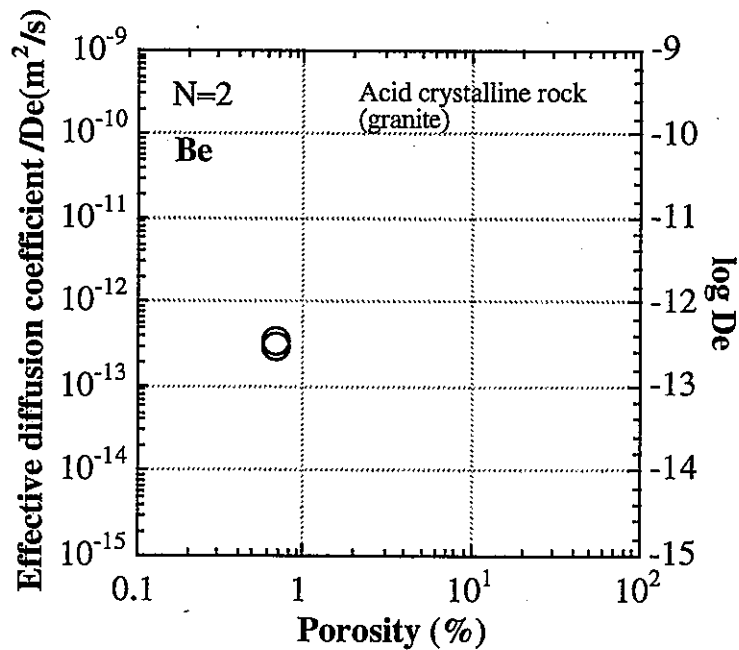
Figure 2.4.2-1 shows a histogram for  $D_e$  values of  $\text{Be}^{2+}$  in acid crystalline rock reported to date. Totally, 2  $D_e$  data have been reported for granite. Both  $D_e$  values are mutually similar. No  $D_a$  datum has been reported here. Since rock capacity factor has not been reported,  $D_a$  can not be estimated.

Figure 2.4.2-2 shows a plot of  $D_e$  values of  $\text{Be}^{2+}$  as a function of rock porosity. The correlations were plotted only for data reported both porosity and  $D_e$ . Totally, 2 plots have been reported for  $D_e$ . Here, since all reported porosities are the same, porosity dependency for  $D_e$  can not be discussed.

The  $D_0$  of  $\text{Be}^{2+}$  is estimated to be  $6.0 \times 10^{-10} \text{ m}^2/\text{s}$  at  $25 \text{ }^\circ\text{C}$  [Marcus, 1997] by the same theory as Ba as described in section 2.4.1(Ba). The FF and Gf were also calculated by the same theory as those of Ba. Detailed discussion for correlation between porosity and  $D_e$  will be made summarizing in section 2.4.20.



**Figure 2.4.2-1** Histogram for  $De$  values of  $Be^{2+}$  ion in acid crystalline rock (granite)  
The average value of  $De$  of  $Be^{2+}$  in granite was estimated to be  $3.18 \times 10^{-13} \text{ m}^2/\text{s}$ .



**Figure 2.4.2-2**  $De$  values of  $Be^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite)

The correlations were plotted only for data reported both porosity and  $De$ . Since all  $De$  data have been obtained for the same porosity for granite, the effect of porosity on  $De$  can not be discussed here.



## References

Brookins, D. G. (1988): pH-Eh Diagrams for Geochemistry, Springer-Verlag, Berlin, Germany, pp.160~161.

Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.

Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1993): Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, **30** (8), pp.796~803, the Atomic Energy Society of Japan.

### 2.4.3 Chlorine (Cl)

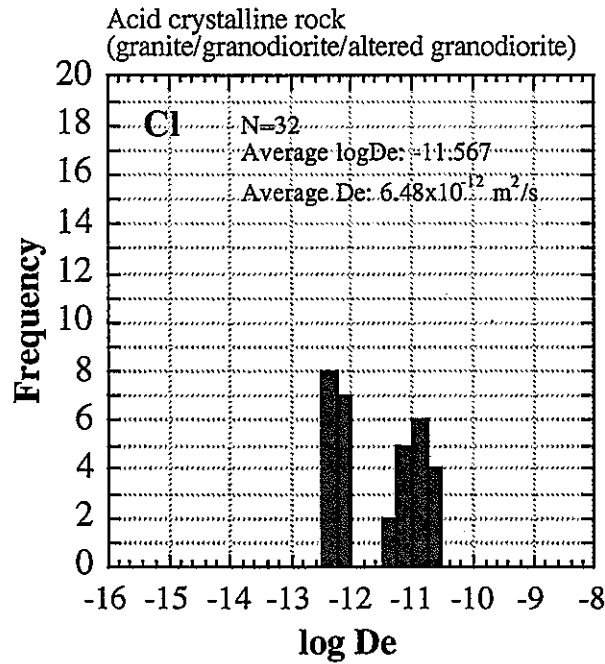
The  $D_e$  values for Cl ( $Cl^-$ ) in acid crystalline rock have been reported for granodiorite [Sato et al., 1992, 1993, 1997a, 1997b, 1997c; Sato and Shibutani, 1996; PNC, 1996], granite [Sato and Yui, 1992], altered granodiorite [Sato et al., 1997b, 1997c; Sato and Shibutani, 1996; PNC, 1996] and fracture fillings [Sato et al., 1997b, 1997c; Sato and Shibutani, 1996; PNC, 1996]. Though the fracture fillings are not included in acid crystalline rock, since a series of experiments have been carried out as a link in the chain of nuclide migration study in fractured crystalline rocks being composed of granodiorite as a host rock, these minerals were also regarded as one of the acid crystalline rocks here. The difference between granodiorite and altered granodiorite is the content of constituent chlorite. The content of constituent chlorite in the altered granodiorite is a little higher than that in the unaltered granodiorite. The content of biotite in the unaltered is a little higher instead of chlorite. But, the difference is quite small. The chemical behaviour and treatment of Cl in performance assessment of the geological disposal of HLW are as described in section 2.1.1(Cl).

Totally, 32  $D_e$  values, obtained by through-diffusion method, have been reported for granite, granodiorite and altered granodiorite up to the present. The  $D_a$  data have been also estimated, based on the time-lag method [e.g. Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor. All measurements have been carried out under aerobic conditions. In situ groundwater or distilled water was used as porewater in the experiments. The measurements have been performed at 25 °C or room temperature. The reported porewater pH was in a range of 5.5 to 8.92.

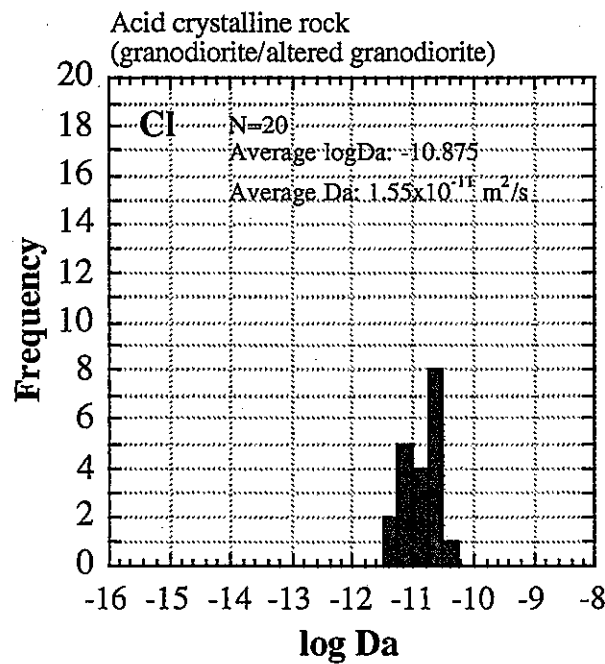
Figures 2.4.3-1 and 2.4.3-2 show histograms for  $D_e$  and  $D_a$  values of  $Cl^-$  in acid crystalline rock reported to date, respectively. Totally, 32  $D_e$  data have been reported. Some variation is found in  $D_e$ . The width of variation in  $D_e$  is nearly equal to two orders of magnitude. This is presumed to be because of the variation of rock porosity which range is between 0.88 and 5.6 %. Whilst, for  $D_a$ , 20 data have been reported in total for granodiorite and altered granodiorite. The width of variation in  $D_a$  was approximately one order of magnitude, being not so large.

Figures 2.4.3-3 and 2.4.3-4 show plots of  $D_e$  and  $D_a$  values of  $Cl^-$  as a function of rock porosity, respectively. For  $D_e$ , totally, 31 data have been reported together with rock porosity. The  $D_e$  clearly shows a tendency to increase with increasing rock porosity. Whilst, for  $D_a$ , 19 data have been reported for granodiorite and altered granodiorite. Significant porosity dependency for  $D_a$  is not found. Almost all  $D_a$  values are around  $10^{-11}$  m<sup>2</sup>/s in a porosity range of 1 to 5.6 %.

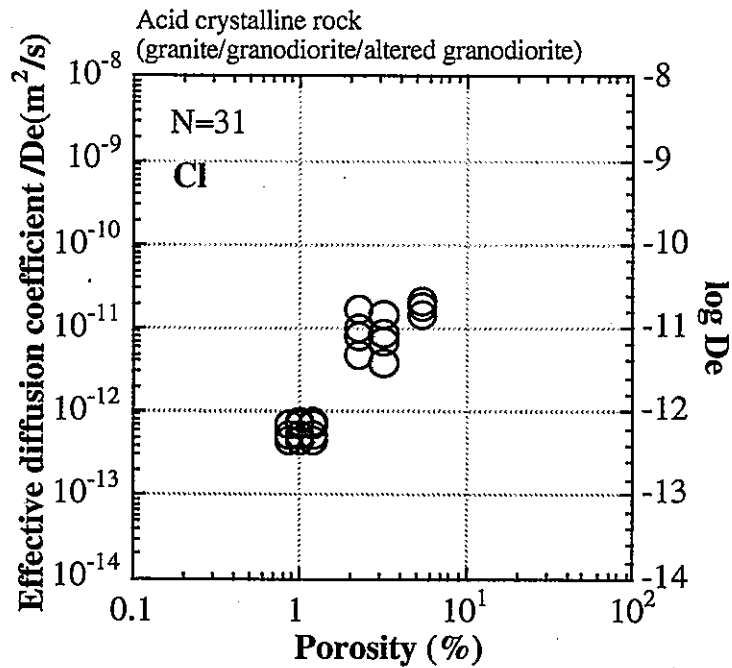
The  $D_o$  of  $Cl^-$ , FF and Gf were estimated in the same way as section 2.1.1(Cl). For FF and Gf will be discussed in more detail in section 2.4.20.



**Figure 2.4.3-1** Histogram for De values of Cl<sup>-</sup> ion in acid crystalline rock (granite, granodiorite, altered granodiorite)  
The width of variation in De is nearly equal to two orders of magnitudes. This may be due to the variation of porosity which range is between 0.88 and 5.6 %.

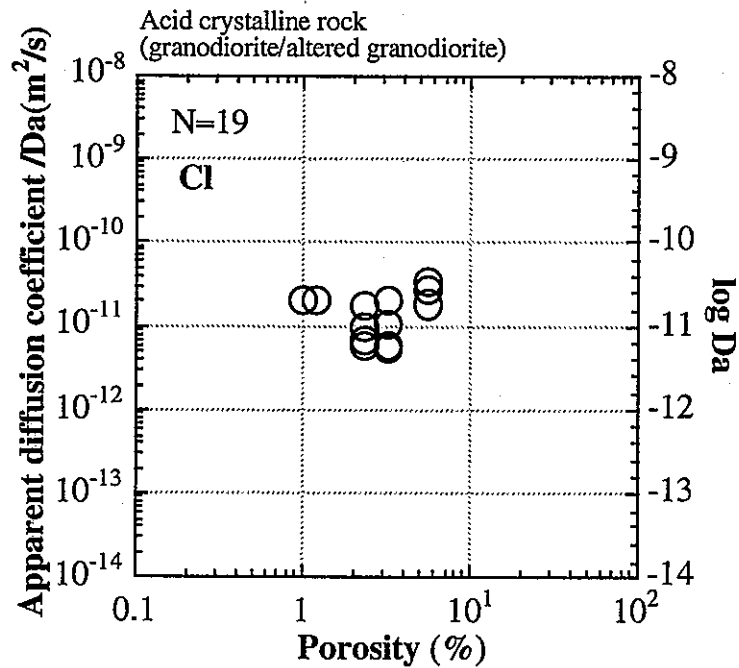


**Figure 2.4.3-2** Histogram for Da values of Cl<sup>-</sup> ion in acid crystalline rock (granodiorite, altered granodiorite)  
Not so large variation is found in Da.



**Figure 2.4.3-3** De values of Cl<sup>-</sup> ion as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite)

The De of Cl<sup>-</sup> clearly shows a tendency to increase with increasing rock porosity.



**Figure 2.4.3-4** Da values of Cl<sup>-</sup> ion as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)

Significant porosity dependency for Da is not found. Almost all Da values are around 10<sup>-11</sup> m<sup>2</sup>/s in a porosity range of 1 to 5.6 %.

## References

- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~207 [In Japanese].
- Sato, H., Yui, M. and Ishikawa, H. (1992): A Study on Pore Structure of Rocks, PNC Technical Report, PNC TN8410 92-222 [In Japanese].
- Sato, H. and Yui, M. (1992): Measurement of Ionic Diffusion Coefficients in Rocks, 1992 Fall Meeting of the Atomic Energy Society of Japan, F15, p.323 [In Japanese].
- Sato, H., Shibutani, T., Yoshikawa, H. and Yui, M. (1993): A Study on Diffusion Mechanism for Cs, Ni and Sm in Granodiorite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M29, p.667 [In Japanese].
- Sato, H. and Shibutani, T. (1996): Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste, PNC Technical Report, PNC TN1100 96-010, pp.139~144 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997a): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, **26**, pp.119~133, Elsevier Science.
- Sato, H., Shibutani, T., Tachi, Y., Ota, K., Amano, K. and Yui, M. (1997b): Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127.
- Sato, H., Tachi, Y., Shibutani, T. and Yui, M. (1997c): Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan, L12, p.561 [In Japanese].

## 2.4.4 Cobalt (Co)

The De values for Co ( $\text{Co}^{2+}$ ) in acid crystalline rock have been reported for granite sampled from Inada, Ibaraki, Japan [Idemitsu et al., 1992; Yamaguchi et al., 1993]. Cobalt is not directly included in important elements for performance assessment of the geological disposal of HLW, but it is considered to be able to be treated in the same as  $\text{Ba}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Sr}^{2+}$ ,  $\text{Ra}^{2+}$  and  $\text{Be}^{2+}$ , because they chemically show similar behaviour if they have the same chemical form in solution. Cobalt can chemically take Co(II) and (III) in solution [Brookins, 1988], but Co(II) stably exists in wide pH and Eh conditions, and it is not sensitive to redox condition. The predominant species in solution is  $\text{Co}^{2+}$  < around pH 8,  $\text{CoCO}_3$  between 8 and 11,  $\text{Co(OH)}_2$  between 11 and 12, and  $\text{HCoO}_2^-$  > 12 in the system Co-S-C-O-H. Under relatively high oxidizing conditions,  $\text{Co}_3\text{O}_4$  is predominant in a pH range < around 6. Besides CoS is predominant in a pH range > around 5 under strongly reducing conditions.

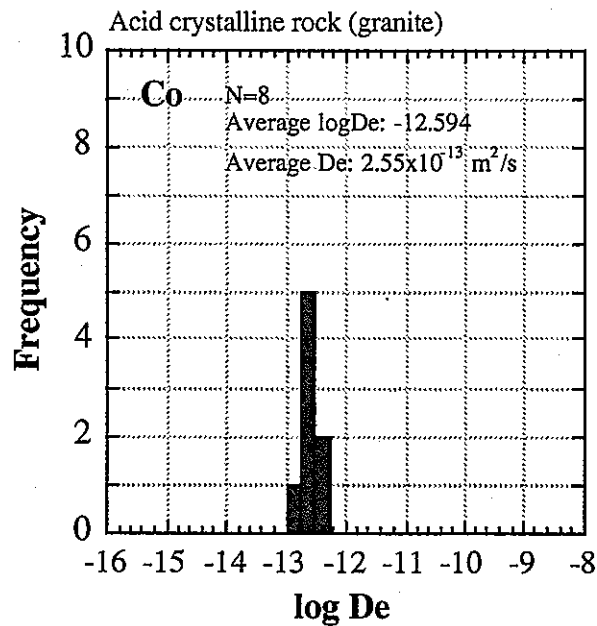
Cobalt-60 is regarded as an important nuclide in low-level radioactive waste (LLW)(half-life: 5.271 y  $\beta^-$  decay property: 0.31 MeV,  $\gamma$  decay property: 1.17 and 1.33 MeV). All measurements have been carried out under aerobic conditions, because it is not redox sensitive.

The De data have been obtained by through-diffusion method. Whilst, Da values have been obtained by in-diffusion method. A solution of 0.1 M KCl was used as porewater in the through-diffusion experiments to keep the ionic strength of porewater. On the other hand, deionized water was used as porewater in in-diffusion experiments. All measurements for De have been performed at 25 °C and those for Da have been performed at 30 °C. The reported porewater pH was 4 in the through-diffusion experiments for De, but it ranged from 4.3 to 9.4 in the in-diffusion experiments for Da. Besides all through-diffusion experiments for De have been carried out using a mixed solution of  $\text{Ba(NO}_3)_2$ ,  $\text{BeSO}_4$ ,  $\text{CoCl}_2$ ,  $\text{MgCl}_2$ ,  $\text{NiCl}_2$  and  $\text{Sr(NO}_3)_2$ , what is called a multi-tracer or a cocktail tracer. All in-diffusion experiments for Da have been carried out using a mixture solution of Cs-134, Sr-85 and Co-60.

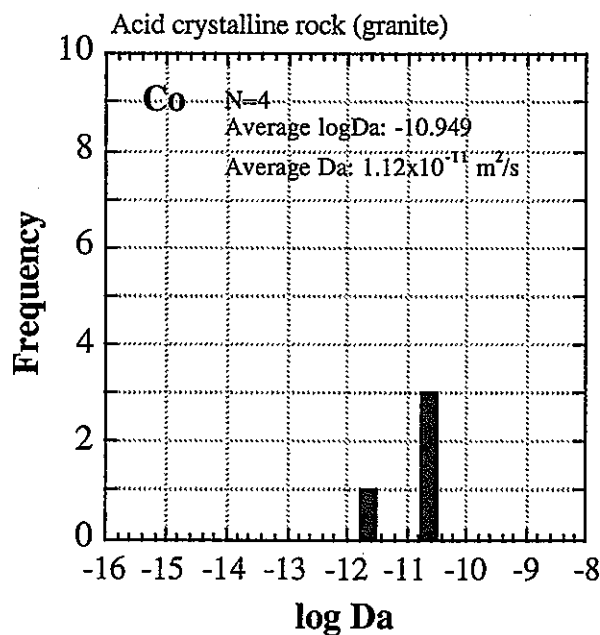
Figures 2.4.4-1 and 2.4.4-2 show histograms for De and Da values of  $\text{Co}^{2+}$  in acid crystalline rock reported to date, respectively. Totally, 8 De data have been reported for granite. The width of variation in De is narrow. This is presumed to be due to the same porosity for granite. For Da, totally, 4 data have been reported. The width of variation in Da is approximately one order of magnitude even though rock porosity is the same. This is presumed to be due to various pH of the porewater.

Figures 2.4.4-3 and 2.4.4-4 show plots of De and Da values of  $\text{Co}^{2+}$  as a function of rock porosity reported to date, respectively. Totally, 8 plots have been reported for De. Since all De data are plotted on the same porosity, porosity dependency for De can not be discussed here. For Da, totally, 4 plots have been reported. As described above, though rock porosity is the same and De values are approximately the same, a variation of approximately one order of magnitude is found in Da. Since the pore structural factors of granite were almost the same, it is presumed that Kd of  $\text{Co}^{2+}$  on granite changed depending on the pH of the porewater. Since all Da data are also plotted on the same porosity, the effect of porosity on Da can not be discussed.

The  $D_o$  of  $Co^{2+}$  is estimated to be  $7.3 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C [Marcus, 1997] by the Nernst-Einstein equation [Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991]. The FF and Gf of granite were calculated in the same way as Ba, Be, Cl, etc. Porosity dependencies for FF and Gf will be discussed in more detail summarizing in section 2.4.20.

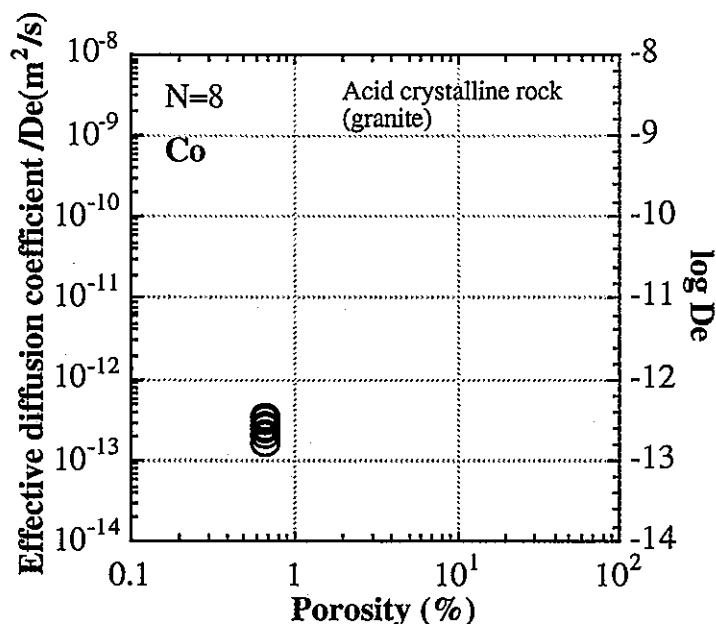


**Figure 2.4.4-1** Histogram for De values of  $\text{Co}^{2+}$  ion in acid crystalline rock (granite)  
The average value of De of  $\text{Co}^{2+}$  in granite was estimated to be  $2.55 \times 10^{-13} \text{ m}^2/\text{s}$ . The width of variation in De is small. This is considered to be due to the same porosity.

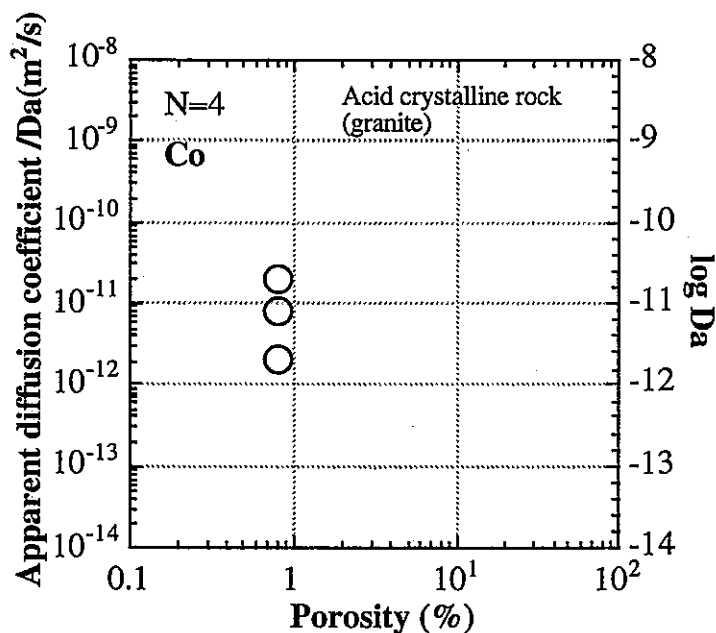


**Figure 2.4.4-2** Histogram for Da values of  $\text{Co}^{2+}$  ion in acid crystalline rock (granite)  
The average value of Da of  $\text{Co}^{2+}$  in acid crystalline rock was estimated to be  $1.12 \times 10^{-11} \text{ m}^2/\text{s}$ . A variation of approximately one order of magnitude is found in Da even though rock porosity is the same. This is presumed to be due to various pH of the porewater.





**Figure 2.4.4-3**  $D_e$  values of  $\text{Co}^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite)  
 All  $D_e$  values are approximately the same.



**Figure 2.4.4-4**  $D_a$  values of  $\text{Co}^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite)  
 A variation of approximately one order of magnitude is found in  $D_a$  even though rock porosity is the same. However, the pH of the porewater was various in diffusion experiments. This various pH of the porewater may have caused this variation in  $D_a$ .

## References

- Brookins, D. G. (1988): pH-Eh Diagrams for Geochemistry, Springer-Verlag, Berlin, Germany, pp.71~72.
- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.
- Idemitsu, K., Furuya, H., Hara, T. and Inagaki, Y. (1992): Migration of Cesium, Strontium and Cobalt in Water-Saturation Inada Granite, Journal of Nuclear Science and Technology, Vol.29, No.5, pp.454~460, the Atomic Energy Society of Japan.
- Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.
- Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1993): Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796~803, the Atomic Energy Society of Japan.

## 2.4.5 Cesium (Cs)

The  $D_e$  values for Cs ( $\text{Cs}^+$ ) in acid crystalline rock have been reported for granodiorite [Sato et al., 1993, 1997a, 1997b, 1997c; Sato and Shibutani, 1994, 1996; PNC, 1994, 1996], altered granodiorite [Sato et al., 1997b, 1997c; Sato and Shibutani, 1996; PNC, 1996] and fracture fillings [Sato et al., 1997b, 1997c; Sato and Shibutani, 1996; PNC, 1996]. The reason which the fracture fillings are included in acid crystalline rock is as described in section 2.4.3(C1). Since Cs is not redox sensitive, all measurements have been carried out under aerobic conditions.

For Cs,  $^{135}\text{Cs}$  is regarded as one of the important elements for performance assessment of the geological disposal of HLW. The chemical behaviour, decay property and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.2(Cs).

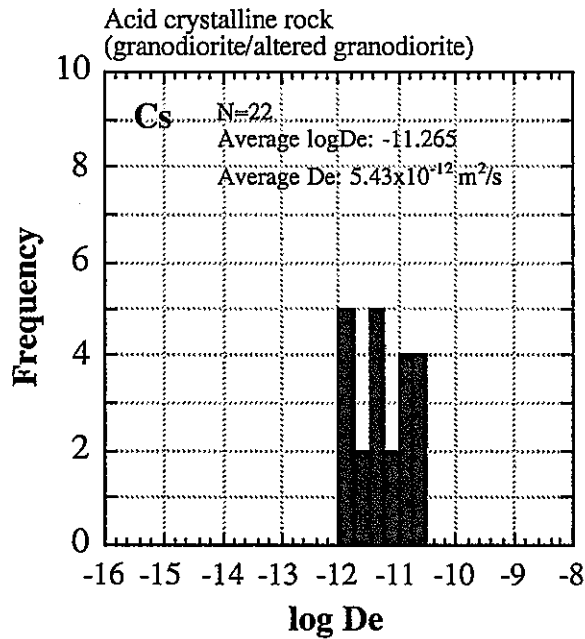
All  $D_e$  data have been obtained by through-diffusion method. The  $D_a$  data have been obtained from the time-lag method [e.g. Muurinen et al., 1987] in the through-diffusion experiments, in-diffusion experiments and an advection diffusion experiment. In the measurements by the time-lag method,  $D_a$  values for granodiorite [Sato et al., 1997a, 1997b, 1997c; Sato and Shibutani, 1994, 1996; PNC, 1994, 1996], altered granodiorite [Sato et al., 1993, 1997b, 1997c; Sato and Shibutani, 1996; PNC, 1996] and fracture fillings [Sato et al., 1997b, 1997c; Sato and Shibutani, 1996; PNC, 1996] have been reported. In the advection diffusion experiment,  $D_a$  for granite [Mukai et al., 1998] has been reported. In the measurements by in-diffusion method,  $D_a$  values for biotitic granite [Tsukamoto and Ohe, 1988] and granite [Idemitsu et al., 1992] have been reported. In situ groundwater, distilled water and deionized water were used as porewater in the experiments. All through-diffusion experiments have been performed at 25 °C (only one experiment has been done at 20 °C) and advection diffusion experiment has been done at room temperature. In-diffusion experiments have been carried out in a temperature range of 30 to 70 °C. The pH of the porewater ranged from 4.2 to 9.4.

Figures 2.4.5-1 and 2.4.5-2 show histograms for  $D_e$  and  $D_a$  values of  $\text{Cs}^+$  in acid crystalline rock reported to date, respectively. Totally, 22  $D_e$  data have been reported for granodiorite and altered granodiorite (including fracture fillings). The width of variation in  $D_e$  is approximately one order of magnitude, being not so large. Whilst, for  $D_a$ , totally, 30 data have been reported for granite, granodiorite, altered granodiorite and biotitic granite. The variation in  $D_a$  is also narrow except only one datum. Only one datum was extremely low. This low  $D_a$  datum was obtained from advection diffusion experiment which is one of the dynamic diffusion methods.

Figures 2.4.5-3 and 2.4.5-4 show plots of  $D_e$  and  $D_a$  values of  $\text{Cs}^+$  as a function of rock porosity for acid crystalline rock, respectively. The correlations were plotted only for data reported both porosity and  $D_e$  or  $D_a$ . For  $D_e$ , 21 plots have been reported in total. The  $D_e$  clearly shows a tendency to increase with increasing rock porosity. Whilst, 28  $D_a$  plots have been totally reported. The  $D_a$ , different from  $D_e$ , does not change as a function of rock porosity. The  $D_a$  approximately shows  $10^{-11}$  m<sup>2</sup>/s in a porosity range of 0.374 to 5.6 %. The sorptivity of Cs is generally said to be depended on the ionic strength of porewater and cation exchange capacity (CEC), but the ionic strength of porewater in this case is not various.

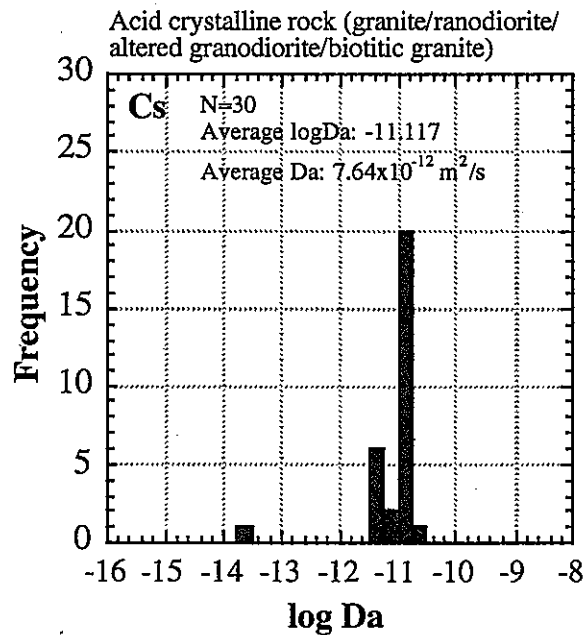
However, temperature and the pH of the porewater were various. Besides CEC is in the order of fracture fillings > altered granodiorite = granodiorite [Sato et al., 1997b]. Therefore, a variation of these parameters may have caused the change in sorption.

The  $D_0$  of  $Cs^+$ , FF and Gf were estimated in the same way as section 2.1.2(Cs). Porosity dependencies for FF and Gf will be discussed in detail in section 2.4.20.



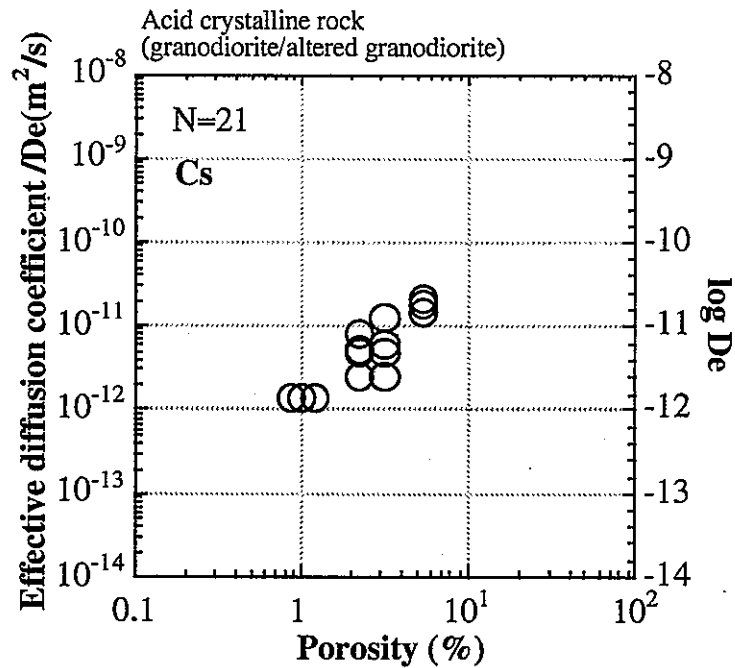
**Figure 2.4.5-1** Histogram for  $De$  values of  $Cs^+$  ion in acid crystalline rock (granodiorite, altered granodiorite)

The width of variation in  $De$  is approximately one order of magnitude, being not so large.



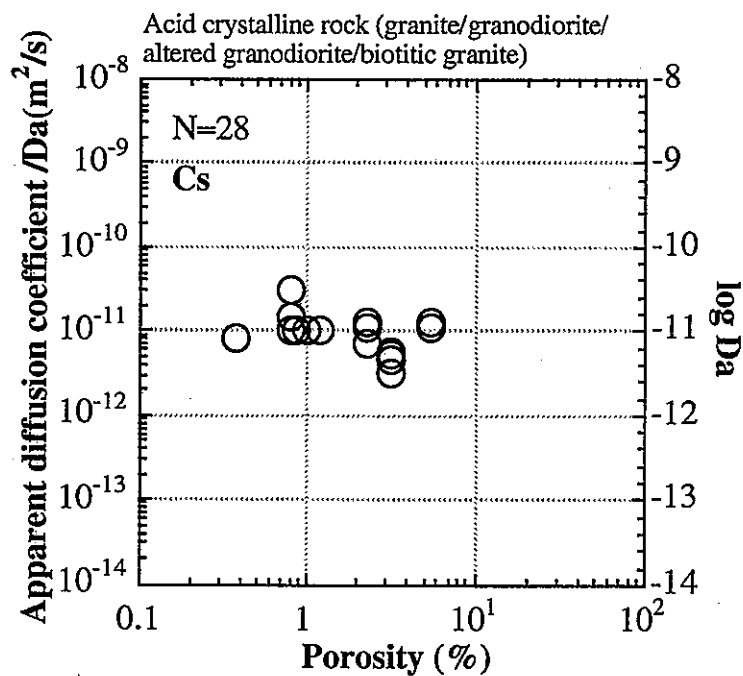
**Figure 2.4.5-2** Histogram for  $Da$  values of  $Cs^+$  ion in acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite)

The variation in  $Da$  is also narrow except only one extremely low datum. The datum was obtained from advection diffusion experiment which is one of the dynamic diffusion methods.



**Figure 2.4.5-3**  $D_e$  values of  $Cs^+$  ion as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)

The  $D_e$  clearly shows a tendency to increase with increasing rock porosity.



**Figure 2.4.5-4**  $D_a$  values of  $Cs^+$  ion as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite)

The  $D_a$  does not change as a function of rock porosity, showing approximately  $10^{-11} m^2/s$  in a porosity range of 0.374 to 5.6 %.

## References

- Idemitsu, K., Furuya, H., Hara, T. and Inagaki, Y. (1992): Migration of Cesium, Strontium and Cobalt in Water-Saturation Inada Granite, *Journal of Nuclear Science and Technology*, Vol.29, No.5, pp.454~460, the Atomic Energy Society of Japan.
- Mukai, S., Kitao, H. and Kataoka, S. (1998): Advection Diffusion Experiment Using Se and Cs in Fractured Rocks, 1998 Fall Meeting of the Atomic Energy Society of Japan, M25, p.842 (1998) [In Japanese].
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, *Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X* (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~207 [In Japanese].
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1994): Current Status for Research and Development of Geological Disposal -FY1993-, PNC Technical Report, PNC TN1410 94-094, pp.131~133 [In Japanese].
- Sato, H., Shibutani, T., Yoshikawa, H. and Yui, M. (1993): A Study on Diffusion Mechanism for Cs, Ni and Sm in Granodiorite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M29, p.667 [In Japanese].
- Sato, H. and Shibutani, T. (1994): Study on Adsorption and Diffusion of Nuclides in Buffer Material and Geosphere, PNC Technical Review, No.91, PNC Technical Report, PNC TN8410 94-284, pp.71~89, Power Reactor and Nuclear Fuel Development Corporation (PNC) [In Japanese].
- Sato, H. and Shibutani, T. (1996): Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste, PNC Technical Report, PNC TN1100 96-010, pp.139~144 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997a): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, *Journal of Contaminant Hydrology*, 26, pp.119~133, Elsevier Science.
- Sato, H., Shibutani, T., Tachi, Y., Ota, K., Amano, K. and Yui, M. (1997b): Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127.
- Sato, H., Tachi, Y., Shibutani, T. and Yui, M. (1997c): Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic

Energy Society of Japan, L12, p.561 [In Japanese].

Tsukamoto, M. and Ohe, T. (1988): Diffusion of Radioactive Cesium into Granitic Core Sample, 1988 Fall Meeting of the Atomic Energy Society of Japan, J45, p.169 [In Japanese].

## 2.4.6 Tritiated water (HTO)

The  $D_e$  values for tritiated water (HTO) in acid crystalline rock have been reported for granodiorite [PNC, 1996; Sato and Shibutani, 1994; Sato et al., 1997a, 1997b], altered granodiorite [PNC, 1996; Sato and Shibutani, 1994; Sato et al., 1997a, 1997b] and fracture fillings [Sato et al., 1997a, 1997b]. The reason which the fracture fillings are included in acid crystalline rock is as described in section 2.4.3(CI). Tritiated water (half-life: 12.3 y,  $\beta^-$  decay property) is not directly included in important nuclides for performance assessment of the geological disposal of HLW, but it is generally regarded as a non-sorbing nuclide. Therefore, it is generally used as basic data to estimate pore structural factors such as FF and Gf. Since HTO is not concerned with redox condition, all measurements have been carried out under aerobic conditions. Since HTO is one which part of water molecule was labelled by  $^3\text{H}$ , which is a radioactive hydrogen, it is considered to be a neutral species in solution and does not occur interaction with the surface of minerals composing rock.

All  $D_e$  data have been obtained by through-diffusion method. The  $D_a$  data have been derived by the time-lag method [e.g. Muurinen et al., 1987] from breakthrough curves in the through-diffusion experiments. In situ groundwater was used as porewater for all measurements. All measurements have been performed at 23 °C. The pH of the porewater ranged from 8.7 to 8.92, but the pH of the porewater doesn't influence on HTO behaviour.

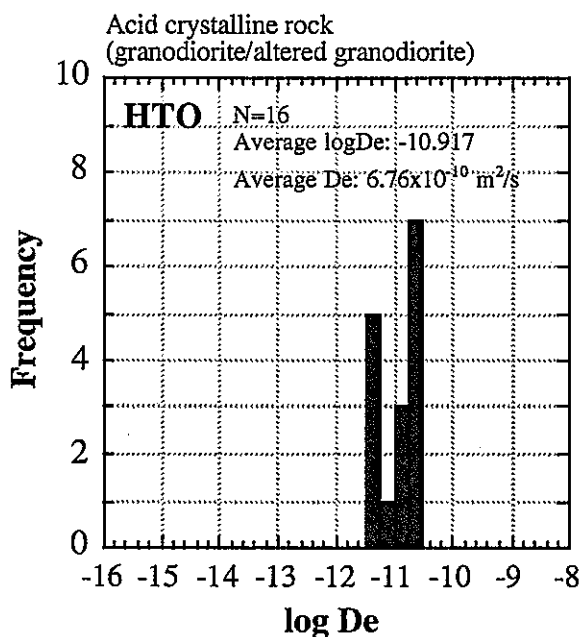
Figures 2.4.6-1 and 2.4.6-2 show histograms for  $D_e$  and  $D_a$  values of HTO in acid crystalline rock reported to the present, respectively. Totally, 16  $D_e$  data have been reported for granodiorite and altered granodiorite (including fracture fillings). The width of variation in  $D_e$  of HTO is small, being within one order of magnitude. Whilst, for  $D_a$ , 16 data have been totally reported for granodiorite and altered granodiorite. The width of variation in  $D_a$  of HTO is also small in the same as that in  $D_e$ , being within one order of magnitude.

Figures 2.4.6-3 and 2.4.6-4 show plots of  $D_e$  and  $D_a$  values of HTO as a function of rock porosity for acid crystalline rock, respectively. Totally, 16  $D_e$  plots have been reported. The  $D_e$  shows a tendency to increase with increasing rock porosity. Also for  $D_a$ , totally, 16 plots have been reported. However,  $D_a$  does not clearly show porosity dependency, being approximately constant between  $10^{-10}$  and  $10^{-9}$  m<sup>2</sup>/s. This  $D_a$  level is higher than values obtained under the same conditions by one to two orders of magnitudes. The  $D_a$  values are generally below  $10^{-11}$  m<sup>2</sup>/s.

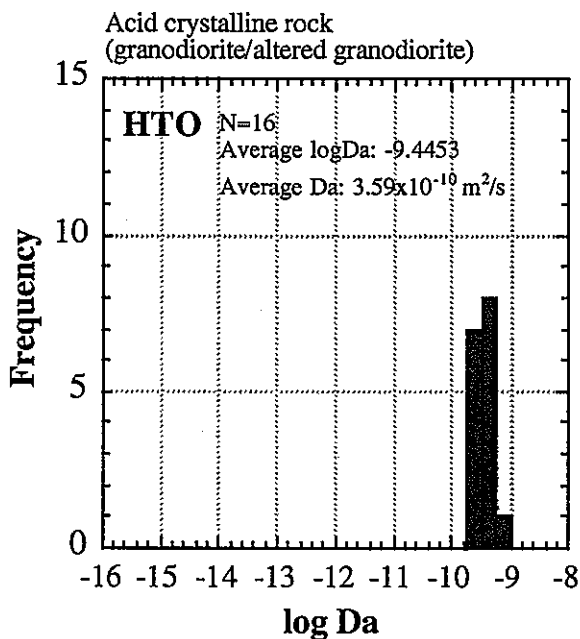
Though  $D_0$  of HTO has not been directly reported, for heavy water, a temperature dependency of  $D_0$  values has been reported [Chemical Handbook, 1975]. If  $D_0$  of heavy water is equal to that of HTO, that of HTO is estimated to be  $2.14 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C. The FF and



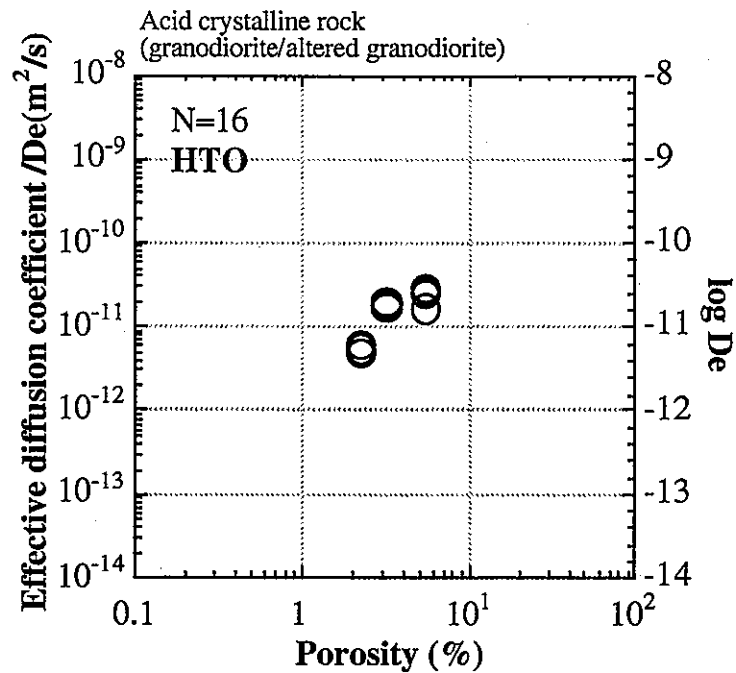
Gf values of individual rocks were calculated in the same way as Ba, Be, Cl, etc. Porosity dependencies for FF and Gf will be discussed in detail in section 2.4.20.



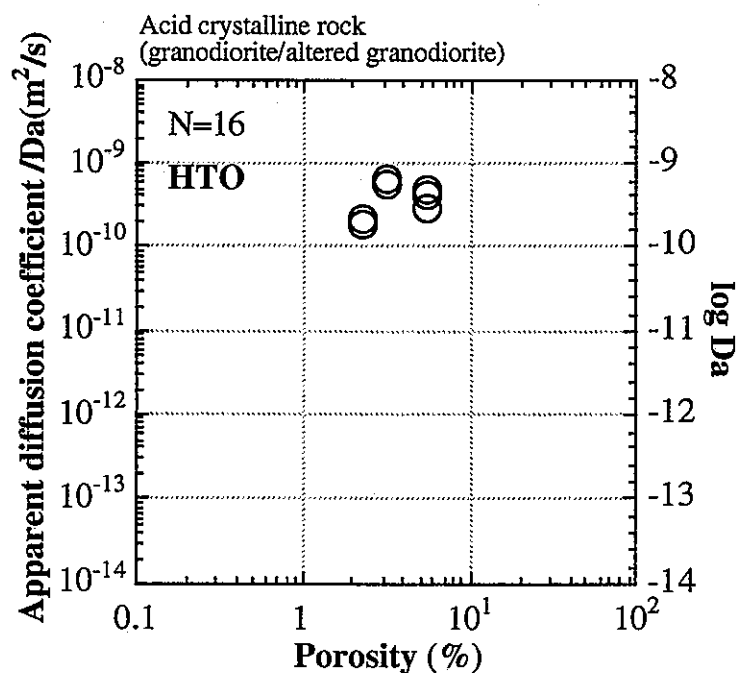
**Figure 2.4.6-1** Histogram for De values of HTO in acid crystalline rock (granodiorite, altered granodiorite)  
The width of variation in De of HTO is small, being within one order of magnitude.



**Figure 2.4.6-2** Histogram for Da values of HTO in acid crystalline rock (granodiorite, altered granodiorite)  
The width of variation in Da of HTO is also small in the same as that in De, being within one order of magnitude.



**Figure 2.4.6-3**  $D_e$  values of HTO as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)  
The  $D_e$  clearly shows a tendency to increase with increasing rock porosity as reported generally.



**Figure 2.4.6-4**  $D_a$  values of HTO as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)  
No significant porosity dependency is found for  $D_a$  in a porosity range of 2.3 to 5.6 %.

## References

- Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.205~207 [In Japanese].
- Sato, H. and Shibutani, T. (1996): Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste, PNC Technical Report, PNC TN1100 96-010, pp.139~144, Power Reactor and Nuclear Fuel Development Corporation [In Japanese].
- Sato, H., Shibutani, T., Tachi, Y., Ota, K., Amano, K. and Yui, M. (1997a): Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127, Power Reactor and Nuclear Fuel Development Corporation.
- Sato, H., Tachi, Y., Shibutani, T. and Yui, M. (1997b): Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan [In Japanese].

## 2.4.7 Iodine (I)

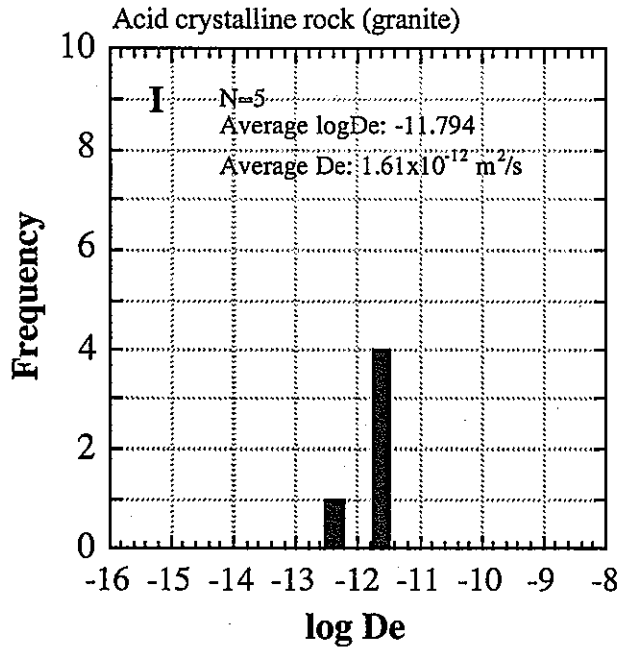
The  $D_e$  values for I ( $I^-$ ) in acid crystalline rock have been reported for granite [Kita et al., 1989; Nishiyama et al., 1990; Yamaguchi et al., 1997]. All data ( $D_e$  and  $D_a$ ) have been obtained under aerobic conditions, because I (iodine) is not sensitive to redox condition. Iodine is not included in important elements for performance assessment of the geological disposal of HLW, but it (especially  $^{129}I$ ) is regarded as one of the important elements for performance assessment in LLW. The chemical behaviour and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.3(I).

The  $D_e$  data have been obtained by through-diffusion method and  $D_a$  data have been derived by the time-lag method [e.g. Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor. Distilled water, deionized water and in situ groundwater were used as porewater in the through-diffusion experiments. All measurements have been performed at room temperature. The pH of the porewater is not reported. The porosities of rocks used in the measurements ranged between 0.4 and 0.8 %.

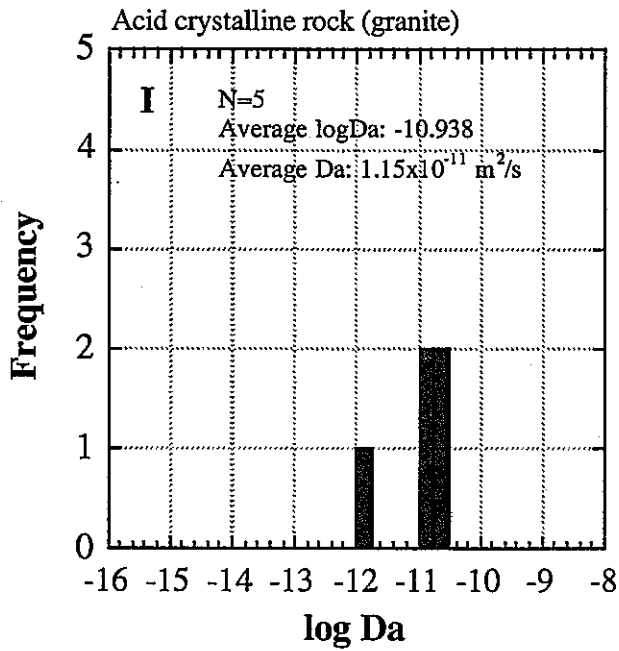
Figures 2.4.7-1 and 2.4.7-2 show histograms for  $D_e$  and  $D_a$  values of  $I^-$  in acid crystalline rock reported to date, respectively. Totally, 5  $D_e$  data have been reported for granite. For  $D_a$ , totally, 5 data have been reported for granite. For both diffusion coefficients, a variation of approximately one order of magnitude is found, though the width of variation in porosity is small to be 0.4-0.8 %. The probable reason is the change in pore structural factor caused by the change in porosity. Since I (iodine) is generally regarded as a non-sorbing element, both diffusion coefficients are considered to have depended on rock porosity.

Figures 2.4.7-3 and 2.4.7-4 show plots of  $D_e$  and  $D_a$  values of  $I^-$  as a function of rock porosity for acid crystalline rock reported to date, respectively. Five plots have been totally reported for both  $D_e$  and  $D_a$ . Both  $D_e$  and  $D_a$  show a tendency to increase with increasing rock porosity. The  $D_e$  generally depends on porosity, but  $D_a$  does not depend on porosity from the past examples. Whether  $D_a$  values of  $I^-$  in this case depends on porosity or not is difficult to discuss, because the number of data is too short.

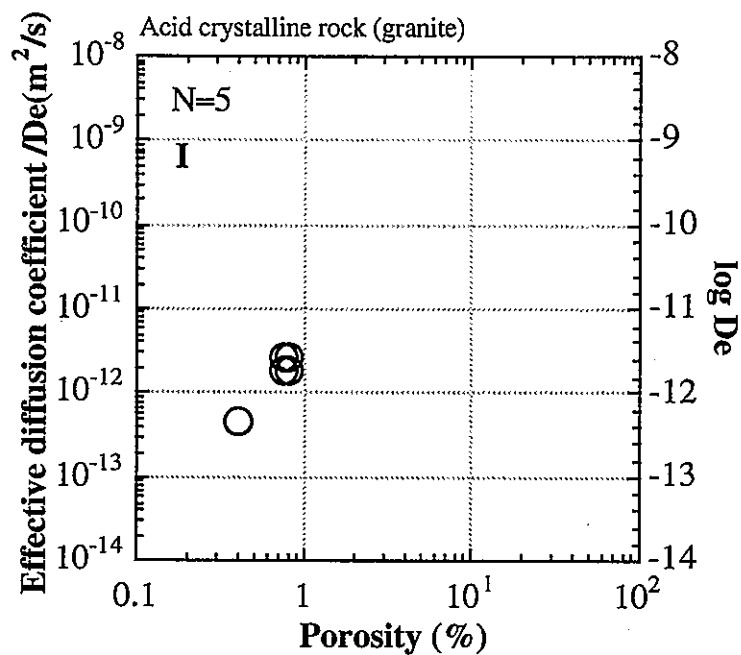
The  $D_0$  of  $I^-$  ( $2.0 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C), FF and Gf were estimated in the same as section 2.1.3(I). Porosity dependencies for FF and Gf will be discussed in detail in section 2.4.20.



**Figure 2.4.7-1** Histogram for De values of I<sup>-</sup> ion in acid crystalline rock (granite)  
 A variation of approximately one order of magnitude is found in De, though the width of variation in porosity is small to be 0.4~0.8 %.

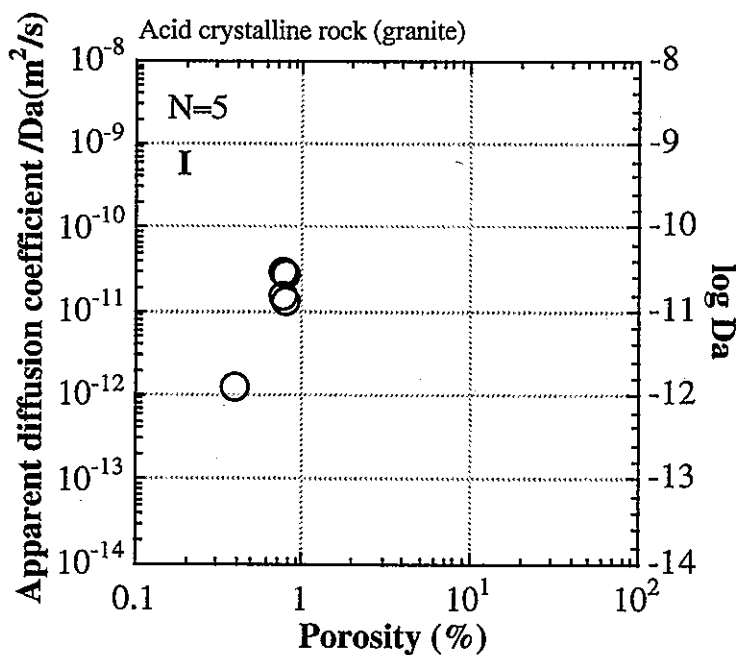


**Figure 2.4.7-2** Histogram for Da values of I<sup>-</sup> ion in acid crystalline rock (granite)  
 A variation of approximately one order of magnitude is found in Da, though the width of variation in porosity is small to be 0.4~0.8 %.



**Figure 2.4.7-3** De values of I<sup>-</sup> ion as a function of rock porosity for acid crystalline rock (granite)

The De shows a tendency to increase with increasing rock porosity.



**Figure 2.4.7-4** Da values of I<sup>-</sup> ion as a function of rock porosity for acid crystalline rock (granite)

The Da also shows a tendency to increase with increasing rock porosity in the same as De.

## References

- Kita, H., Iwai, T. and Nakashima, S. (1989): Diffusion Coefficient Measurement of an ion in Pore Water of Granite and Tuff, *Journal of the Japan Society of Engineering Geology*, **30-2**, pp.26~32 [In Japanese].
- Nishiyama, K., Nakashima, S., Tada, R. and Uchida, T. (1990): Diffusion of an Ion in Rock Pore Water and Its Relation to Pore Characteristics, *Mining Geology*, **40** (5), pp.323~336.
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, *Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X* (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Yamaguchi, T. and Nakayama, S. (1997): Effect of Sorption on Diffusivity in Micropores, 1997 Annual Meeting of the Atomic Energy Society of Japan, L10, p.559 [In Japanese].

## 2.4.8 Potassium (K)

The  $D_e$  values for K ( $K^+$ ) in acid crystalline rock have been reported for granite [Conca et al., 1992; Conca and Wright, 1993]. Potassium is not sensitive to redox condition. Therefore, all measurements have been carried out under aerobic conditions. Potassium is not particularly regarded as an important element in performance assessment of the geological disposal of HLW, but K chemically shows similar behaviour to alkaline metals such as Na and Cs in solution. The chemical behaviour and treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.4(K).

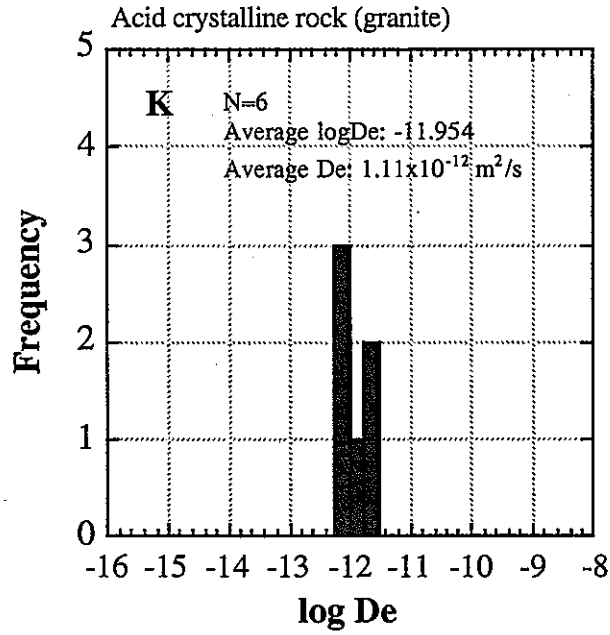
All  $D_e$  data have been obtained by electrical conductivity method. However,  $D_a$  can not be obtained from this method. Therefore, no  $D_a$  for K has been reported. In the measurements, rock samples were initially saturated with a 0.1 M KCl solution and then electrical conductivities were measured. The  $D_e$  values were determined by the Nernst-Einstein equation [Robinson and Stokes, 1959]. The principle of these measurements is as described in more detail in section 1.3.6. In these measurements, since an electrolyte solution which concentration is known is used instead of tracer such as KCl, the porewater and tracer are the same. Although it is not described, it seems that all measurements have been performed at room temperature. The pH of the porewater is not reported.

Figure 2.4.8-1 shows a histogram for  $D_e$  values of  $K^+$  in acid crystalline rock reported to date. Totally, 6  $D_e$  data have been reported for granite. No  $D_a$  has been reported. The width of variation in  $D_e$  is within one order of magnitude. This may be because rock porosities for all samples are the same.

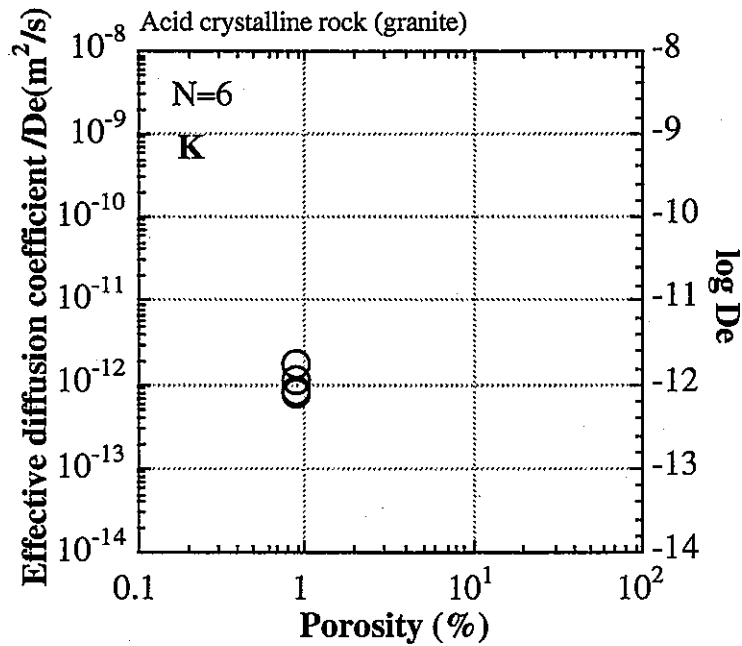
Figure 2.4.8-2 shows a plot of  $D_e$  values of  $K^+$  as a function of rock porosity for acid crystalline rock. Since all  $D_e$  data were obtained for samples with the same porosity, porosity dependencies for  $D_e$  can not be discussed here.

The  $D_o$  of  $K^+$ , FF and Gf were estimated in the same way as section 2.1.4(K). Porosity dependency for FF and Gf will be also discussed in more detail summarizing in section 2.4.20.





**Figure 2.4.8-1** Histogram for  $D_e$  values for  $K^+$  ion in acid crystalline rock (granite)  
 The width of variation in  $D_e$  is small to be within one order of magnitude. This is because rock porosities for all samples are the same.



**Figure 2.4.8-2**  $D_e$  values of  $K^+$  ion as a function of rock porosity for acid crystalline rock (granite)  
 Since rock porosities for all samples are the same, porosity dependency for  $D_e$  can not be discussed here.

## References

- Conca, J. L. and Wright, J. (1992): Direct Determinations of Unsaturated Flow and Transport, Proceedings of the Twelfth Annual American Geophysical Union Hydrology Days Meeting, Fort Collins, March 1992, pp.103~116.
- Conca, J. L., Apped, M. J. and Arthur, R. C. (1993): Direct Determinations of Transport Parameters in Repository Materials, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management XVI (Interrante, C. G. and Pabalan, R. T. eds.), Vol.294, pp.839~844, Pittsburgh, Pennsylvania.
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.

## 2.4.9 Magnesium (Mg)

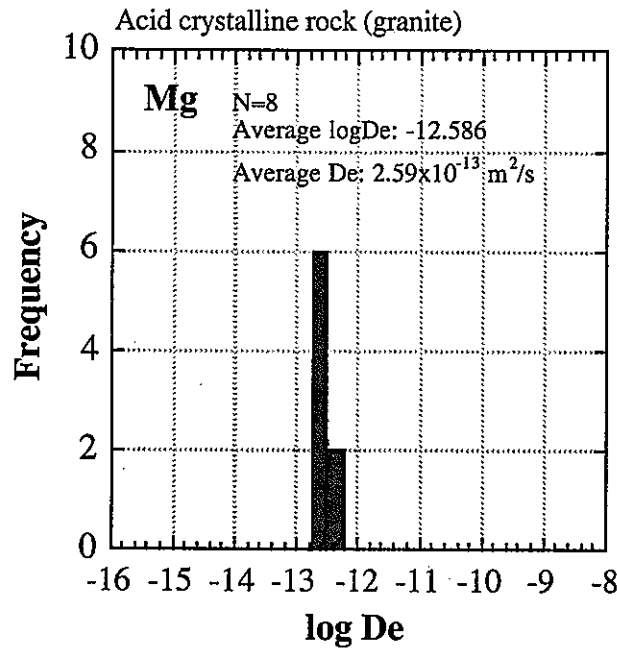
The  $D_e$  values for Mg ( $Mg^{2+}$ ) in acid crystalline rock have been reported for granite [Yamaguchi et al., 1993]. Since Mg is not sensitive to redox condition, all measurements have been carried out under aerobic conditions. Magnesium is not accounted as one of the important elements for performance assessment of the geological disposal of HLW. Since Mg is also one of the alkaline earth metals, it is presumed that elements which belong to the same group chemically show similar behaviour in solution. Furthermore,  $Mg^{2+}$  is considered to be able to be treated in the same as  $Ba^{2+}$ ,  $Be^{2+}$ ,  $Ni^{2+}$ ,  $Sr^{2+}$ ,  $Co^{2+}$  and  $Ra^{2+}$  if they take such chemical forms in solution.

All  $D_e$  values have been obtained by through-diffusion method, but no  $D_a$  has been reported. Since rock capacity factor has not been also reported,  $D_a$  can not be estimated. All measurements have been carried out in a 0.1 M KCl porewater system to keep the ionic strength of porewater in both cells. All experiments have been performed at 25 °C. Besides the reported porewater pH was 4 for all experiments. All measurements have been carried out using a mixed solution of  $Ba(NO_3)_2$ ,  $BeSO_4$ ,  $CoCl_2$ ,  $MgCl_2$ ,  $NiCl_2$  and  $Sr(NO_3)_2$ , what is called a multi-tracer or a cocktail tracer.

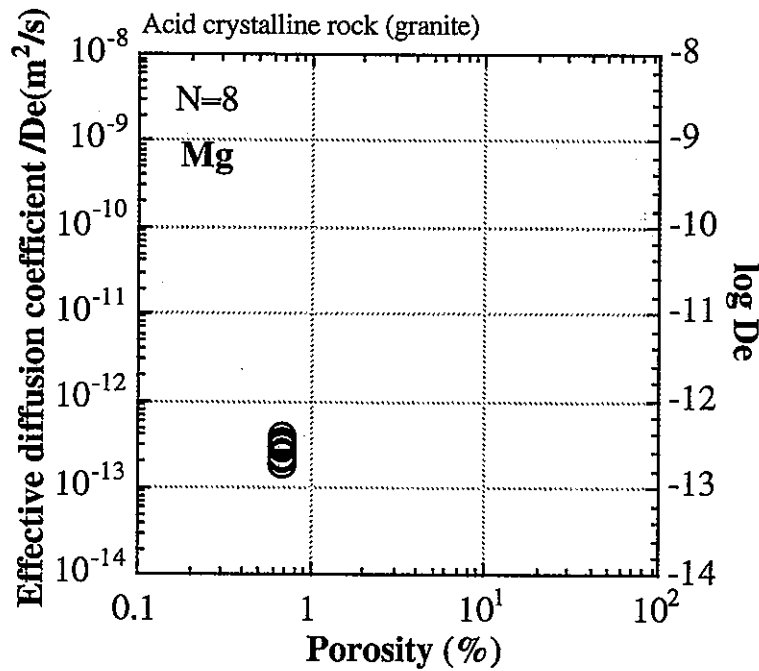
**Figure 2.4.9-1** shows a histogram for  $D_e$  values of  $Mg^{2+}$  in acid crystalline rock reported to date. Totally, 8 data have been reported for  $D_e$  of  $Mg^{2+}$  in granite. The width of variation in  $D_e$  was small to be within one order of magnitude. This is because all rock porosities are the same. As described above, no datum for  $D_a$  has been reported.

**Figure 2.4.9-2** shows a plot of  $D_e$  values of  $Mg^{2+}$  as a function of rock porosity for acid crystalline rock. Although the correlations were plotted for data reported both porosity and  $D_e$ , 8 plots have been totally reported. Since the reported porosities were all 0.68 %, porosity dependency for  $D_e$  can not be discussed. The reported  $D_e$  values were all in the order of  $10^{-13}$   $m^2/s$  and the width of variation in  $D_e$  was quite small to be within a factor of 2 in a deviation.

The  $D_o$  of  $Mg^{2+}$  is estimated to be  $7.1 \times 10^{-10}$   $m^2/s$  at 25 °C [Marcus, 1997] by the Nernst-Einstein equation [Robinson and Stokes, 1959; Li and Gregory, 1974; Chemical Handbook, 1975; Daniel and Shackelford, 1988; Shackelford and Daniel, 1991]. The FF and Gf values were also calculated based on  $D_e$ ,  $D_o$  and porosity. For porosity dependency for FF and Gf will be discussed summarizing in section 2.4.20.



**Figure 2.4.9-1** Histogram for De values of  $\text{Mg}^{2+}$  ion in acid crystalline rock (granite)  
The width of variation in rock porosity is small to be within one order of magnitude. This may be because all rock porosities are the same.



**Figure 2.4.9-2** De values of  $\text{Mg}^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite)  
The width of variation in De is quite small to be within a factor of 2 in a deviation.

## References

Chemical Handbook, Basic Version II ("Kagaku-binran"), 2nd ed., Chemical Society of Japan ("Nihon-kagakukai"), Maruzen, Tokyo (1975) [In Japanese].

Daniel, D. E. and Shackelford, C. D. (1988): Disposal Barriers That Release Contaminants only by Molecular Diffusion, Nuclear and Chemical Waste Management, Vol.8, pp.299~305.

Li, Y. and Gregory, S. (1974): Diffusion of Ions in Sea Water and in Deep-Sea Sediments, Geochimica et Cosmochimica Acta, Vol.38, pp.703~714.

Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.

Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.

Shackelford, C. D. and Daniel, D. E. (1991): Diffusion in Saturated Soil. I: Background, Journal of Geotechnical Engineering, Vol.117, No.3, American Society of Civil Engineering.

Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1993): Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796~803, the Atomic Energy Society of Japan.

### 2.4.10 Sodium (Na)

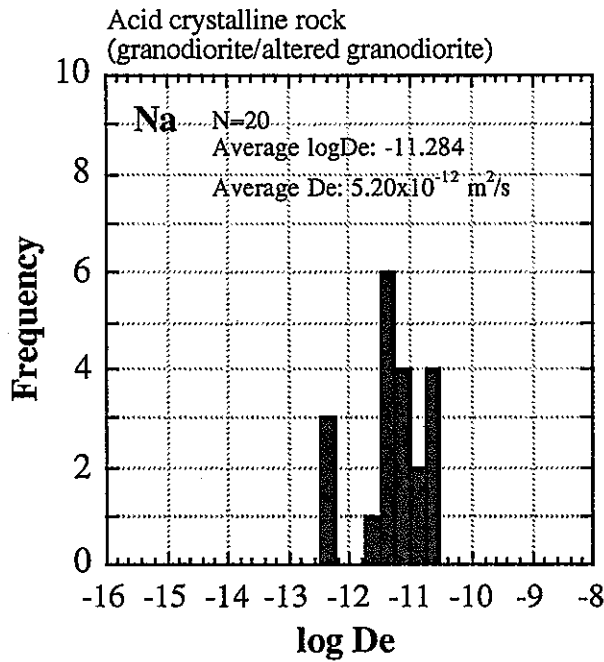
The  $D_e$  values for Na ( $\text{Na}^+$ ) in acid crystalline rock have been reported for granodiorite [Sato et al., 1997a, 1997b, 1997c; PNC, 1996; Sato and Shibutani, 1996], altered granodiorite [Sato et al., 1997b, 1997c; PNC, 1996; Sato and Shibutani, 1996] and fracture fillings [Sato et al., 1997b, 1997c; PNC, 1996; Sato and Shibutani, 1996]. All measurements have been carried out under aerobic conditions. Sodium is not considered as one of the important elements for performance assessment of the geological disposal of HLW. As described in sections 2.1.5 and 2.3.5(Na), Na chemically shows similar behaviour to alkaline metals such as  $\text{Cs}^+$  and  $\text{K}^+$  in solution. Therefore, it is considered that even no datum for Cs which is one of the important elements for performance assessment is existing, these data for Na and K can be used as supplementary data. The chemical behaviour and basic treatment in performance assessment of the geological disposal of HLW are the same as section 2.1.5(Na).

All  $D_e$  values for Na have been obtained by through-diffusion method and  $D_a$  values were derived based on the time-lag method [e.g. Muurinen et al., 1987] or based on  $D_e$  and rock capacity factor. The porewater initially saturated is distilled water for measurements for granodiorite [Sato et al., 1997a] and in-situ groundwater for those for granodiorite, altered granodiorite and fracture fillings [Sato et al., 1997b, 1997c; PNC, 1996; Sato and Shibutani, 1996]. All measurements have been performed at 25 °C. The reported porewater pH was in a range of 5.5 to 8.92 and NaCl was used as a tracer solution for the measurements. The concentration of the tracer is also various from 0.05 M to 10 %. For experimental period, although for all measurements are not reported, almost all measurements are 30 days.

Figures 2.4.10-1 and 2.4.10-2 show histograms for  $D_e$  and  $D_a$  values of  $\text{Na}^+$  for acid crystalline rock reported to date, respectively. Totally, 20  $D_e$  and 16  $D_a$  data have been reported for granodiorite and altered granodiorite (for fracture fillings are also included). For  $D_e$ , the width of variation is approximately 2 orders of magnitude. For  $D_a$ , the width of variation is narrower than that for  $D_e$ , being within 2 orders of magnitude.

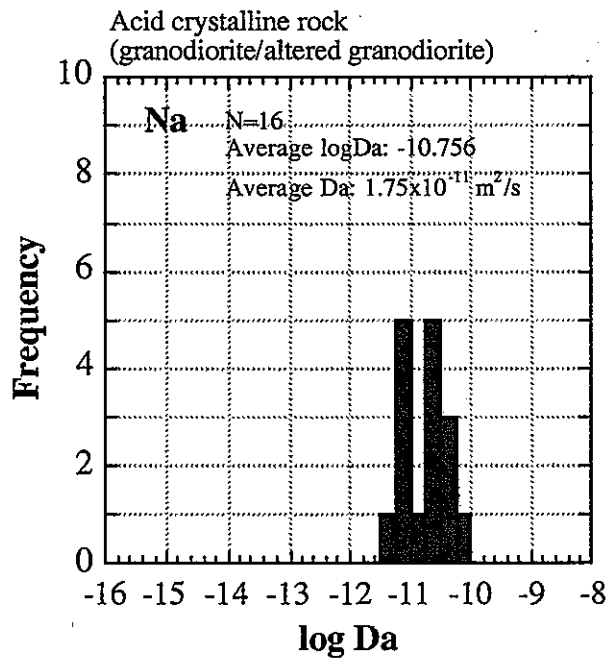
Figures 2.4.10-3 and 2.4.10-4 show plots of  $D_e$  and  $D_a$  values of  $\text{Na}^+$  as a function of rock porosity for acid crystalline rock, respectively. The  $D_e$  clearly shows a tendency to increase with increasing rock porosity. On the other hand, no significant tendency is found for  $D_a$ . Although the same porosity, the width of variation in  $D_a$  is about one order of magnitude. This may be due to the variation in sorptivity onto these rocks.

The  $D_o$  of  $\text{Na}^+$ , FF and Gf were estimated in the same way as section 2.1.5(Na). Porosity dependencies for FF and Gf will be described in detail in section 2.4.20.



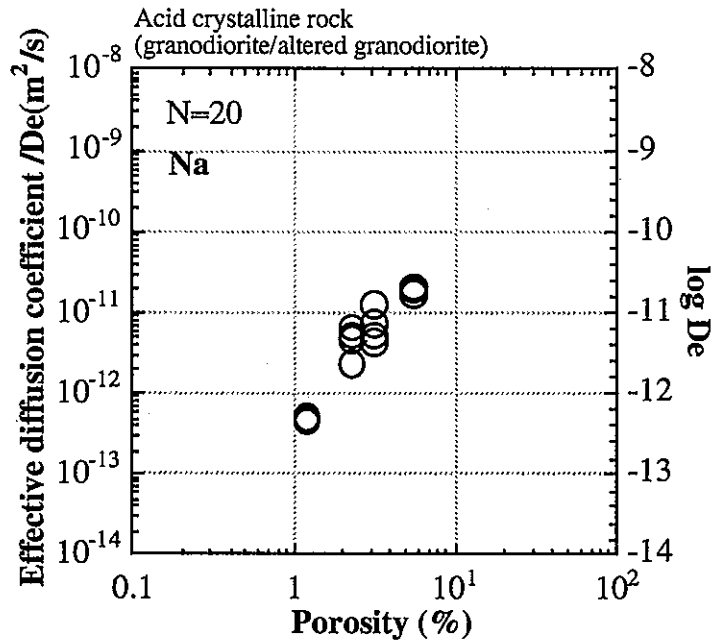
**Figure 2.4.10-1** Histogram for De values of Na<sup>+</sup> ion in acid crystalline rock (granodiorite, altered granodiorite)

The De values have a width of variation of 2 orders of magnitude and the average value of De was estimated to be  $5.20 \times 10^{-12} \text{ m}^2/\text{s}$ .



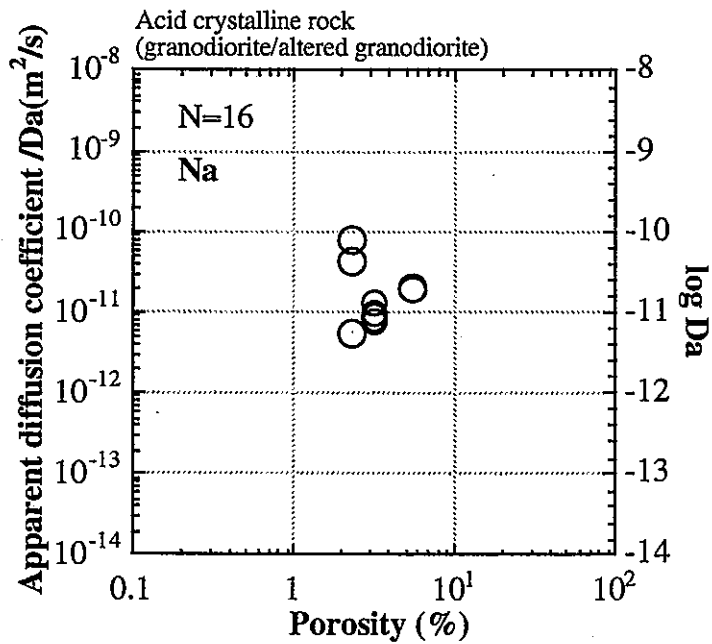
**Figure 2.4.10-2** Histogram for Da values of Na<sup>+</sup> ion in acid crystalline rock (granodiorite, altered granodiorite)

For Da, the width of variation is smaller than that for De, being within 2 orders of magnitude.



**Figure 2.4.10-3** De values of Na<sup>+</sup> ion as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)

The De clearly depends on rock porosity as shown in figure, increasing with increasing porosity.



**Figure 2.4.10-4** Da values of Na<sup>+</sup> ion as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)

No significant tendency is found between Da and rock porosity and the width of variation in Da is about one order of magnitude although the same porosity.



## References

- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.205~207 [In Japanese].
- Sato, H. and Shibutani, T. (1996): Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste, PNC Technical Report, PNC TN1100 96-010, pp.139~144 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997a): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, **26**, pp.119~133, Elsevier Science.
- Sato, H., Shibutani, T., Tachi, Y., Ota, K., Amano, K. and Yui, M. (1997b): Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127.
- Sato, H., Tachi, Y., Shibutani, T. and Yui, M. (1997c): Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan, L12, p.561 [In Japanese].

### 2.4.11 Nickel (Ni)

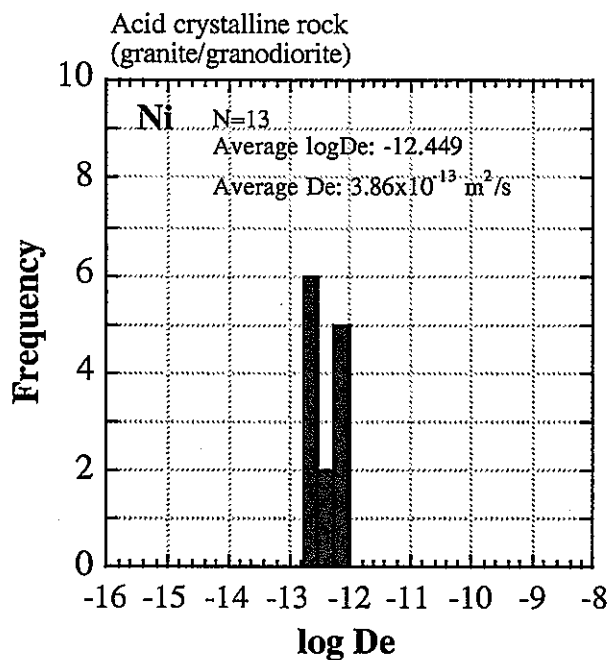
The De values for Ni ( $\text{Ni}^{2+}$ ) in acid crystalline rock have been reported for granite [Yamaguchi et al., 1993] and granodiorite [Sato et al., 1993; Sato and Shibutani, 1994; PNC, 1994, 1996]. All measurements have been carried out under aerobic conditions. Nickel is considered as one of the important elements for performance assessment of the geological disposal of HLW, because Ni-59 is produced as an activation product of the corrosion products in a power reactor. The chemical behaviour and basic treatment in performance assessment of the geological disposal of HLW are as described in section 2.1.6(Ni).

All De data have been obtained by through-diffusion method and Da values have been derived based on the time-lag method [e.g. Muurinen et al., 1987] or based on De and rock capacity factor. The measurements have been carried out in a distilled water system [Sato et al., 1993; Sato and Shibutani, 1994; PNC, 1994, 1996] and in a 0.1 M KCl solution system [Yamaguchi et al., 1993]. All measurements have been performed at 25 °C. Moreover, the pH of the porewater used was between 4 and 6 in the measurements. A nickel dichloride ( $\text{NiCl}_2$ ) solution was used as a tracer in all measurements. For some measurements, a mixed solution of  $\text{Ba}(\text{NO}_3)_2$ ,  $\text{BeSO}_4$ ,  $\text{CoCl}_2$ ,  $\text{MgCl}_2$ ,  $\text{NiCl}_2$  and  $\text{Sr}(\text{NO}_3)_2$ , what is called a multi-tracer or cocktail tracer was used [Yamaguchi et al., 1993]. The species of Ni in the porewater in this solution condition is considered to be  $\text{Ni}^{2+}$  from the pH of the porewater and redox condition.

Figures 2.4.11-1 and 2.4.11-2 show histograms for De and Da values of  $\text{Ni}^{2+}$  in acid crystalline rock, respectively. Totally, 13 De and 5 Da data have been reported for granite and granodiorite. The width of variation in De is small to be within one order of magnitude. This may be because the variation of rock porosity is small to be 0.68 to 1.2 %. For Da, the variation is smaller than that for De. For Da, the variation of rock porosity is smaller than that for De, being between 0.88 and 1 %.

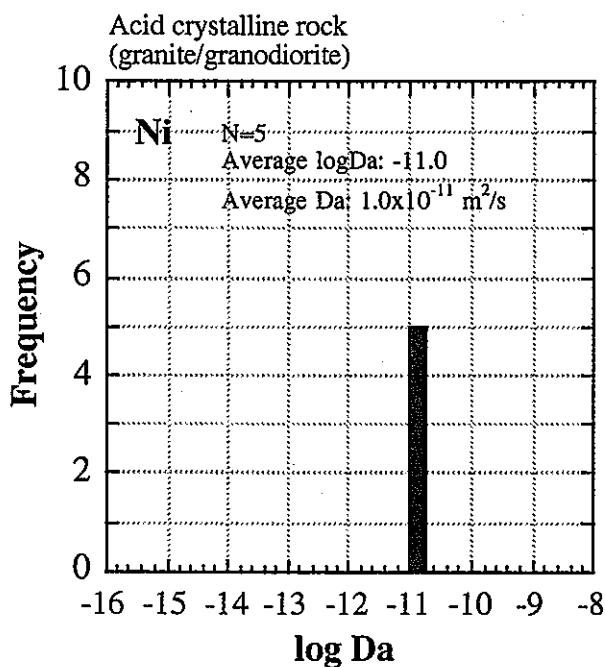
Figures 2.4.11-3 and 2.4.11-4 show plots of De and Da values of  $\text{Ni}^{2+}$  as a function of rock porosity for acid crystalline rock, respectively. Totally, 12 De plots have been reported to date. Although De seems to show a tendency to increase with increasing rock porosity, whether De depends on rock porosity or not is difficult to judge because the porosity range is narrow. For Da, 4 plots have been totally reported up to the present. For Da, no porosity dependency is found although it is unclear because the rock porosity is too narrow to discuss for porosity dependency.

The Do of  $\text{Ni}^{2+}$ , FF and Gf were estimated in the same way as section 2.1.6(Ni). Porosity dependencies for FF and Gf will be described in detail in section 2.4.20.



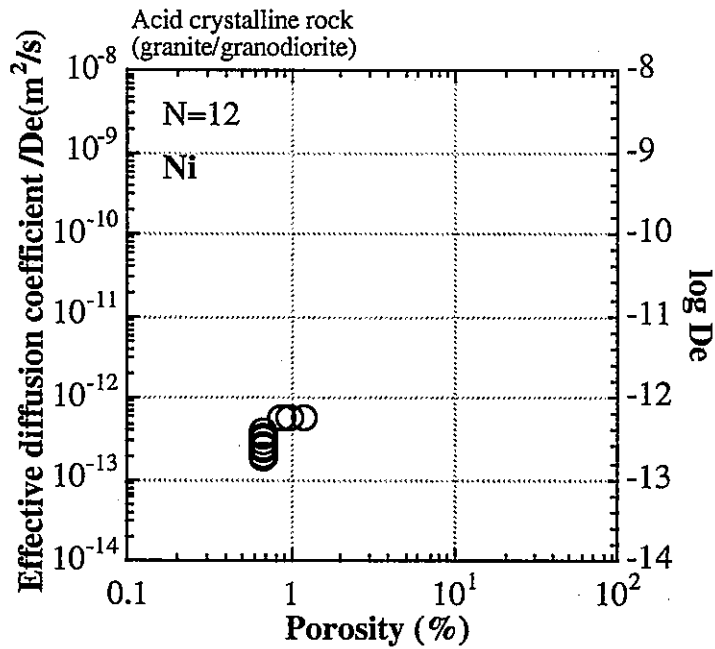
**Figure 2.4.11-1** Histogram for De values of Ni<sup>2+</sup> ion in acid crystalline rock (granite, granodiorite).

The width of variation in De is within one order of magnitude, being small. This may be because the variation of rock porosity is small to be in a range of 0.68 to 1.2 %.



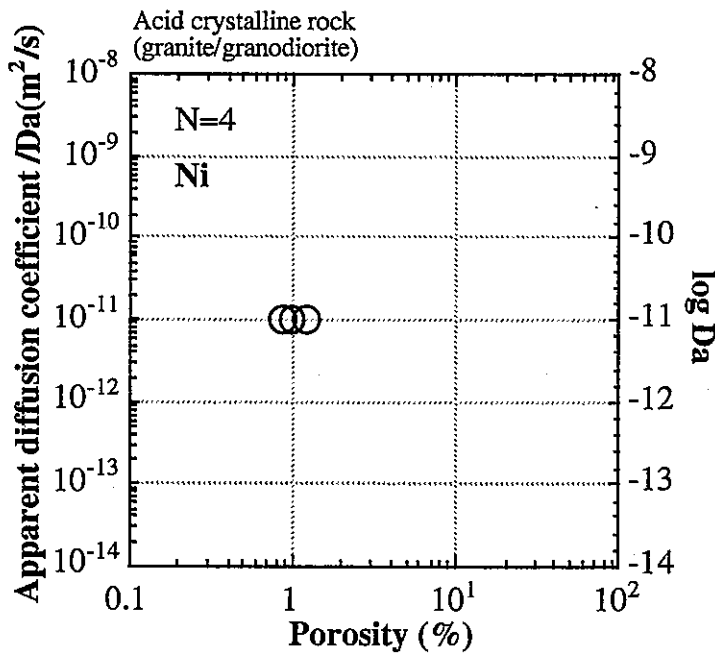
**Figure 2.4.11-2** Histogram for Da values of Ni<sup>2+</sup> ion in acid crystalline rock for acid crystalline rock (granite, granodiorite)

For Da, the variation of rock porosity is smaller than that for De, being between 0.88 to 1.0 %.



**Figure 2.4.11-3**  $D_e$  values of  $Ni^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite, granodiorite)

Although  $D_e$  seems to show a tendency to increase with increasing rock porosity, it is difficult to judge whether  $D_e$  depends on rock porosity or not, because the porosity range is narrow.



**Figure 2.4.11-4**  $D_a$  values of  $Ni^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite, granodiorite)

For  $D_a$ , no porosity dependency is found. However, since the porosity range is narrow, it is difficult to judge whether there is porosity dependency in  $D_a$  or not.

## References

- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1994): Current Status for Research and Development of Geological Disposal -FY1993-, PNC Technical Report, PNC TN1410 94-094, pp.131~133 [In Japanese].
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].
- Sato, H., Shibutani, T., Yoshikawa, H. and Yui, M. (1993): A Study on Diffusion Mechanism for Cs, Ni and Sm in Granodiorite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M29, p.667 [In Japanese].
- Sato, H. and Shibutani, T. (1994): Study on Adsorption and Diffusion of Nuclides in Buffer Material and Geosphere, PNC Technical Review, No.91, PNC TN8410 94-284, pp.71~89 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, **26**, pp.119~133, Elsevier Science.
- Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1993): Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, **30** (8), pp.796~803.

## 2.4.12 Neptunium (Np)

The De values of Np ( $\text{NpO}_2^+$ ) in acid crystalline rock have been reported for granite [Yamaguchi et al., 1997, 1998; Kumata et al., 1990]. Since Np takes Np(III), (IV), (V), (VI) and (VII) in solution and is sensitive to redox condition, some measurements have been carried out under reducing conditions [Yamaguchi et al., 1998]. Neptunium generally takes Np(V) under aerobic conditions. Neptunium is regarded as one of the important elements for performance assessment of the geological disposal of HLW and Np-237 (decay property:  $\alpha$ , half-life: 2.14E6 y) and Np-239 (decay property:  $\beta^-$ , half-life: 2.355 d) are accounted for performance assessment. In this case, both nuclides are included in decay chain nuclides. The half-life of Np-239 is quite short, but this is one of the decay chain nuclides, constantly continuing to exist as a daughter nuclide. Therefore, this nuclide is also included as an important element for performance assessment.

The species of Np in solution is affected by not only redox condition, but also is affected by the pH of the solution and the concentrations of carbonates in the solution. For example,  $\text{NpO}_2^+$  (neptunyl) is dominant in a relatively low pH region under aerobic conditions, but carbonate complexes such as  $\text{NpO}_2\text{CO}_3^-$  become predominant in a high pH region. To FRHP (Fresh-Reducing-High pH) porewater and SRHP (Saline-Reducing-High pH) porewater,  $\text{Np}(\text{OH})_4(\text{aq})$  and  $\text{Np}(\text{OH})_2(\text{CO}_3)_2^{2-}$ , and  $\text{Np}(\text{OH})_4(\text{aq})$  have been predicted to be predominant, respectively [JNC, 1999].

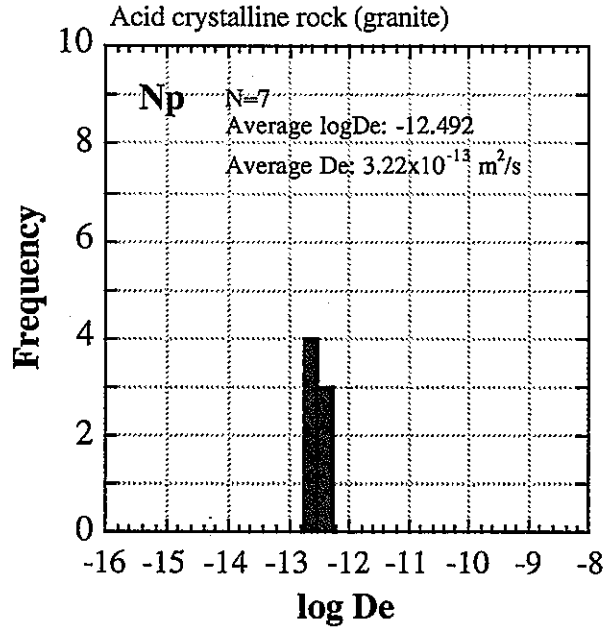
All De values have been obtained by through-diffusion method and Da values have been derived based on the time-lag method [e.g. Muurinen et al., 1987] or De and rock capacity factor. The measurements have been carried out in a natural groundwater system [Kumata et al., 1990; Yamaguchi et al., 1997] and in a 1M  $\text{NaHCO}_3$  solution system [Yamaguchi et al., 1998]. Besides the measurements have been performed at room temperature [Kumata et al., 1990; Yamaguchi et al., 1997] and at  $26 \pm 1$  °C [Yamaguchi et al., 1998]. The through-diffusion experiments have been carried out in a pH range of 7.4 to 8.6. The measurements have been performed under aerobic conditions [Kumata et al., 1990; Yamaguchi et al., 1997] and under reducing conditions (Eh vs. SHE:  $-500 \sim -400$  mV) [Yamaguchi et al., 1998]. The valences of Np in this case are considered to be V and IV for the former and later, respectively.

Figures 2.4.12-1 and 2.4.12-2 show histograms for De and Da values of Np in acid crystalline rock reported, respectively. Totally, 7 De and 7 Da values have been reported for granite. In these studies, all granite samples were sampled from Inada, Ibaraki, Japan. The variation in De is small to be about a half order of magnitude. Whilst, the variation in Da is 2 orders of magnitude, although the variation of rock porosity is small to be from 0.49 to 0.74 %. Since all experimental conditions are not the same, comparison can not be simply made. In particular, Da is affected by chemical and redox conditions, because this parameter includes sorption property. Since the width of variation in De was small, such wide variation in Da is considered to be due to the variation of sorption property.

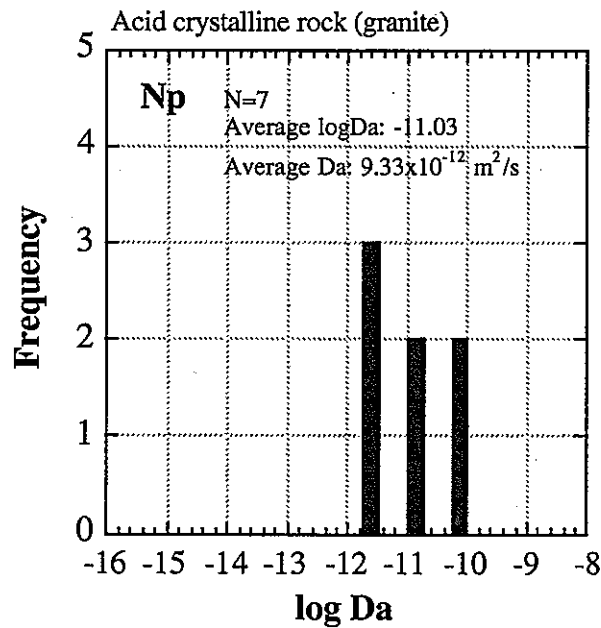
Figures 2.4.12-3 and 2.4.12-4 show plots of De and Da values of  $\text{NpO}_2^+$  as a function of rock porosity for acid crystalline rock, respectively. Since the width of variation in porosity was small, clear porosity dependency for Da is not found. For Da, a variation of

approximately one order of magnitude is found. This is considered to be due to the variation of sorption property.

The  $D_0$  of  $\text{NpO}_2^+$  is estimated to be  $1.2 \times 10^{-9} \text{ m}^2/\text{s}$  at  $25 \text{ }^\circ\text{C}$  by the Nernst-Einstein equation [e.g. Marcus, 1997]. The FF and Gf values were also calculated for  $\text{NpO}_2^+$ . Porosity dependencies for FF and Gf will be discussed in detail in section 2.4.20.



**Figure 2.4.12-1** Histogram for De values of Np in acid crystalline rock (granite)  
The width of variation in De is small to be about a half order of magnitude.



**Figure 2.4.12-2** Histogram for Da values of Np in acid crystalline rock (granite)  
The width of variation in Da is near 2 orders of magnitude. Since all experimental conditions are not the same, it is considered that the variation of Da was also wide, because Da includes sorption property and is affected by chemical and redox conditions.



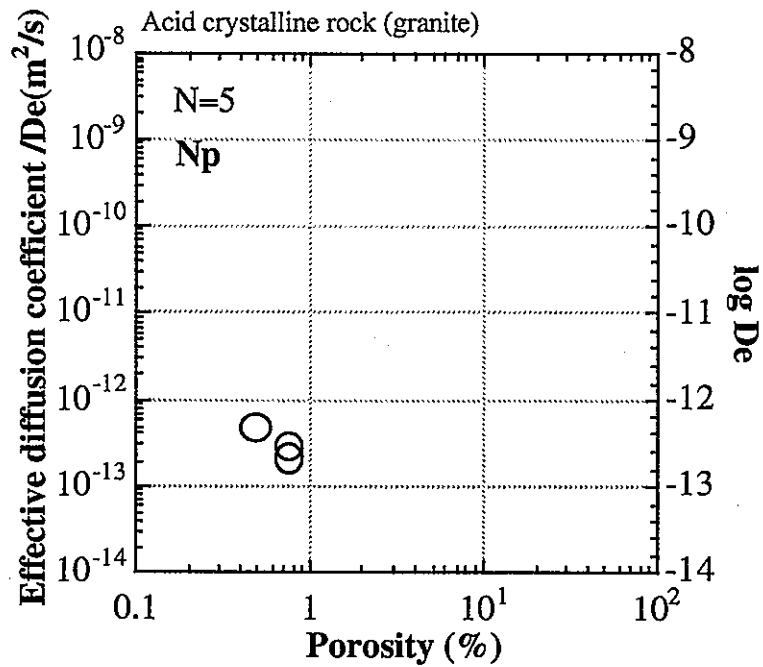


Figure 2.4.12-3  $D_e$  values of  $N_p$  as a function of rock porosity for acid crystalline rock (granite)

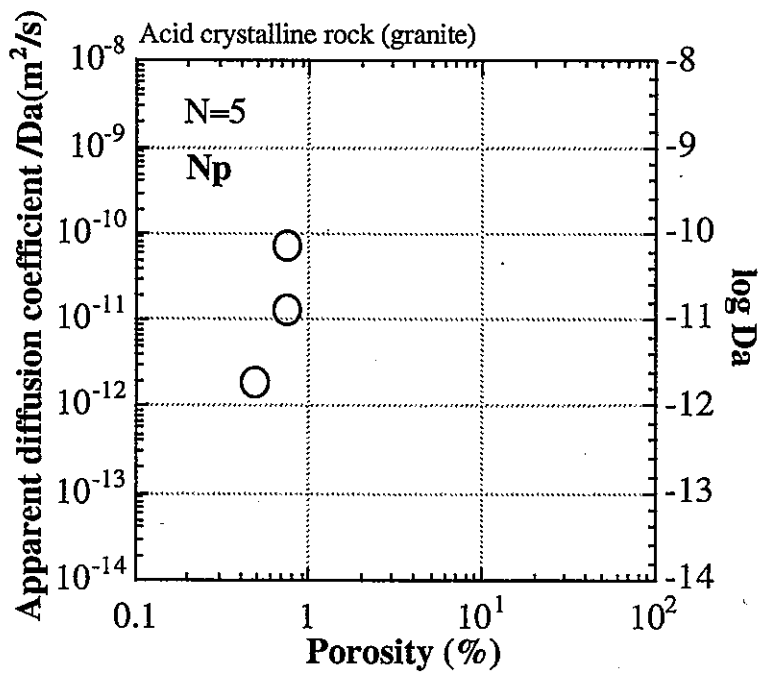


Figure 2.4.12-4  $D_a$  values of  $N_p$  as a function of rock porosity for acid crystalline rock (granite)

The variation in  $D_a$  is large, even though the variation of rock porosity is small.

## References

- Japan Nuclear Cycle Development Institute (JNC) (1999): H12 Project to Establish Technical Basis for HLW Disposal in Japan, Supporting Report 3, Safety Assessment, JNC Technical Report, JNC TN1400 99-013.
- Kumata, M., Iwai, T., Sagawa, T., Suzuki, T. and Nishiyama, K. (1990): Diffusion Experiment of a Radionuclide in Granite Rock Cores, JAERI Report, JAERI-M 90-179 [In Japanese].
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.168.
- Muurinen, A., Penttilä-Hiltunen, P. and Rantanen, J. (1987): Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite, Materials Research Society Symposium Proceedings on the Scientific Basis for Nuclear Waste Management X (Bates, J. K. and Seefeldt, W. B. eds.), Vol.84, pp.803~811.
- Yamaguchi, T., Sakamoto, Y., Nakayama, S. and Vandergraaf, T. T. (1997): Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, Journal of Contaminant Hydrology, **26**, pp.109~117, Elsevier Science, B. V.
- Yamaguchi, T., Nakayama, S. and Saito, M. (1998): Np and Pu Diffusion Experiments in Granite under Antoxic Conditions, 1998 Fall Meeting of the Atomic Energy Society of Japan, M17, p.834 [In Japanese].

### 2.4.13 Plutonium (Pu)

The De values of Pu in acid crystalline rock have been reported for granite [Yamaguchi, 1996]. Plutonium takes Pu(III), (VI), (V), (VI) and (VII) in solution and Pu(IV) is the most stable in solution. Plutonium is accounted as one of the important elements for performance assessment of the geological disposal of HLW, and Pu-238 (decay property:  $\alpha$ , half-life: 87.7 y), Pu-239 (decay property:  $\alpha$ , half-life: 2.41E4 y), Pu-240 (decay property:  $\alpha$ , half-life: 6550 y), Pu-241 (decay property:  $\beta^-$ , half-life: 14.4 y) and Pu-242 (decay property:  $\alpha$ , half-life: 3.76E5 y) are taken into account for performance assessment. All nuclides are included in decay chain nuclides in this case. The half-life of Pu-238 is short, but this is one of the decay chain nuclides, constantly continuing to exist as a daughter nuclide.

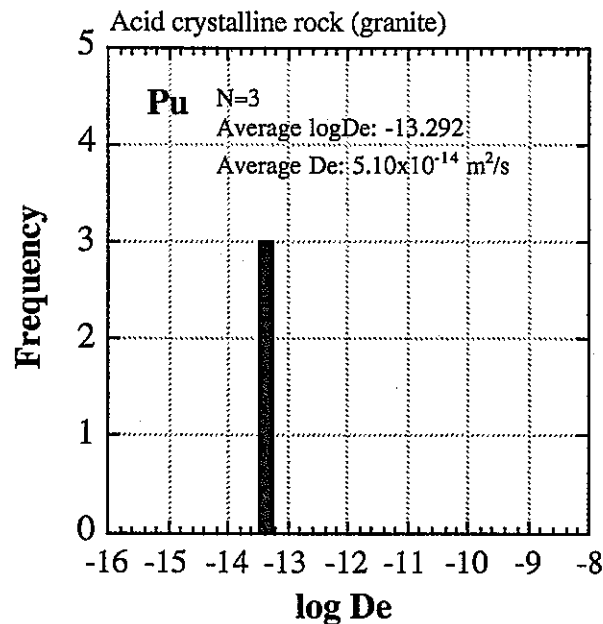
The species of Pu in solution is affected by redox condition, the pH of the solution and the concentrations of dissolved carbonates in the solution. To FRHP (Fresh-Reducing-High pH) porewater,  $\text{Pu}(\text{OH})_2(\text{CO}_3)_2^{2-}$ ,  $\text{Pu}(\text{CO}_3)_2^-$ ,  $\text{Pu}(\text{CO}_3)_3^{3-}$  and  $\text{PuCO}_3^+$  have been predicted to be predominant, but to SRHP (Saline-Reducing-High pH) porewater,  $\text{PuOH}^{2+}$ ,  $\text{PuCO}_3^+$  and  $\text{Pu}(\text{OH})_2^+$  have been predicted to be predominant [JNC, 1999]. To porewater due to reaction between concrete supports (low alkaline cement) and FRHP groundwater,  $\text{Pu}(\text{CO}_3)_2^-$ ,  $\text{Pu}(\text{OH})_2(\text{CO}_3)_2^{2-}$  and  $\text{Pu}(\text{CO}_3)_3^{3-}$  have been predicted to be predominant [JNC, 1999].

According to Brookins [Brookins, 1988], in the system Pu-(C)-O-H, under relatively oxidizing conditions,  $\text{PuO}_2(\text{OH})_2\text{CO}_3^{2-}$  is predominant in a wide pH range. In the system Pu-O-H-C,  $\text{Pu}(\text{OH})_5^-$  is predominant in a pH range of < around 6 under both aerobic and reducing conditions. But, for both systems, in a low pH reageon,  $\text{Pu}^{3+}$  is predominant.

All De values have been obtained by through-diffusion method, but no Da has been reported. The measurements have been carried out in a mixture solution of  $\text{NaHCO}_3$  and  $\text{NaNO}_2$ , in which total concentrations of  $\text{CO}_3^{2-}$  and  $\text{NO}_2^-$  are 0.1 and 0.01 M, respectively. All measurements have been performed at 25 °C and the pH of the porewater was 9.3. Although Pu is redox sensitive, all measurements have been carried out under aerobic conditions. The dominant chemical species of Pu in the solution was estimated to be  $\text{Pu}(\text{OH})_2(\text{CO}_3)_2^{2-}$  in the literature from the pH and chemical conditions of the porewater [Yamaguchi, 1996].

Figure 2.4.13-13 shows a histogram for De values of Pu in acid crystalline rock reported so far. Totally, 3 De data have been reported for granite. It is reported that the error of De values was evaluated to be  $5.1 \pm 2.2 \times 10^{-14} \text{ m}^2/\text{s}$  [Yamaguchi, 1996]. No directly obtained Da has been reported. Besides, since no rock capacity factor is reported, Da can not be calculated. No porosity is also reported, porosity dependency for De can not be discussed here.

The Do for Pu carbonate complex is not reported. The Do for Pu is reported only for  $\text{PuO}_2^{2+}$  ( $7.9 \times 10^{-10} \text{ m}^2/\text{s}$  at 25 °C) [Marcus, 1997]. Since Do of carbonate complex can not be estimated, neither FF nor Gf can be discussed here.



**Figure 2.4.13-1** Histogram for De values of Pu in acid crystalline rock (granite)  
Totally, 3 De data have been reported for granite. For porosity dependency, since no porosity is reported, it can not be calculated.

## References

- Brookins, D. G. (1988): pH-Eh Diagrams for Geochemistry, Springer-Verlag, Berlin, Germany, pp.144~145.
- Japan Nuclear Cycle Development Institute (JNC) (1999): H12 Project to Establish Technical Basis for HLW Disposal in Japan, Supporting Report 3, Safety Assessment, JNC Technical Report, JNC TN1400 99-013.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.169.
- Yamaguchi, T. (1996): Diffusivity of Actinides in a Granite from Inada, Ibaraki, Japan, PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.156~160, Power Reactor and Nuclear Fuel Development Corporation.

## 2.4.14 Selenium (Se)

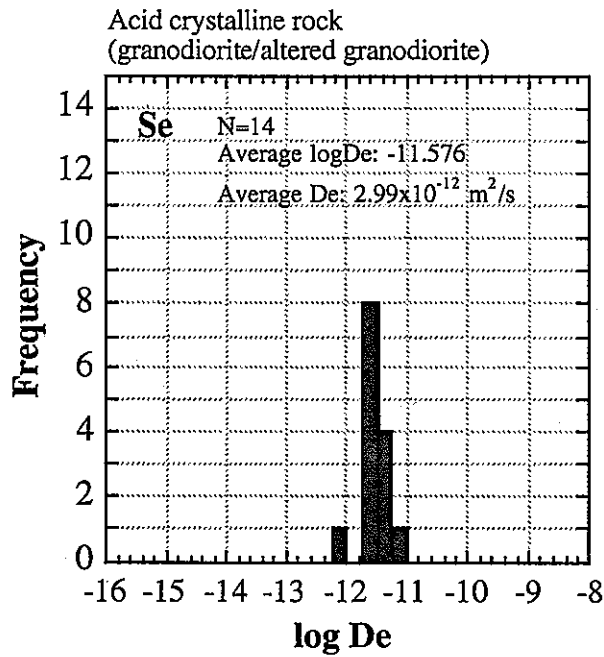
The De values of Se ( $\text{SeO}_3^{2-}$ ,  $\text{SeO}_4^{2-}$ ) in acid crystalline rock have been reported for granodiorite [Sato and Shibutani, 1996; Sato et al., 1997a, 1997b; PNC, 1996; Mukai et al., 1998], altered granodiorite [Sato and Shibutani, 1996; Sato et al., 1997a, 1997b; PNC, 1996] and fracture fillings [Sato et al., 1997a, 1997b]. The fracture fillings are also included in acid crystalline rock here as described in section 2.4.3(CI). Totally, 14 De and 16 Da data have been reported for Se. Among these, 6 De and 7 Da data for granodiorite, 5 De and 6 Da data for altered granodiorite, and 3 De and 3 Da data for fracture fillings have been reported. Since Se is sensitive to redox condition, the chemical species in solution and the valence change depending on redox condition. Selenium can chemically take Se(-II), (0), (II), (IV) and (VI), but since Se(II) is unstable, it does not normally exist as described in section 2.1.7(Se). The chemical behaviour of Se in solution and general sorptivity are as described in section 2.1.7(Se). Selenium is accounted as one of the important elements for performance assessment of the geological disposal of HLW. Under reducing conditions which is considered to be deep geological environment, the dominant species of Se is predicted to be  $\text{HSe}^-$  (hydroselenide). Also to FRHP and SRHP porewater in the second progress report [JNC, 1999], the dominant species of Se is  $\text{HSe}^-$ . The chemical behaviour and treatment of Se in performance assessment of the geological disposal of HLW are as described in section 2.1.7(Se).

The De data have been obtained by through-diffusion method and advection diffusion experiment, and Da values have been derived from De and rock capacity factor or from analysis by the time-lag method. The measurements have been carried out in a distilled water system [Mukai et al., 1998] and in a groundwater system [Sato and Shibutani, 1996; Sato et al., 1997a, 1997b; PNC, 1996]. The measurements have been carried out under anaerobic conditions [Sato and Shibutani, 1996; Sato et al., 1997a, 1997b; PNC, 1996] and under aerobic conditions [Mukai et al., 1998]. For the measurements conducted under anaerobic conditions, all experiments were performed in an  $\text{N}_2$ -atmosphere glove-box, in which the concentration of oxygen was kept less than 1ppm. Whilst, for that conducted under aerobic conditions, the experiment was carried out in the outside of the glove-box. In this case, the species of Se in the porewater is predicted to be  $\text{SeO}_3^{2-}$  (selenite) for measurements in the glove-box and  $\text{SeO}_4^{2-}$  (selenate) for that in the outside of the glove-box. All measurements have been carried out at 22 °C or room temperature. Besides, the measurements have been carried out in a pH range of 8.70 to 8.92.

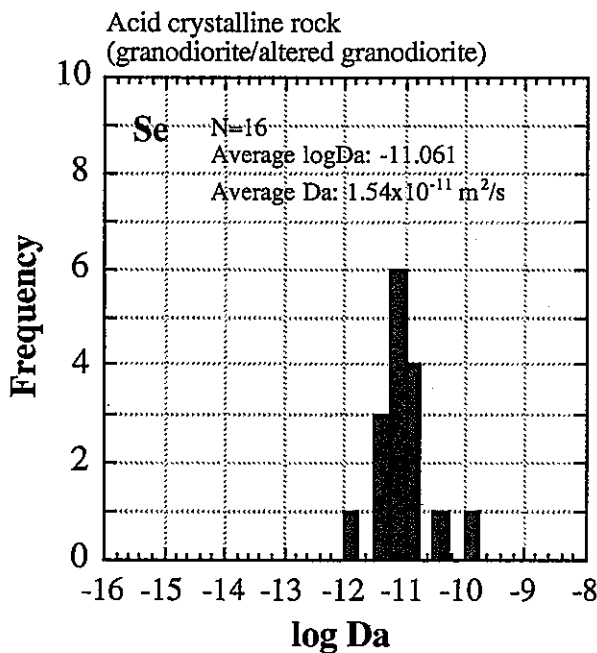
Figures 2.4.14-1 and 2.4.14-2 show histograms for De and Da values of Se in acid crystalline rock reported to date, respectively. For De, 14 data have been totally reported for  $\text{SeO}_3^{2-}$  so far. The width of variation in De is approximately one order of magnitude, being relatively small. The variation of porosity is also between 2.3 and 5.6 %, being not so large. Besides obtained conditions are also similar. Thus it is considered that those De data are similar. For Da, 15 and 1 data have been totally reported for  $\text{SeO}_3^{2-}$  and  $\text{SeO}_4^{2-}$ , respectively. For  $\text{SeO}_3^{2-}$ , all measurements have been carried out under anaerobic conditions, but for  $\text{SeO}_4^{2-}$ , the measurement has been performed under aerobic conditions. The width of variation in Da is over 2 orders of magnitude, being relatively wide. This may be because of the differences of porosity and redox condition.

Figures 2.4.14-3 and 2.4.14-4 show plots of De and Da values of  $\text{SeO}_3^{2-}$  as a function of rock porosity for acid crystalline rock. Since no datum for porosity has been reported for  $\text{SeO}_4^{2-}$ , all plots are for  $\text{SeO}_3^{2-}$ . Totally, 14 De and 15 Da data have been reported together with porosity. The De of  $\text{SeO}_3^{2-}$  shows a tendency to decrease with decreasing rock porosity. On the other hand, Da shows similar values between  $10^{-12}$  and  $10^{-11}$  m<sup>2</sup>/s, and porosity dependency is not found.

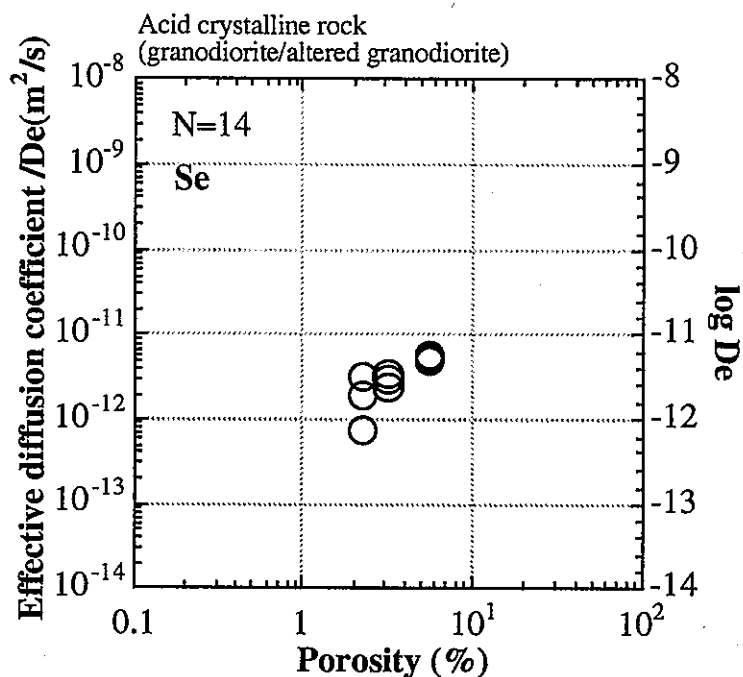
The Do values of  $\text{SeO}_3^{2-}$  and  $\text{SeO}_4^{2-}$  are estimated to be  $8.1 \times 10^{-10}$  and  $1.0 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C, respectively by the Nernst-Einstein equation [Marcus, 1997]. The FF and Gf were also estimated in the same way as section 2.1.7(Se). The discussion of these parameters will be done in detail in section 2.4.20.



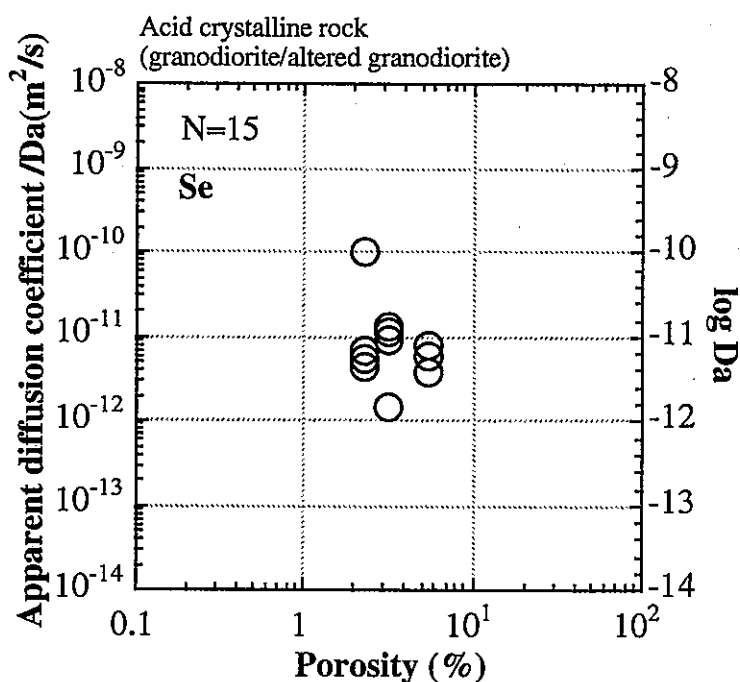
**Figure 2.4.14-1** Histogram for De values of  $\text{SeO}_3^{2-}$  ion in acid crystalline rock (granodiorite, altered granodiorite)  
The width of variation in De is approximately one order of magnitude, being small.



**Figure 2.4.14-2** Histogram for Da values of  $\text{SeO}_3^{2-}$  or  $\text{SeO}_4^{2-}$  ion in acid crystalline rock (granodiorite, altered granodiorite)  
The width of variation in Da is over 2 orders of magnitude, being relatively wide. This may be because of the differences of porosity and redox condition.



**Figure 2.4.14-3**  $D_e$  values of  $SeO_3^{2-}$  ion as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)  
The  $D_e$  shows a tendency to decrease with decreasing rock porosity between 2.3 and 5.6 %.



**Figure 2.4.14-4**  $D_a$  values of  $SeO_3^{2-}$  ion as a function of rock porosity for acid crystalline rock (granodiorite, altered granodiorite)  
Porosity dependency for  $D_a$  is not found for  $SeO_3^{2-}$ .



## References

- Japan Nuclear Cycle Development Institute (JNC) (1999): H12 Project to Establish Technical Basis for HLW Disposal in Japan, Supporting Report 3, Safety Assessment, JNC Technical Report, JNC TN1400 99-013.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.170.
- Mukai, S., Kitao, H. and Kataoka, S. (1998): Advection Diffusion Experiment Using Se and Cs in Fractured Rocks, 1998 Fall Meeting of the Atomic Energy Society of Japan, M25, p.842 (1998) [In Japanese].
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.205~207 [In Japanese].
- Sato, H. and Shibutani, T. (1996): Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste, PNC Technical Report, PNC TN1100 96-010, pp.139~144 [In Japanese].
- Sato, H., Shibutani, T., Tachi, Y., Ota, K., Amano, K. and Yui, M. (1997a): Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127.
- Sato, H., Tachi, Y., Shibutani, T. and Yui, M. (1997b): Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan, L12, p.561 [In Japanese].

### 2.4.15 Samarium (Sm)

The De values of Sm ( $\text{Sm}^{3+}$ ) in acid crystalline rock have been reported for granodiorite [Sato et al., 1993, 1997; PNC, 1994, 1996; Sato and Shibutani, 1994]. Totally, 5 De and 5 Da data have been reported for granodiorite. Samarium is one of the important elements for performance assessment of the geological disposal of HLW, and the chemical behaviour, decay property and treatment in performance assessment are described in section 2.1.8(Sm). Since Sm is not sensitive to redox condition, all measurements have been carried out under aerobic conditions.

In the second progress report, to FRHP (Fresh-Reducing-High pH) porewater,  $\text{Sm}(\text{CO}_3)_2^-$ ,  $\text{Sm}(\text{CO}_3)_3^{3-}$  and  $\text{SmCO}_3^+$  have been predicted to be predominant, and to SRHP (Saline-Reducing-High pH) porewater,  $\text{SmOH}^{2+}$ ,  $\text{SmCO}_3^+$  and  $\text{Sm}(\text{OH})_2^+$  have been predicted to be predominant [JNC, 1999]. To porewater due to reaction between concrete supports (low alkaline concrete) and FRHP groundwater, the same species as those to FRHP porewater have been predicted.

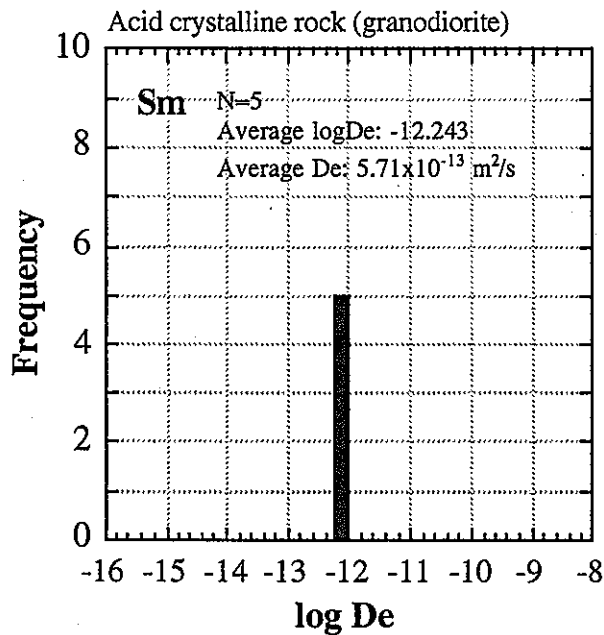
All De data have been obtained by through-diffusion method and Da values have been derived from De and rock capacity factor by the time-lag method. The measurements have been carried out in a distilled water system. The experiments have been carried out at 20°C [Sato et al., 1993; Sato and Shibutani, 1994; PNC, 1994] and 25 °C [PNC, 1996; Sato et al., 1997]. The reported porewater pH was between 5.8 and 6. The species Sm in the porewater in this pH condition is considered to be  $\text{Sm}^{3+}$ . In addition, a mixture solution of 0.1 M-CsCl,  $\text{NiCl}_2$  and  $\text{SmCl}_3$  was used as a tracer.

Figures 2.4.15-1 and 2.4.15-2 show histograms for De and Da values of  $\text{Sm}^{3+}$  in acid crystalline rock reported to date, respectively. For De, 5 data have been reported for  $\text{Sm}^{3+}$ . The width of variation in De is quite small. For Da, 5 data have been reported in total. Also for Da, no variation is found. This reason is considered to be because of a small porosity range (0.88 to 1.2 %).

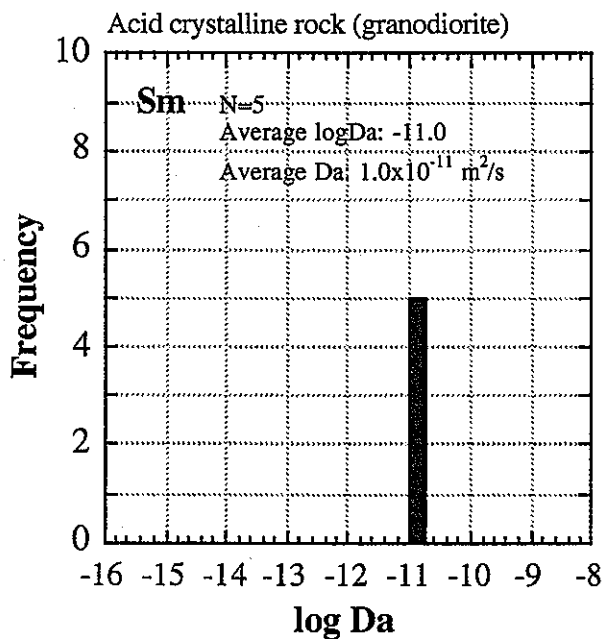
Figures 2.4.15-3 and 2.4.15-4 show plots of De and Da values of  $\text{Sm}^{3+}$  as a function of rock porosity for acid crystalline rock. Totally, 4 data have been reported together with porosity for De. No porosity dependency for De is found because the range of porosity is small to be 0.88 to 1.2 %. Also for Da, 4 data have been totally reported and no porosity dependency is found.

For Sm, De and Da data have been also reported for mudstone, which belongs to sedimentary rock (argillaceous/tuffaceous rock). The obtained porosity condition is also similar to that of granodiorite. Comparing with data for mudstone, both De and Da data for granodiorite are approximately one order of magnitude higher than those for mudstone.

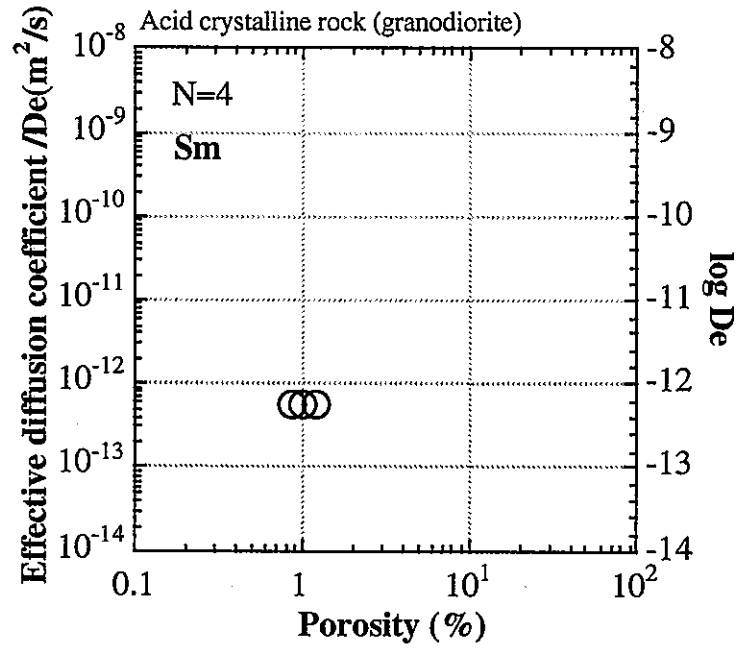
The Do of  $\text{Sm}^{3+}$  is  $6.1 \times 10^{-10} \text{ m}^2/\text{s}$  at 25 °C as described in section 2.1.8(Sm). The FF and Gf were also estimated in the same way as section 2.1.8(Sm). The discussion of these parameters will be done in section 2.4.20.



**Figure 2.4.15-1** Histogram for De values of  $\text{Sm}^{3+}$  ion in acid crystalline rock (granodiorite) The variation of De is quite small. This is considered to be because the variation of porosity is quite small.

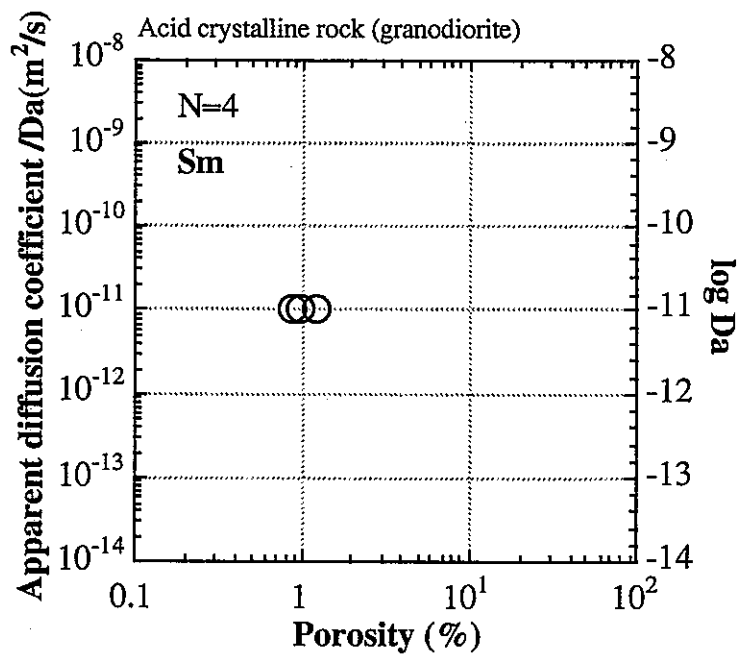


**Figure 2.4.15-2** Histogram for Da values of  $\text{Sm}^{3+}$  ion in acid crystalline rock (granodiorite) The variation of Da is also small similarly to that of De.



**Figure 2.4.15-3**  $D_e$  values of  $Sm^{3+}$  ion as a function of rock porosity for acid crystalline rock (granodiorite)

No porosity dependency for  $D_e$  is formed. This is considered to be because the range of porosity is small.



**Figure 2.4.15-4**  $D_a$  values of  $Sm^{3+}$  ion as a function of rock porosity for acid crystalline rock (granodiorite)

No porosity dependency is found for  $D_a$  similarly to that for  $D_e$ .

## References

- Japan Nuclear Cycle Development Institute (JNC) (1999): H12 Project to Establish Technical Basis for HLW Disposal in Japan, Supporting Report 3, Safety Assessment, JNC Technical Report, JNC TN1400 99-013.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.169.
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1994): Current Status for Research and Development of Geological Disposal -FY1993-, PNC Technical Report, PNC TN1410 94-094, pp.131~133 [In Japanese].
- Power Reactor and Nuclear Fuel Development Corporation (PNC) (1996): Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202~204 [In Japanese].
- Sato, H., Shibutani, T., Yoshikawa, H. and Yui, M. (1993): A Study on Diffusion Mechanism for Cs, Ni and Sm in Granodiorite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M29, p.667 [In Japanese].
- Sato, H. and Shibutani, T. (1994): Study on Adsorption and Diffusion of Nuclides in Buffer Material and Geosphere, PNC Technical Review, No.91, PNC TN8410 94-284, pp.71~89 [In Japanese].
- Sato, H., Shibutani, T. and Yui, M. (1997): Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119~133, Elsevier Science.

### 2.4.16 Strontium (Sr)

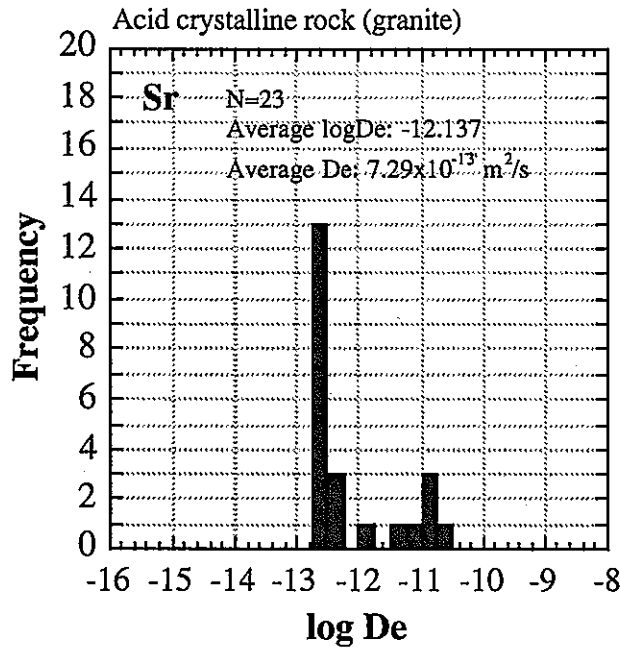
The  $D_e$  values for Sr ( $\text{Sr}^{2+}$ ) in acid crystalline rock have been reported for granite sampled from Inada, Ibaraki, Japan [Yamaguchi et al., 1992, 1993, 1997; Yamaguchi, 1997; Yamaguchi and Nakayam, 1997]. The  $D_a$  values have been also reported for granite [Yamaguchi et al., 1992, 1993, 1997; Yamaguchi and Nakayam, 1997; Idemitsu et al., 1992]. Although Sr-90 (decay property:  $\beta^-$ , half-life: 28.8 y) is initially much included as one of the fission products in vitrified waste, Sr is not included in important elements for performance assessment of the geological disposal of HLW, because its half-life, 28.8 y, is short. However, the chemical behaviour of Sr in solution is well familiar, and much reports and papers for sorption and diffusion have been reported so far. For matrix diffusion in acid crystalline rock, totally, 6 literatures have been reported listed up above. Strontium is not redox sensitive and takes  $\text{Sr}^{2+}$  in a wide pH range, but  $\text{SrSO}_4$  and  $\text{SrCO}_3$  are formed with increasing pH [Brookins, 1988]. Since Sr is one of the alkaline earth metals, it is considered that elements which belong to the same group such as  $\text{Be}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{Ba}^{2+}$  show similar behaviour in solution. It is presumed that  $\text{Co}^{2+}$  and  $\text{Ni}^{2+}$  also show similar diffusion behaviour. Though Sr is possible to receive the effect of porewater chemistry such as  $[\text{CO}_3^{2-}]$  and  $[\text{SO}_4^{2-}]$  [Brookins, 1988], it is not sensitive

to redox condition. Therefore, all measurements have been carried out under aerobic conditions. The  $D_e$  values have been obtained by through-diffusion method. Whilst,  $D_a$  values have been obtained by in-diffusion method. If rock capacity factor was also reported in experiments by the through-diffusion method,  $D_a$  was calculated. As porewater in the through-diffusion experiments, 0.1M KCl [Yamaguchi et al., 1992, 1993, 1997; Yamaguchi, 1996], deionized water [Yamaguchi et al., 1992, 1993; Yamaguchi, 1996] and groundwater [Yamaguchi and Nakayama, 1997] were used. On the other hand, deionized water was used as porewater in the in-diffusion experiments [Idemitsu et al., 1992]. All measurements for  $D_e$  have been performed at 25 °C or room temperature and those for  $D_a$  have been reported at 30 °C. The pH of the porewater was in a range of 4 to 5.5 for through-diffusion experiments, but it ranged from 4.3 to 9.4 for in-diffusion experiments. In through-diffusion experiments, a mixture solution of  $Ba(NO_3)_2$ ,  $BeSO_4$ ,  $CoCl_2$ ,  $MgCl_2$ ,  $NiCl_2$  and  $Sr(NO_3)_2$ , what is called a multi-tracer or a cocktail tracer, a  $Sr(NO_3)_2$  solution and a  $Sr^{12}$  solution were used as a tracer solution. All in-diffusion experiments for  $D_a$  have been carried out using a mixture solution of Cs-134, Sr-85 and Co-60.

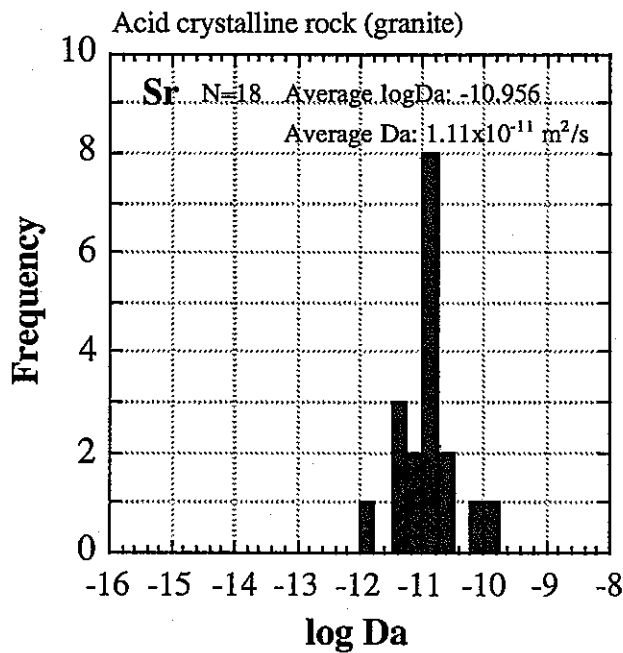
**Figures 2.4.16-1 and 2.4.16-2** show histograms for  $D_e$  and  $D_a$  values of  $Sr^{2+}$  in acid crystalline rock reported to date, respectively. Totally, 23  $D_e$  and 18  $D_a$  data have been reported for granite. The width of variation in  $D_e$  is two orders of magnitude, even though the rock porosities are approximately the same (0.4~0.7 %). This is presumed to be due to various pH of the solution and the ionic strength of the porewater. The width of variation in  $D_a$  is also approximately two orders of magnitude similarly to  $D_e$ . Since  $D_a$  also includes the effect of sorption, such wide variation is considered to be due to the change in sorption caused by various porewater conditions (pH, ionic strength).

**Figures 2.4.16-3 and 2.4.16-4** show plots of  $D_e$  and  $D_a$  values of  $Sr^{2+}$  ion as a function of rock porosity reported to date, respectively. Totally, 17  $D_e$  and 15  $D_a$  plots have been reported together with porosity. For  $D_e$ , since all porosities are similar, porosity dependency for  $D_e$  can not be discussed here. Also for  $D_a$ , porosity dependency can not be discussed from the same reason as  $D_e$ . However, the widths of variation in both diffusion coefficients are approximately 2 orders of magnitude.

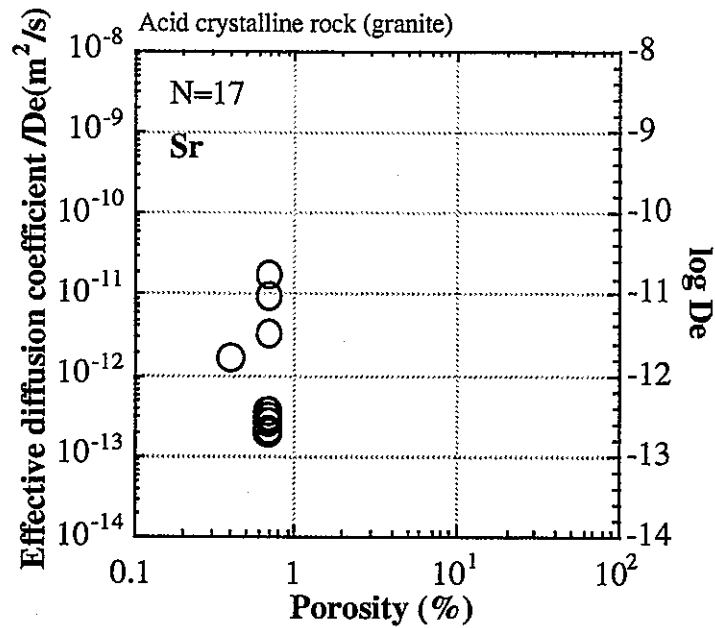
The  $D_o$  of  $Sr^{2+}$  is estimated to be  $7.9 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C [Marcus, 1997] by the Nernst-Einstein equation. The FF and Gf of granite were calculated in the same way as Ba, Be, Cl etc. Porosity dependencies for FF and Gf will be discussed in more detail in section 2.4.20.



**Figure 2.4.16-1** Histogram for De values of Sr<sup>2+</sup> ion in acid crystalline rock (granite)  
The width of variation in De is approximately 2 orders of magnitude.

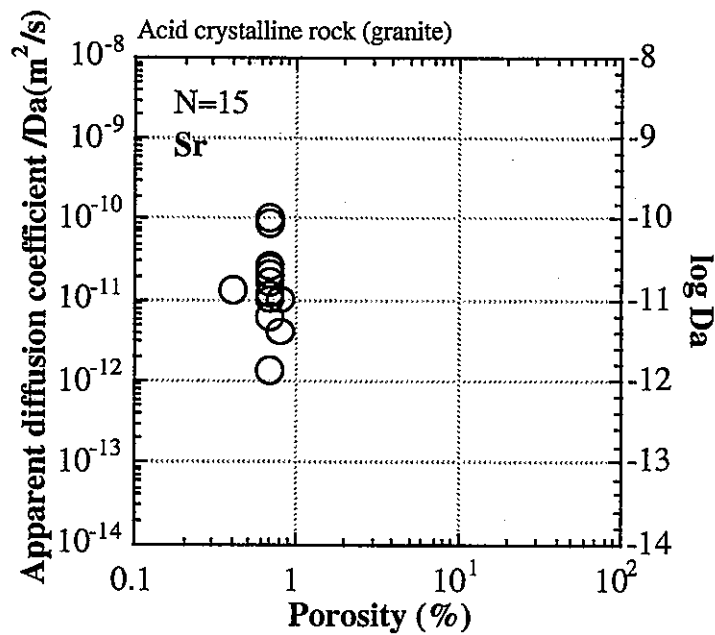


**Figure 2.4.16-2** Histogram for Da values of Sr<sup>2+</sup> ion in acid crystalline rock (granite)  
The width of variation in Da is also approximately 2 orders of magnitude.



**Figure 2.4.16-3** De values of Sr<sup>2+</sup> ion as a function of rock porosity for acid crystalline rock (granite)

Although porosity variation is narrow, the width of variation in De, 2 orders of magnitude, is wide.



**Figure 2.4.16-4** Da values of Sr<sup>2+</sup> ion as a function of rock porosity for acid crystalline rock (granite)

Similarly to De, the width of variation in Da is also approximately 2 orders of magnitude, even though the rock porosities are similar.



## Refernces

Idemitsu, K., Furuya, H., Hara, T. and Inagaki, Y. (1992): Migration of Cesium, Strontium and Cobalt in Water-Saturated Inada Granite, *Journal of Nuclear Science and Technology*, **29** (5), pp.454-460, the Atomic Energy Society of Japan.

Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1992): Surface Diffusion of Strontium in Rocks, 1992 Annual Meeting of the Atomic Energy Society of Japan, C19, p.119 [In Japanese].

Yamaguchi, T., Sakamoto, Y. and Senoo, M. (1993): Consideration on Effective Diffusivity of Strontium in Granite, *Journal of Nuclear Science and Technology*, **30** (8), pp.796~803, the Atomic Energy Society of Japan.

Yamaguchi, T. (1996): Diffusivity of Actinides in a Granite from Inada, Ibaraki, Japan, PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.156~160, Power Reactor and Nuclear Fuel Development Corporation [In Japanese].

Yamaguchi, T., Sakamoto, Y., Nakayama, S. and Vandergraaf, T. T. (1997): Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, *Journal of Contaminant Hydrology*, **26**, pp.109~117, Elsevier Science B. V.

Yamaguchi, T. and Nakayama, S. (1997): Effect of Sorption on Diffusivity in Micropores, 1997 Annual Meeting of the Atomic Energy Society of Japan, L10, p.559 [In Japanese].

### 2.4.17 Technetium (Tc)

The De values for Tc ( $\text{TcO}_4^-$ ) in acid crystalline rock have been reported for granite [PNC, 1991; Sato, 1991]. The Da has been also reported for granite [Sato, 1991]. Technetium, particularly, Tc-99 is accounted as one of the important elements for performance assessment of the geological disposal of HLW [JNC, 1999], because Tc-99, included in HLW as one of the fission products, has a long half-life of  $2.14 \times 10^5$  y (decay property:  $\beta^-$ ). Technetium is an artificial element which does not exist in nature and its stable isotope does not also exist. Technetium is redox sensitive and it is known that the valence depends on redox condition. Technetium can chemically take Tc(0), (II), (III), (IV), (VI)(?) and (VII), but generally takes Tc(VII) in a wide pH range in solution under aerobic conditions [Brookins, 1988]. Pertechnetate ( $\text{TcO}_4^-$ ) predominantly exists under aerobic conditions [Brookins, 1988]. In the Second Progress Report, to FRHP porewater,  $\text{TcO}(\text{OH})_2(\text{aq})$  has been predicted to be predominant, and also to SRHP porewater, the same species as that to FRHP has been predicted to be predominant [JNC, 1999].

All De data have been obtained by through-diffusion method and Da has been derived from De and rock capacity factor. All measurements have been carried out in a distilled water system and aerobic conditions. There is no datum obtained under reducing conditions. Therefore, the predominant species of Tc in distilled water under aerobic conditions is considered to be  $\text{TcO}_4^-$ . The measurements have been performed at 23 °C or room temperature. The reported porewater pH range is between 8.55 and 8.6.

Figures 2.4.17-1 and 2.4.17-2 show histograms for De and Da values of  $\text{TcO}_4^-$  in acid crystalline rock reported to date, respectively. For De, 2 data have been reported for  $\text{TcO}_4^-$ . For Da, only one datum has been reported.

Figures 2.4.17-3 and 2.4.17-4 show plots of De and Da values of  $\text{TcO}_4^-$  as a function of rock porosity for acid crystalline rock. Totally, only one De datum has been reported together with porosity and one Da datum has been reported. Originally, 2 De data have been reported together with porosity, but since rock porosity for one datum was too much high for common granite, the porosity was not adopted to plot.

The Do of  $\text{TcO}_4^-$  is estimated to be  $2.0 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C [Sato et al., 1996] by the Nernst-Einstein equation. Whilst, the ionic radius of pertechnetate ( $\text{TcO}_4^-$ ) is also reported to be 1.4 Å (model radius) [Marcus, 1997] and the Do of  $\text{TcO}_4^-$  is calculated to be  $1.8 \times 10^{-9}$  m<sup>2</sup>/s at 25 °C based on the Einstein-Stokes formula [e.g. Robinson and Stokes, 1959]. Here, the former value,  $2.0 \times 10^{-9}$  m<sup>2</sup>/s, was adopted for the calculations of FF and Gf, because the former was directly measured. The FF and Gf of granite were calculated in the same way as Ba, Be, Cl etc. Porosity dependencies for FF and Gf will be discussed in more detail in section 2.4.20.

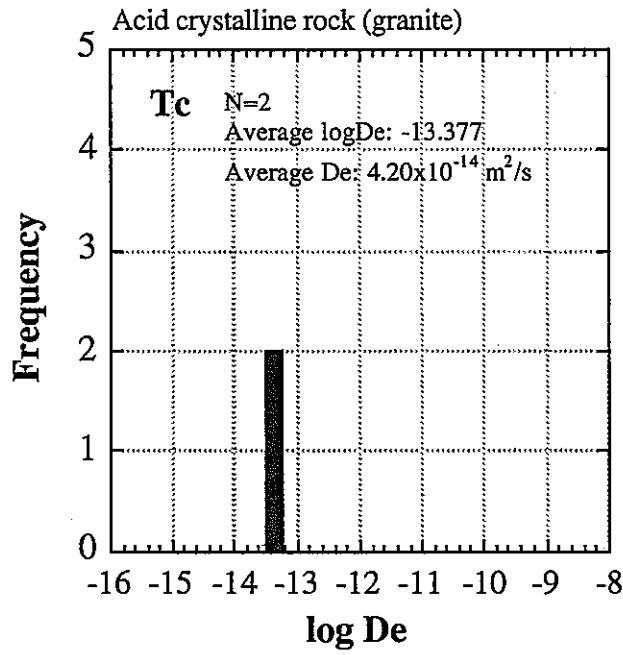


Figure 2.4.17-1 Histogram for De values of  $\text{TcO}_4^-$  ion in acid crystalline rock (granite)

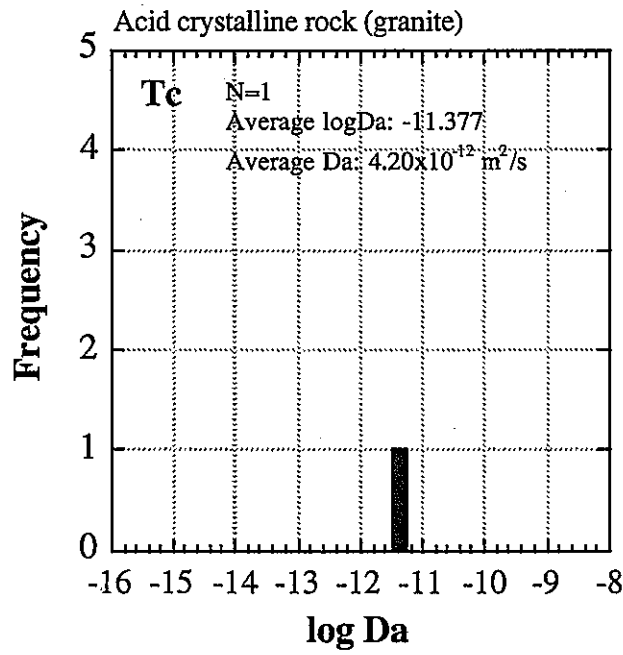


Figure 2.4.17-2 Histogram for Da values of  $\text{TcO}_4^-$  ion in acid crystalline rock (granite)

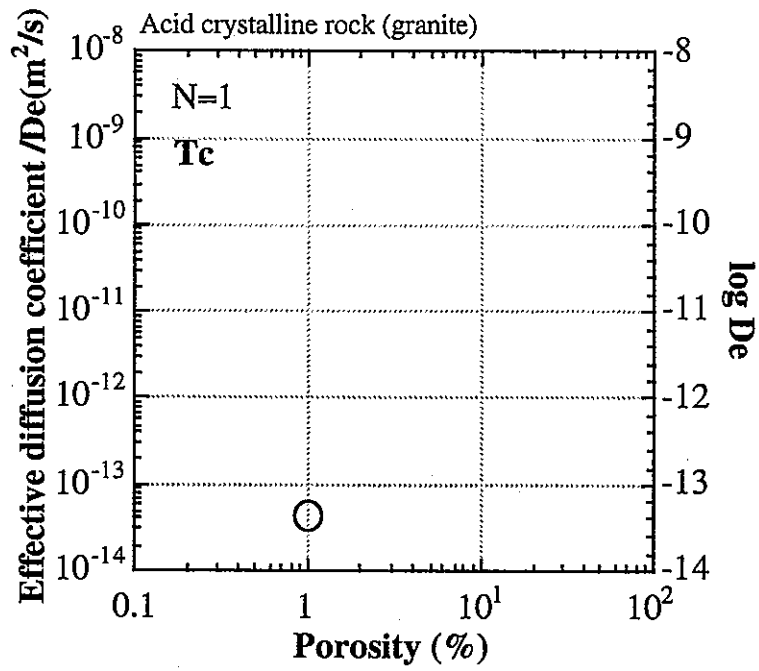


Figure 2.4.17-3 De of TcO<sub>4</sub><sup>-</sup> ion as a function of rock porosity for acid crystalline rock (granite)

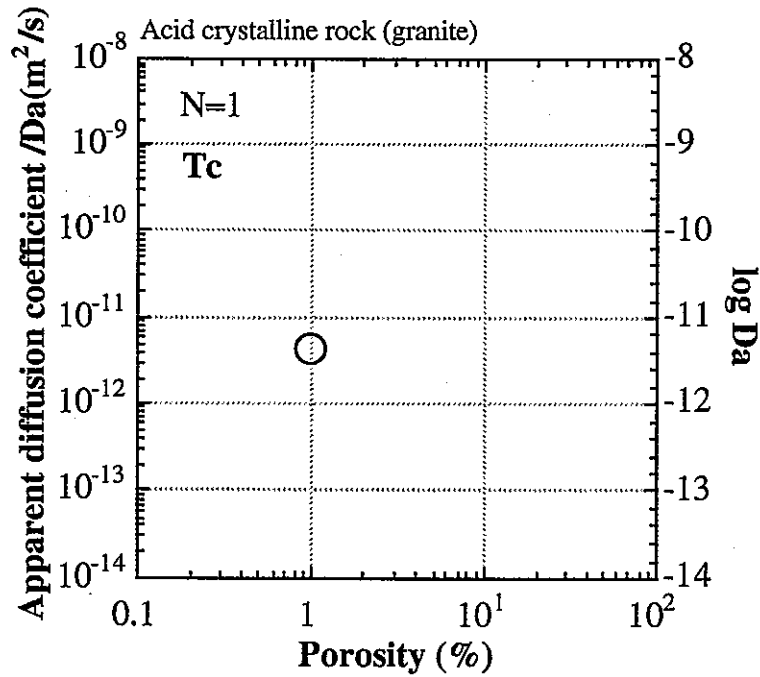


Figure 2.4.17-4 Da of TcO<sub>4</sub><sup>-</sup> ion as a function of rock porosity for acid crystalline rock (granite)

## References

- Brookins, D. G. (1988): pH-Eh Diagrams for Geochemistry, Springer-Verlag, Berlin, Germany, pp.97~99.
- Japan Nuclear Cycle Development Institute (JNC) (1999): H12 Project to Establish Technical Basis for HLW Disposal in Japan, Supporting Report 3, Safety Assessment, JNC Technical Report, JNC TN1400 99-013.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.183.
- Power Reactor and Nuclear Fuel Development Corporation (1991): Current Status for Research and Development of Geological Disposal -FY1989-, PNC Technical Report, PNC TN1410 91-009, pp.81~82 [In Japanese].
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Sato, H. (1991): PNC Technical Report "Report of Information Exchange Meeting on the Performance Assessment Research", PNC TN1410 91-079, pp.275~286, Power Reactor and Nuclear Fuel Development Corporation [In Japanese].
- Sato, H., Yui, M. and Yoshikawa, H. (1996): Ionic Diffusion Coefficients of  $\text{Cs}^+$ ,  $\text{Pb}^{2+}$ ,  $\text{Sm}^{3+}$ ,  $\text{SeO}_4^{2-}$  and  $\text{TcO}_4^-$  in Free Water Determined from Conductivity Measurements, Journal of Nuclear Science and Technology, **33** (12), pp.950~955.

### 2.4.18 Uranium (U)

The De values for U ( $\text{UO}_2^{2+}$ ,  $\text{UO}_2(\text{CO}_3)_3^{4-}$ ) in acid crystalline rock have been reported for granite [Yamaguchi et al., 1993; Yamaguchi, 1996] and biotitic granite [Yamaguchi et al., 1997]. The Da values for  $\text{UO}_2^{2+}$  have been also reported for both granite and biotitic granite [Yamaguchi et al., 1993, 1997; Idemitsu et al., 1992, 1993]. Totally, 9 De and 10 Da data have been reported so far. Uranium is accounted as one of the important elements for performance assessment of the geological disposal of HLW, particularly, U-233, 234, 235, 236 and 238 have been regarded as important nuclides. These decay properties and half-lives are as described in section 2.1.9(U). These nuclides are all produced as decay chain nuclides. Uranium is sensitive to redox condition and can chemically take U(III), (IV), (V) and (VI) in solution, but U(VI) and U(IV) are predominant in solution under aerobic and reducing conditions, respectively. The chemical species also depends on the porewater pH and the concentration of carbonate in the porewater. In the presence of carbonate in the porewater,  $\text{UO}_2^{2+}$ ,  $\text{UO}_2\text{CO}_3^0$ ,  $\text{UO}_2(\text{CO}_3)_2^{2-}$  and  $\text{UO}_2(\text{CO}_3)_3^{4-}$  are predominant in the pH ranges,  $\leq$  around 4, between around 4 and 6, between around 6 and 9, and  $\geq$  9, respectively [Brookins, 1988] under aerobic conditions. Under reducing conditions,  $\text{USiO}_4 \leq$  around pH7 and  $\text{U}(\text{OH})_5^- \geq$  around pH7 are predominant in the presence of Si [Brookins, 1988]. Under the borders of pH for each species are variable depending on the concentrations of U and carbonate and on competing ion. To

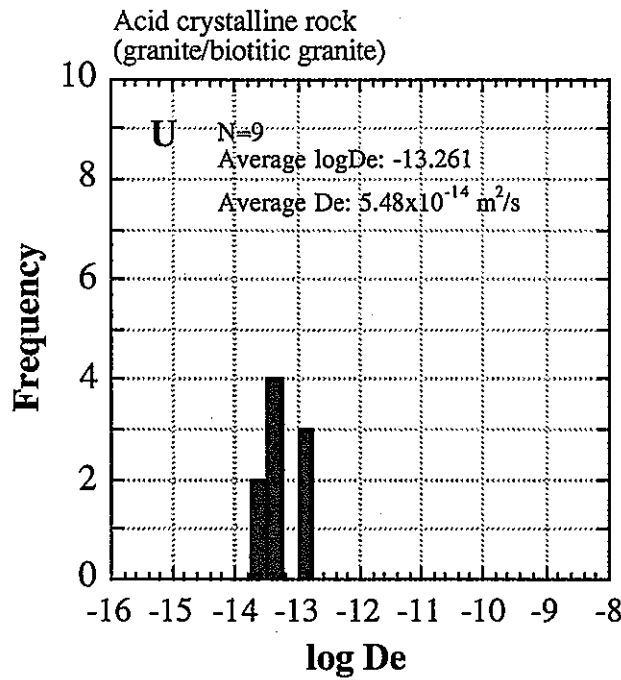
FRHP porewater, in the Second Progress Report,  $U(OH)_2(CO_3)_2^{2-}$  and  $U(OH)_4(aq)$  have been predicted to be predominant [JNC, 1999]. Whilst, to SRHP porewater,  $U(OH)_4(aq)$  has been predicted to be predominant [JNC, 1999].

The  $D_e$  values have been obtained by through-diffusion method and  $D_a$  values have been obtained by both through-diffusion and in-diffusion (back-to-back) methods, and have been derived from  $D_e$  and rock capacity factor or derived from analysis by the time-lag method for some data. The through-diffusion measurements have been carried out in a 0.1M KCl solution system [Yamaguchi et al., 1993, 1997] and in a mixture system of  $NaHCO_3$  and  $NaNO_2$  [Yamaguchi, 1996]. All in-diffusion measurements for  $D_a$  have been carried out in a deionized water system [Idemitsu et al., 1992, 1993]. All measurements for both through-diffusion and in-diffusion experiments have been carried out at 20~30 °C or at room temperature under aerobic conditions. In this case, the valence of U is considered to be hexavalent. The porewater pH was 4, 9.3 and 5 for the KCl,  $NaHCO_3$ - $NaNO_2$  and deionized water systems, respectively.

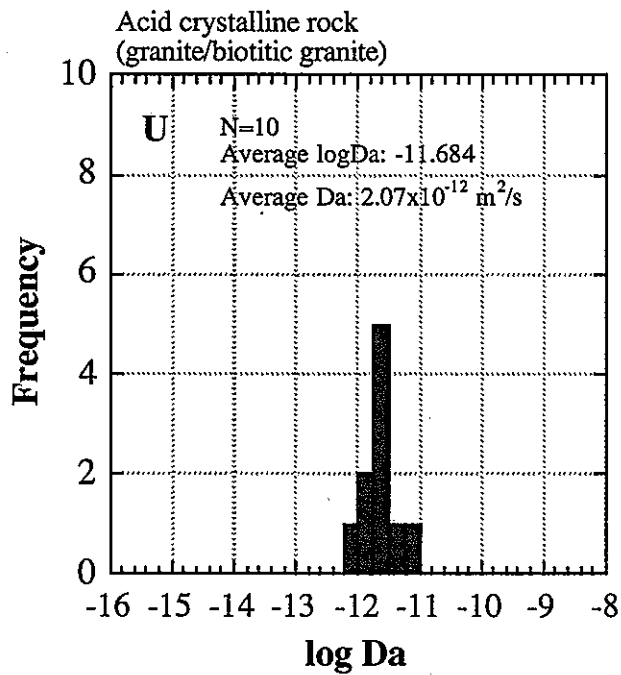
**Figures 2.4.18-1 and 2.4.18-2** show histograms for  $D_e$  and  $D_a$  values of  $UO_2(CO_3)_3^{4-}$  and  $UO_2^{2+}$ , and  $UO_2^{2+}$  in acid crystalline rock reported to date, respectively. The widths of variation in  $D_e$  and  $D_a$  are approximately one order of magnitude, being not so large, although obtained chemical condition is different.

**Figures 2.4.18-3 and 2.4.18-4** show plots of  $D_e$  and  $D_a$  values of  $UO_2(CO_3)_3^{4-}$  and  $UO_2^{2+}$ , and  $UO_2^{2+}$  as a function of rock porosity for acid crystalline rock reported to date, respectively. Totally, 6  $D_e$  and 10  $D_a$  data have been reported together with porosity. Since the widths of variation in porosities are quite small (0.7~0.8 %), porosity dependency for  $D_e$  and  $D_a$  can not be discussed.

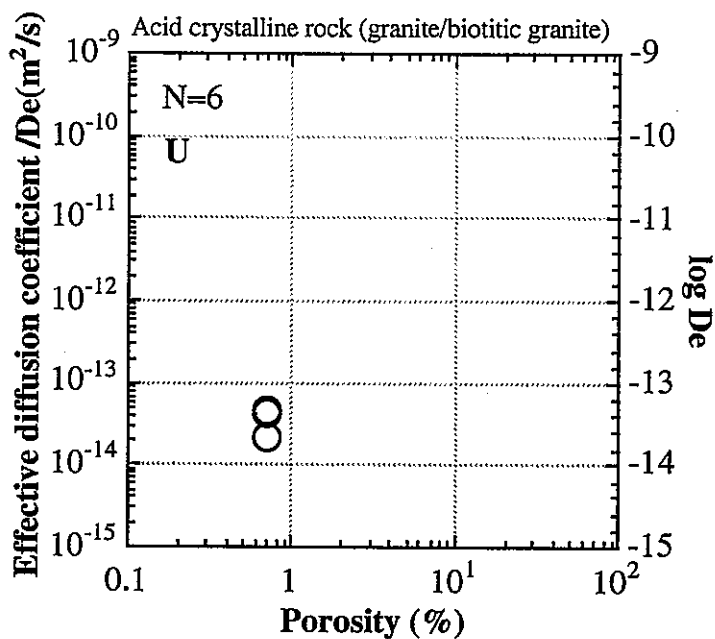
The  $D_o$  of  $UO_2^{2+}$  is estimated to be  $4.3 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C [Marcus, 1997]. However, the  $D_o$  of  $UO_2(CO_3)_3^{4-}$  has not been directly measured to date. The  $D_o$  of this species, estimated to be  $7.2 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C based on the Stokes equation [Robinson and Stokes, 1959] by the ionic radius calculated from the molecular structure [Yamaguchi, 1996], was adopted. The FF and Gf values of granite and biotitic granite were also calculated based on these  $D_o$  values. Porosity dependencies for FF and Gf will be discussed in section 2.4.20.



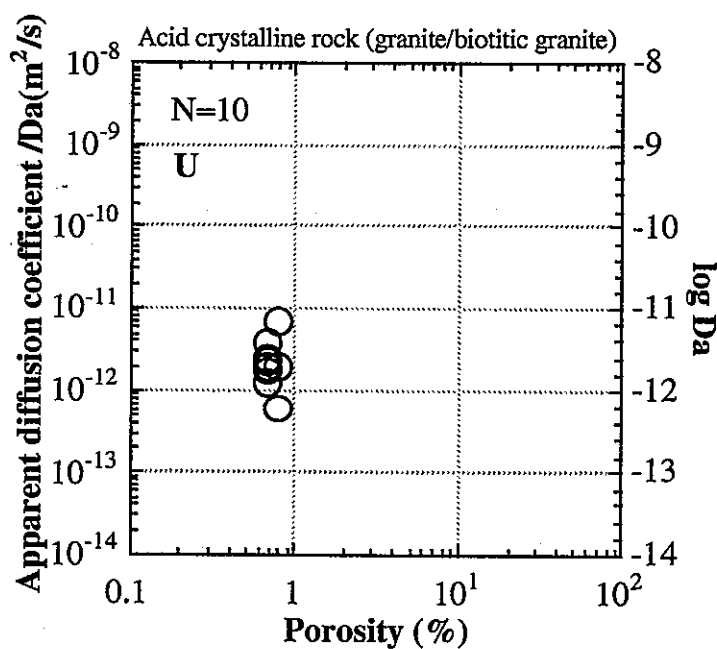
**Figure 2.4.18-1** Histogram for De values of  $\text{UO}_2^{2+}$  and  $\text{UO}_2(\text{CO}_3)_3^{4-}$  ions in acid crystalline rock (granite, biotitic granite)  
The width of variation in De of U is relatively small to be  $10^{-14} \text{ m}^2/\text{s}$  order.



**Figure 2.4.18-2** Histogram for Da values of  $\text{UO}_2^{2+}$  ion in acid crystalline rock (granite, biotitic granite)  
The width of variation in Da is also small approximately one order of magnitude.



**Figure 2.4.18-3** De values of  $\text{UO}_2^{2+}$  and  $\text{UO}_2(\text{CO}_3)_3^{4-}$  ions as a function of rock porosity for acid crystalline rock (granite, biotitic granite)  
 The variation in De is small, although chemical species is different.



**Figure 2.4.18-4** Da values of  $\text{UO}_2^{2+}$  ion as a function of rock porosity for acid crystalline rock (granite, biotitic granite)  
 The variation in Da is also not so large, being approximately one order of magnitude. This is presumed to be due to the effect of porewater pH and the porewater chemistry.



## References

- Brookins, D. G. (1988): pH-Eh Diagrams for Geochemistry, Springer-Verlag, Berlin, Germany, pp.151~157.
- Japan Nuclear Cycle Development Institute (JNC) (1999): H12 Project to Establish Technical Basis for HLW Disposal in Japan, Supporting Report 3, Safety Assessment, JNC Technical Report, JNC TN1400 99-013.
- Idemitsu, K., Furuya, H., Murayama, K. and Inagaki, Y. (1992): Diffusivity of Uranium(VI) in Water-Saturated Inada Granite, Scientific Basis for Nuclear Waste Management XV (Sombret, C. G. ed.), Materials Research Society Symposium Proceedings, Vol.257, pp.625~632, Materials Research Society, Pittsburgh, Pennsylvania.
- Idemitsu, K., Furuya, H., Murayama, K., Asao, H. and Inagaki, Y. (1993): Primary Diffusion Path of Uranium(VI) in Water-Saturated Inada Granite, Proceedings of the International Conference on Nuclear Waste Management and Environmental Remediation (Alexandra, D., Baker, R. and Kohout, R. eds.), the American Society of Mechanical Engineers, pp.207~212.
- Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U.S.A., p.169.
- Robinson, R. A. and Stokes, R. H. (1959): Electrolyte Solutions, 2nd ed., Butterworths, London, UK.
- Yamaguchi, T., Sakamoto, Y. and Sekine, K. (1993): Effective Diffusivity of  $\text{UO}_2^{2+}$  in Granite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M28, p.666.
- Yamaguchi, T. (1996): Diffusivity of Actinides in a Granite from Inada, Ibaraki, Japan, PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.156~160, Power Reactor and Nuclear Fuel Development Corporation [In Japanese].
- Yamaguchi, T., Sakamoto, Y., Nakayama, S. and Vandergraaf, T. T. (1997): Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, Journal of Contaminant Hydrology, 26, pp.109~117, Elsevier Science B. V.

## 2.4.19 Uranine (Sodium Fluorescein: $C_{20}H_{10}O_5Na_2$ )

The  $D_e$  values for uranine ( $C_{20}H_{10}O_5Na_2$ : Sodium fluorescein) in acid crystalline rock have been reported for granite [Kumata et al., 1990; Yamaguchi et al., 1997]. The  $D_a$  values have been also reported for the same rock. Totally, 6 data have been reported for both parameters so far. Different from specific element and species described so far, uranine is one of the hydrocarbons. Uranine is not accounted as one of the important elements for performance assessment of the geological disposal of HLW. Uranine is an aromatic organic compound,  $C_{20}H_{10}O_5Na_2$ , with a molecular weight of 376.28 and its molecules are only slightly hydrated in water, because they are large size and low charged [Yamaguchi et al., 1997]. Besides, the radius of uranine molecule in water is estimated to be 0.5~0.6 nm based on the C-C bonding length (0.139 nm). Therefore, uranine was used as a non-sorbing tracer.

The  $D_e$  values have been all obtained by through-diffusion method and  $D_a$  values have been also obtained analytically from the same method. The details of porewater chemistry is not reported for all cases, but distilled water has been used in some cases reported [Kumata et al., 1990]. All measurements have been carried out at room temperature under aerobic conditions. No information of porewater pH has been reported.

**Figures 2.4.19-1 and 2.4.19-2** show histograms for  $D_e$  and  $D_a$  values of uranine in acid crystalline rock reported to date, respectively. Since the width of variation in porosity (0.7~0.74 %) is very small, the width of variation in  $D_e$  also seems to be small. Although  $D_a$  includes sorption property, since uranine is non-sorbing, the width of variation also seems to be small.

**Figures 2.4.19-3 and 2.4.19-4** show plots of  $D_e$  and  $D_a$  values of uranine as a function of rock porosity. As described above, since there is no variation in porosity, porosity dependencies for  $D_e$  and  $D_a$  can not be here discussed.

The  $D_o$  of uranine, not directly measured, has been estimated to be  $4.5 \times 10^{-10}$  m<sup>2</sup>/s at 25 °C based on the Hyduk-Landie equation [Skagius and Neretnieks, 1985]. The same  $D_o$  has been estimated based on the Stokes equation by the ionic radius calculated from the molecular structure [Yamaguchi et al., 1997]. The FF and Gf values of granite were also calculated based on this  $D_o$ . Porosity dependencies for FF and Gf will be discussed in section 2.4.20.

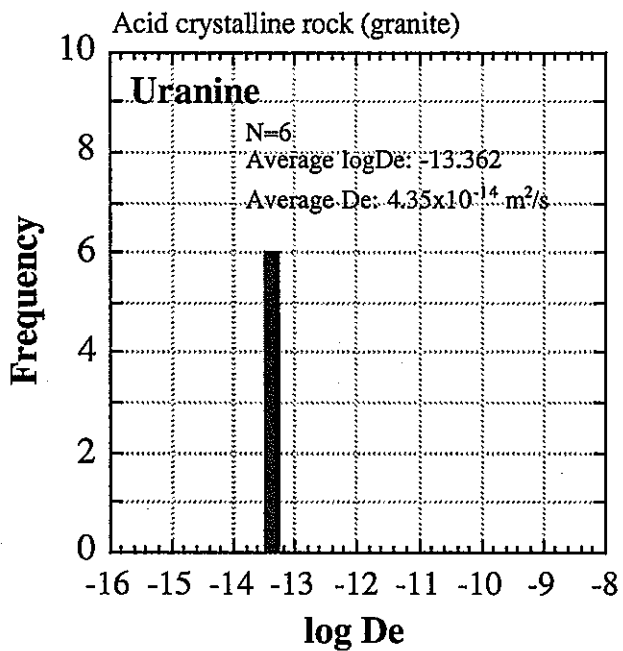


Figure 2.4.19-1 Histogram for De values of uranine in acid crystalline rock (granite)

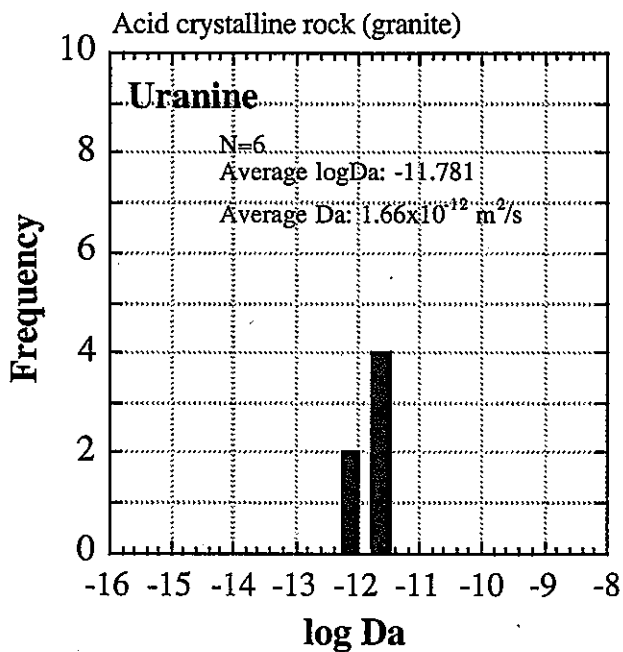


Figure 2.4.19-2 Histogram for Da values of uranine in acid crystalline rock (granite)

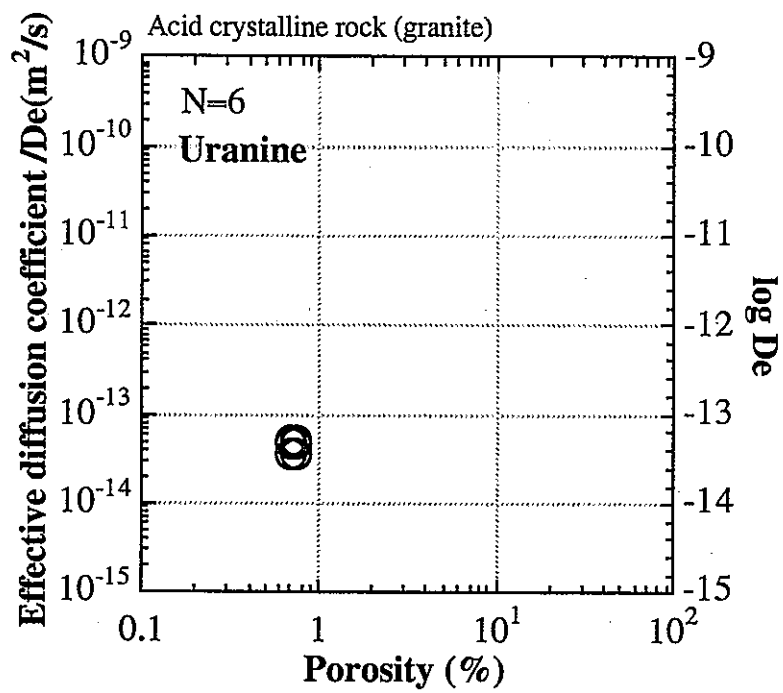


Figure 2.4.18-3  $D_e$  values of uranine as a function of rock porosity for acid crystalline rock (granite)

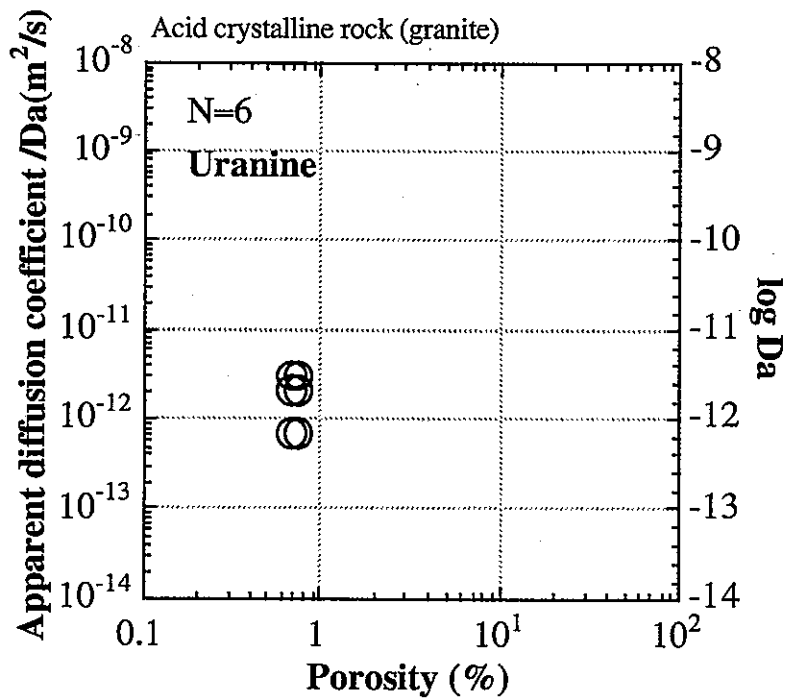


Figure 2.4.18-4  $D_a$  values of uranine as a function of rock porosity for acid crystalline rock (granite)

## References

- Kumata, M., Iwai, T., Sagawa, T., Suzuki, T. and Nishiyama, K. (1990): Diffusion Experiment of a Radionuclide in Granite Rock Cores, JAERI Report, JAERI-M 90-179 [In Japanese].
- Skagius, K. and Neretnieks, I. (1985): Properties and Diffusivities of Some Non-sorbing Species in Crystalline Rocks, SKB 85-03.
- Yamaguchi, T., Sakamoto, Y., Nakayama, S. and Vandergraaf, T. T. (1997): Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, *Journal of Contaminant Hydrology*, **26**, pp.109~117, Elsevier Science B. V.

### 2.4.20 Summary for diffusivity data in acid crystalline rock

Totally, 18 elements and one tracer (hydrocarbon) were discussed in sections 2.4.1~2.4.19. Figure 2.4.20-1 shows a histogram of De values in acid crystalline rock for 18 elements and one hydrocarbon reported in each section. Although the average value of De for all elements and one hydrocarbon in acid crystalline rock was estimated to be  $1.18 \times 10^{-12} \text{ m}^2/\text{s}$ , relatively a wide range of variation is found in De. The width of variation in De is over 3 orders of magnitude. This is considered to attribute to the variation of porosity, but the variation in porosity is not so large, being in a range of 0.4 to 5.6 %. Even though so, the width of variation in De is quite large, compared with that in sedimentary rock. This is considered to be due to porosity and difference in pore structure such as tortuosity.

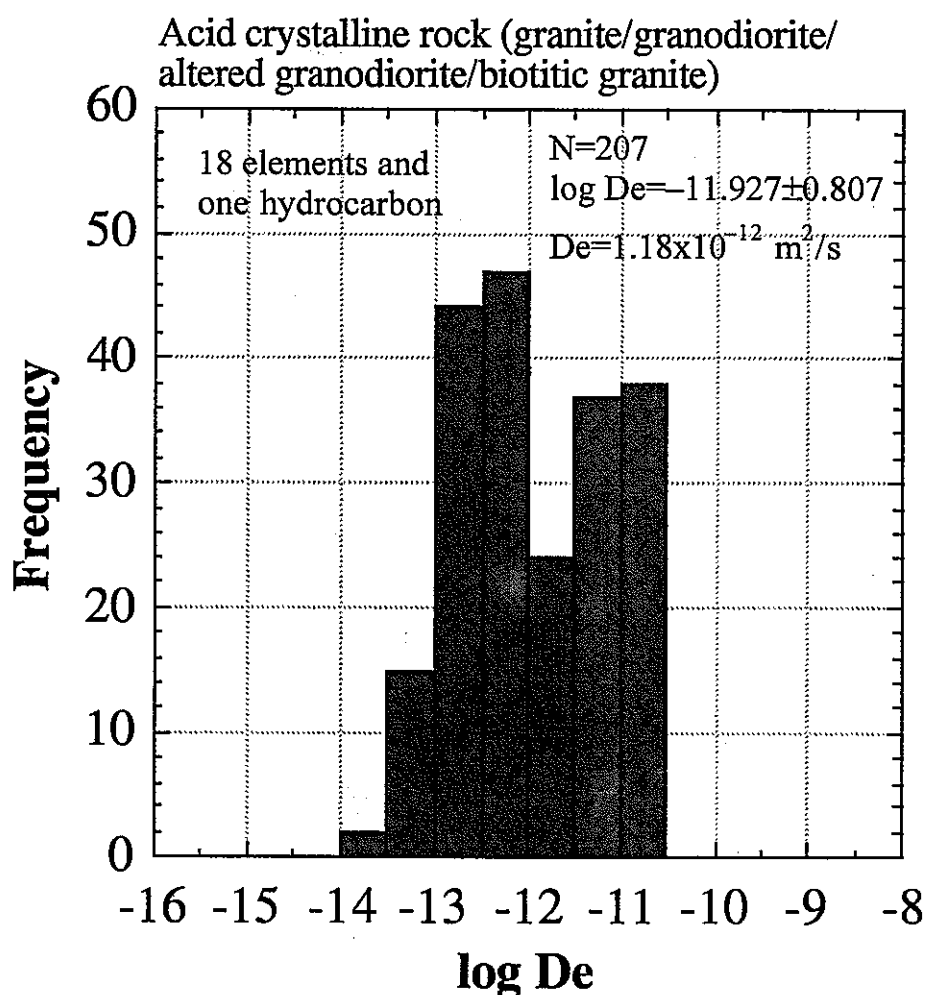


Figure 2.4.20-1 Histogram for De values in acid crystalline rock for 18 elements and one hydrocarbon (granite, granodiorite, biotitic granite)

The average value of De for all elements and one hydrocarbon in acid crystalline rock was estimated to be  $1.18 \times 10^{-12} \text{ m}^2/\text{s}$ . Since the differences in porosity and pore structure are not considered in the histogram, relatively a wide range of variation is found in De.

Figure 2.4.20-2 shows a histogram of Da values in acid crystalline rock for 13 elements and one hydrocarbon (no Da has been reported for Ba, Be, K Mg and Pu) treated in sections 2.4.1~2.4.19. The average value of Da in acid crystalline rock was estimated to be  $1.26 \times 10^{-11} \text{ m}^2/\text{s}$  and a variation of 3 orders of magnitude was found in Da. The Da also includes sorption onto minerals composing rock, so the difference in Da can not be simply explained by porosity and pore structure. The Da is affected by both sorption and De. Since De depends on the pore structure of rock and sorption also depends on the kind of rock, it is presumed that the variation of Da was caused by the differences of constituent minerals and pore structure by the difference of the kind of rock.

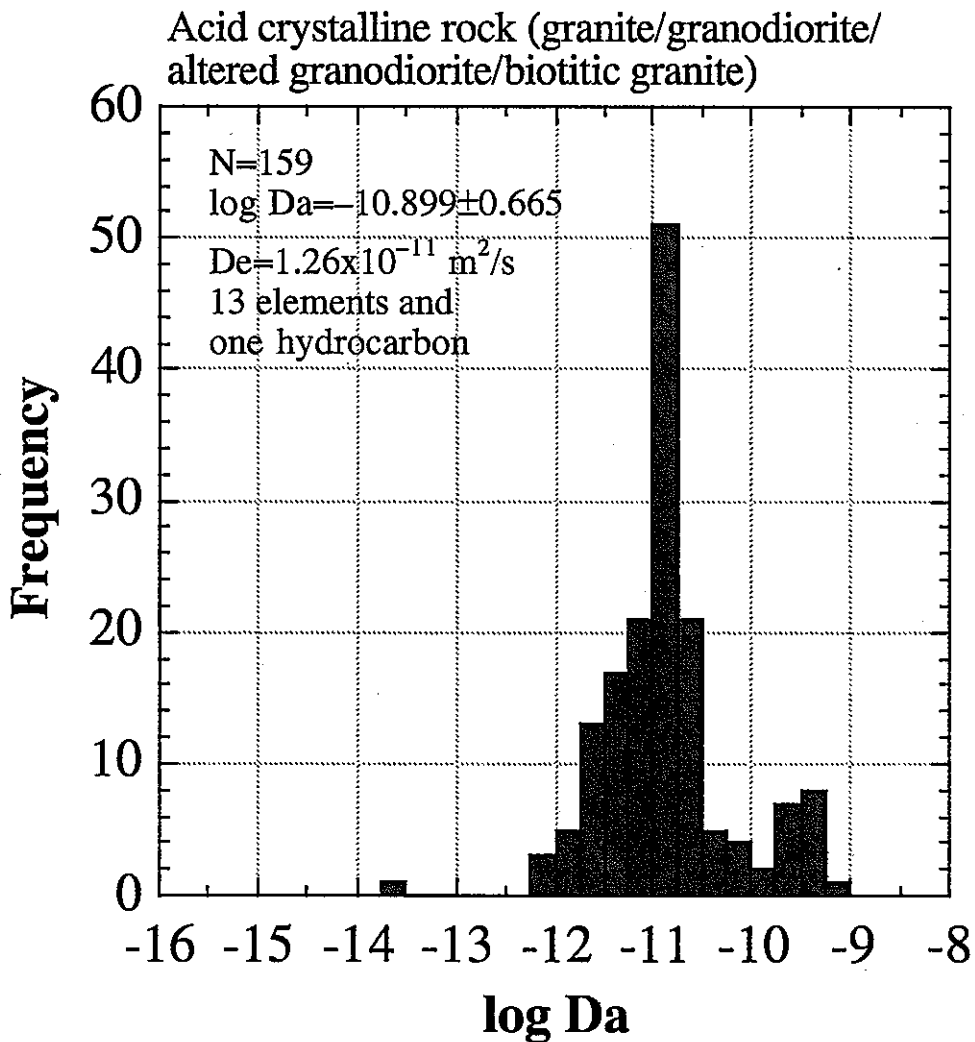


Figure 2.4.20-2 Histogram for Da values in acid crystalline rock (granite, granodiorite, biotitic granite)

The average value of Da in acid crystalline rock was estimated to be  $1.26 \times 10^{-11} \text{ m}^2/\text{s}$ . The width of variation in Da is quite large.

Figure 2.4.20-3 shows a porosity dependency of  $D_e$  values for 17 elements and one hydrocarbon (uranine) discussed in sections 2.4.1~2.4.19. Totally, 188 plots have been reported for  $D_e$  together with porosity. The  $D_e$  shows a tendency to increase with increasing rock porosity. However, the width of variation in  $D_e$  is quite large than that of sedimentary rock. No significant tendency by the difference of the kind of element on  $D_e$  is also particularly found. It is considered that  $D_e$  is strongly affected by pore structure.

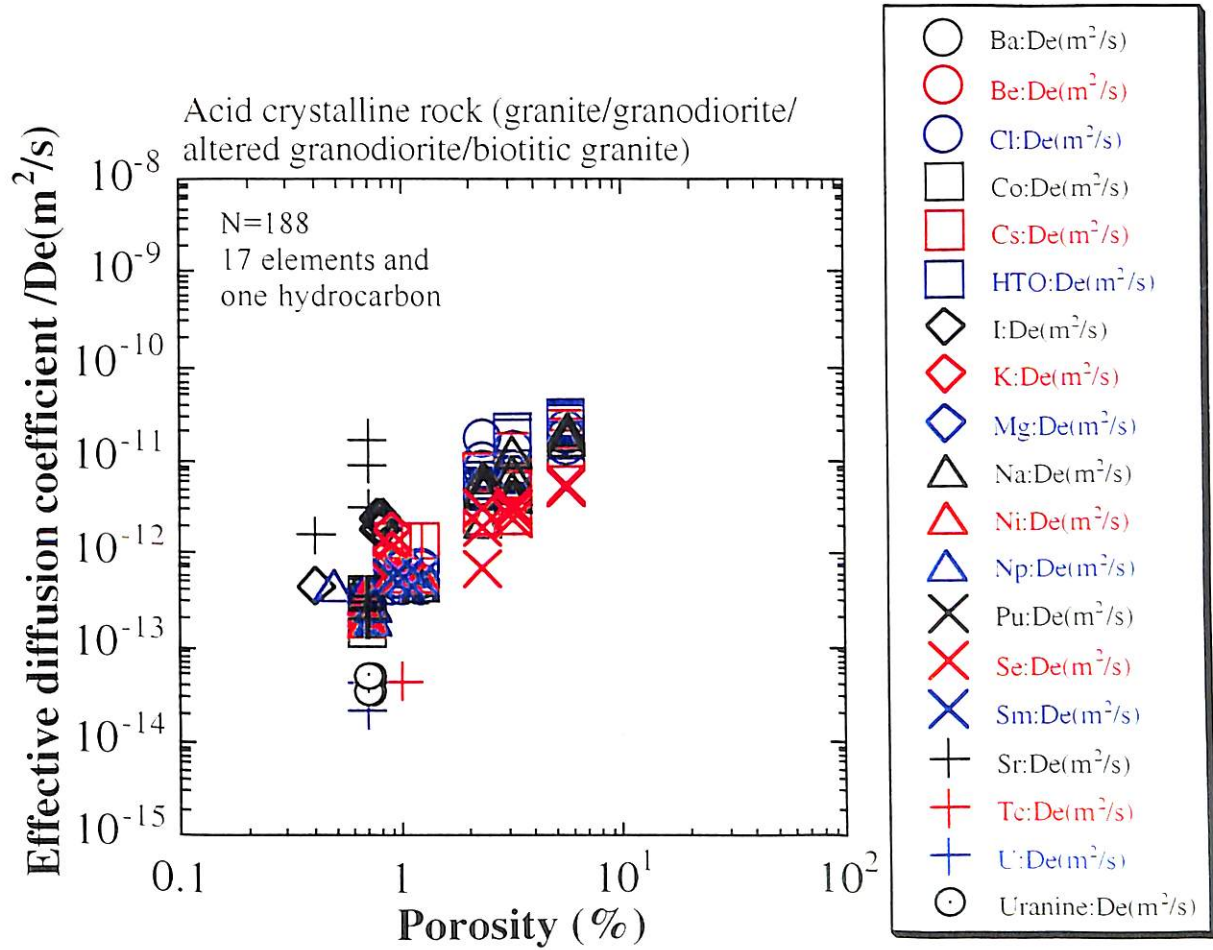


Figure 2.4.20-3  $D_e$  values for 17 elements and one tracer (hydrocarbon) as a function of rock porosity for acid crystalline rock (granite, granodiorite, biotitic granite)



Figure 2.4.20-4 shows a least squares fitting curve for  $D_e$  values as a function of porosity. In the derivation of empirical equation,  $D_e$  data for all elements (17 elements and one hydrocarbon) were taken into account without distinguishing the kind of element. From the fitting, the empirical equation:  $D_e = (10^{-12.20 \pm 0.033}) \cdot \phi^{2.0 \pm 0.090}$  was derived, where  $\phi$  is the porosity in % and the errors stand for the standard deviations.

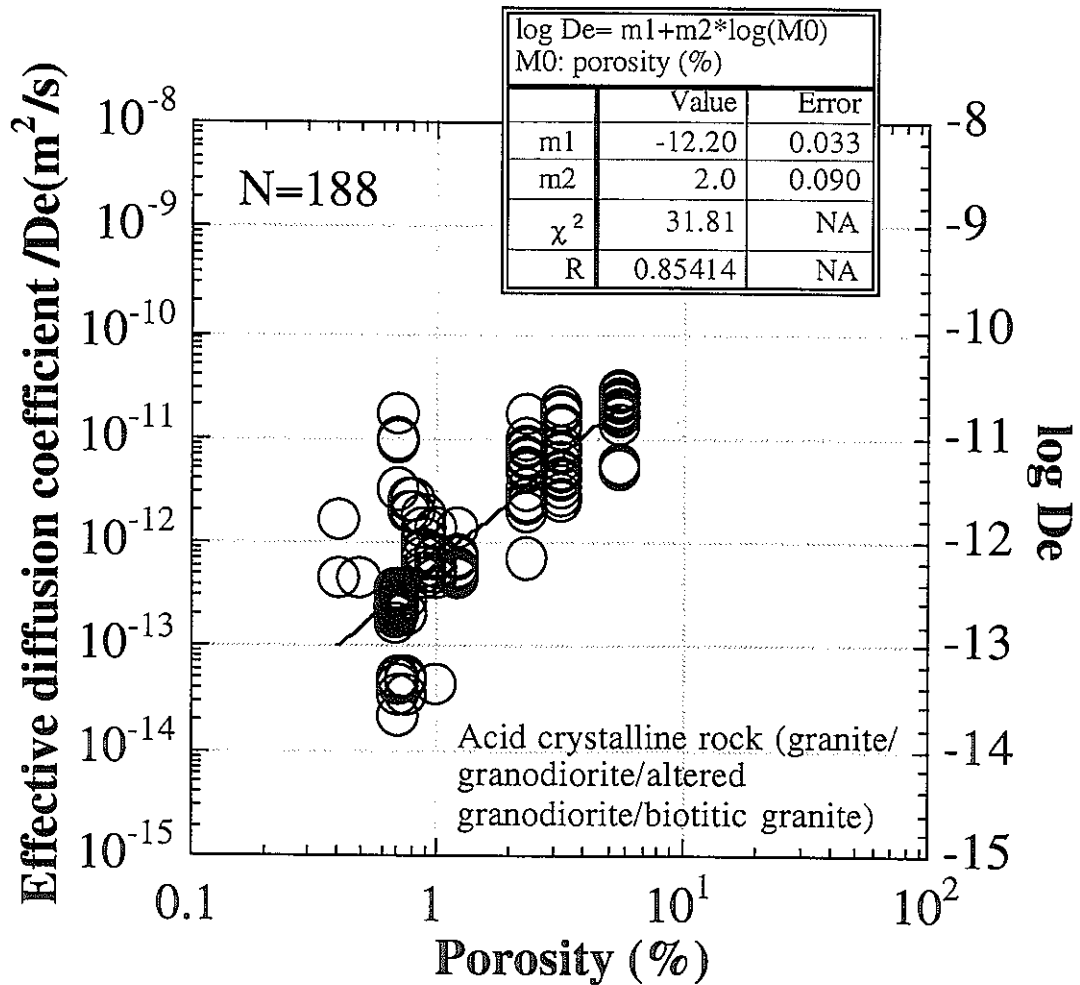


Figure 2.4.20-4 Empirical equation for  $D_e$  values as a function of rock porosity for acid crystalline rock (granite, granodiorite, biotitic granite)

Figure 2.4.20-5 shows a width of variation in  $De$  values as a function of rock porosity. Two lines were drawn to be parallel to the empirical equation of  $De$  values as a function of rock porosity, which is shown in Figure 2.4.20-4 and to pass on the maximum and minimum values of plotted  $De$  values. Although some plots are out of the lines, those data are all conservative in performance assessment. The width of variation in  $De$  is approximately 2 orders of magnitude for the same porosity. The lower and lower lines do not have statistical meaning, merely showing measured maximum and minimum values.

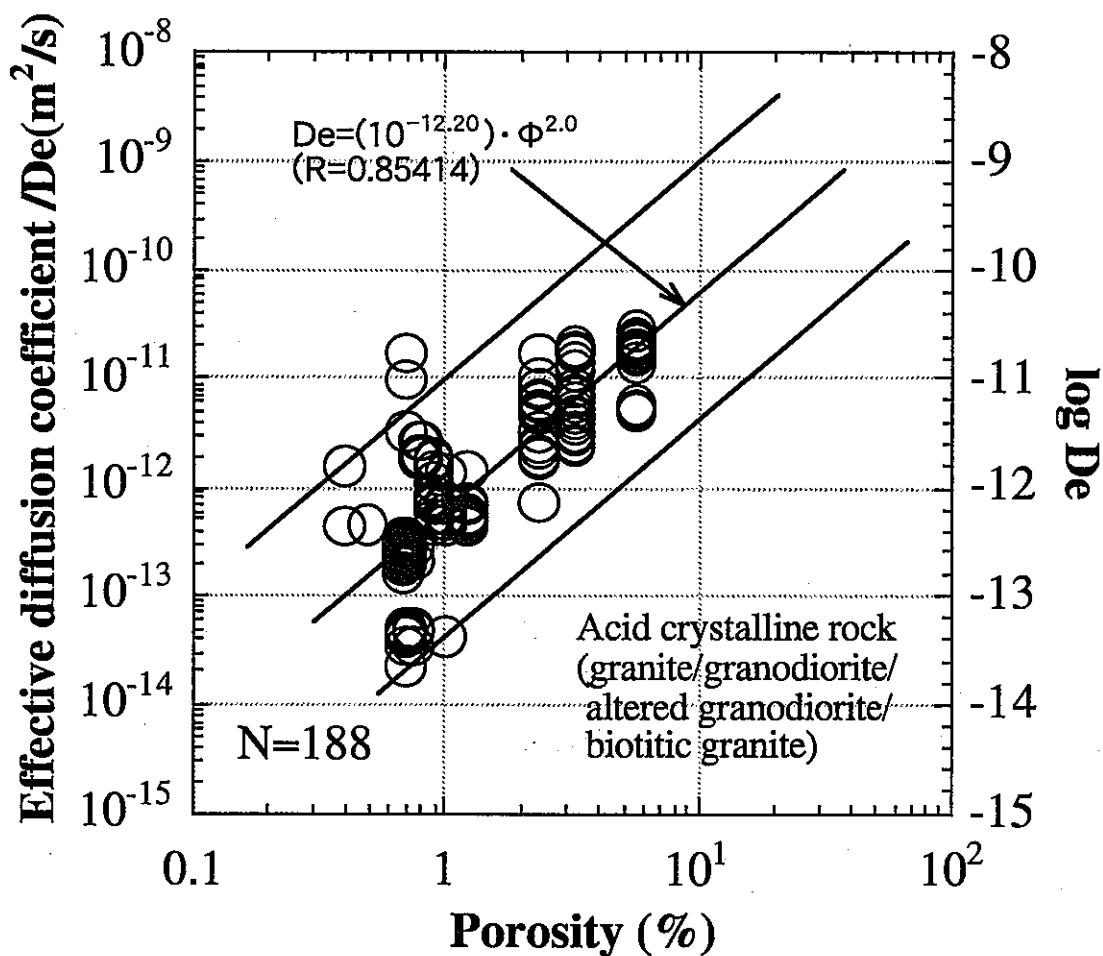


Figure 2.4.20-5 A width of variation in  $De$  values as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite)

2.4.20-6 shows a least squares fitting curve for Da values as a function of rock porosity. Although Da shows a tendency to increase with increasing rock porosity, clear trend is not found. The width of variation in Da is quite large. This is considered to be due to the effect of rock porosity and pore structural factor by the difference of the kind of rock and the effect of sorption by the differences of the kinds of rock and element. For Da, a width of variation of approximately 3 orders of magnitude is in maximum found for the same porosity.

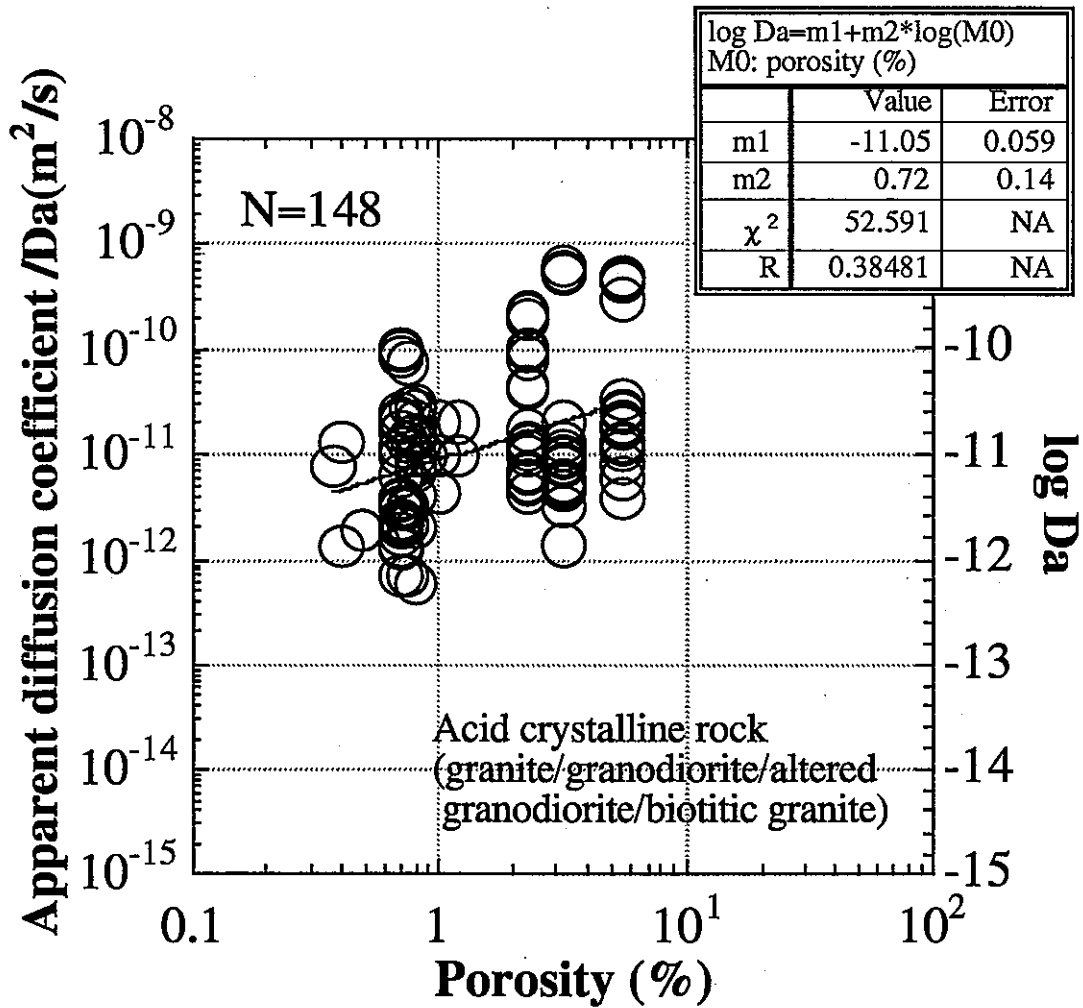


Figure 2.4.20-6 Da values as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite)

Figure 2.4.20-7 shows a correlation of  $D_e$  values as a function of charge number of species. No clearly systematic trend is particularly found. The width of variation in  $D_e$  is approximately 3 orders of magnitude, being very large, although the charge of species is the same. Since porosity and pore structural factors actually depend on the kind of rock and  $D_0$  is also different depending on the kind of species, it is presumed that such a wide range of variation in  $D_e$  was caused by the differences of their factors. However, it seems that the higher the absolute value of the charge of species becomes, the lower  $D_0$  becomes.

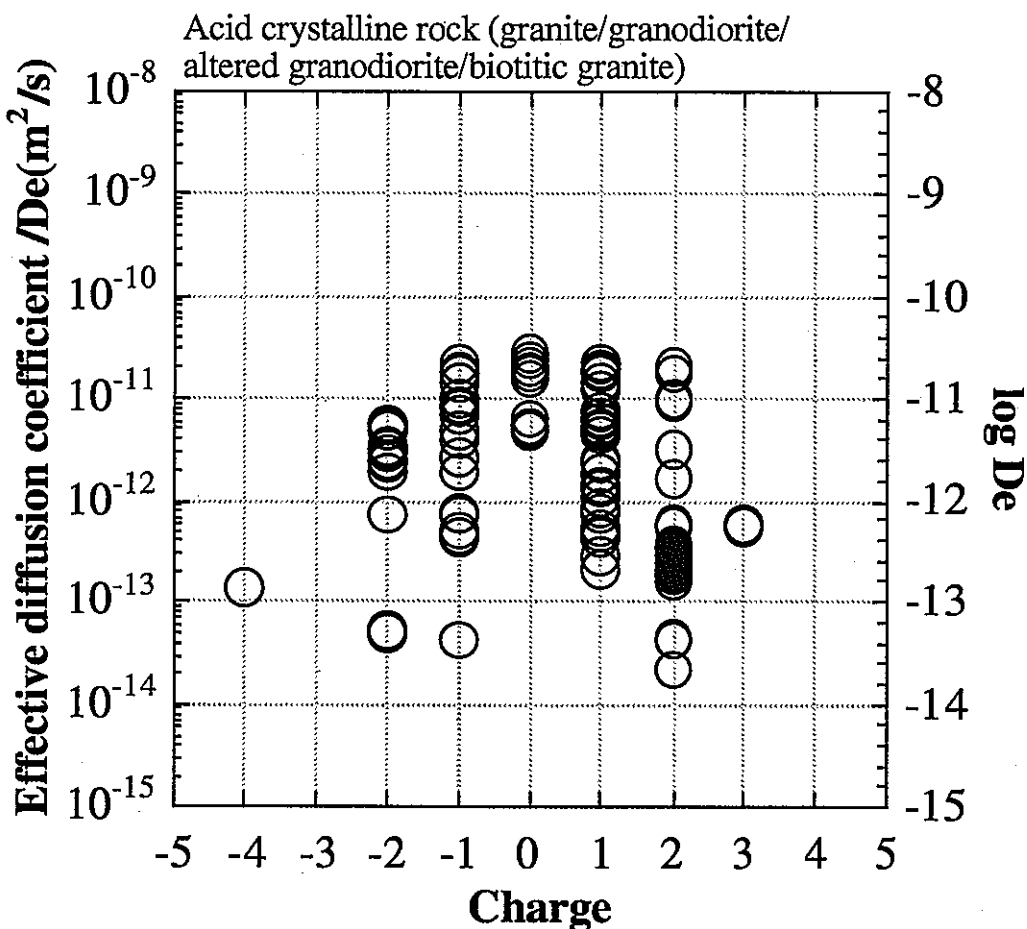


Figure 2.4.20-7  $D_e$  values as a function of charge number of species  
The width of variation in  $D_e$  is approximately 3 orders of magnitude, being large, although the charge number of species is the same. This would be not only because of the variation of porosity, but also because of the difference in  $D_0$  for each species.

Figure 2.4.20-8 shows a correlation of formation factors (FF) as a function of rock porosity. Similarly to De values, FF also shows a tendency to increase with increasing rock porosity. However, relatively a wide range of variation is also found in FF, although rock porosity is the same. This may be due to the differences of tortuosity by the differences between mineral grain sizes composing individual rocks.

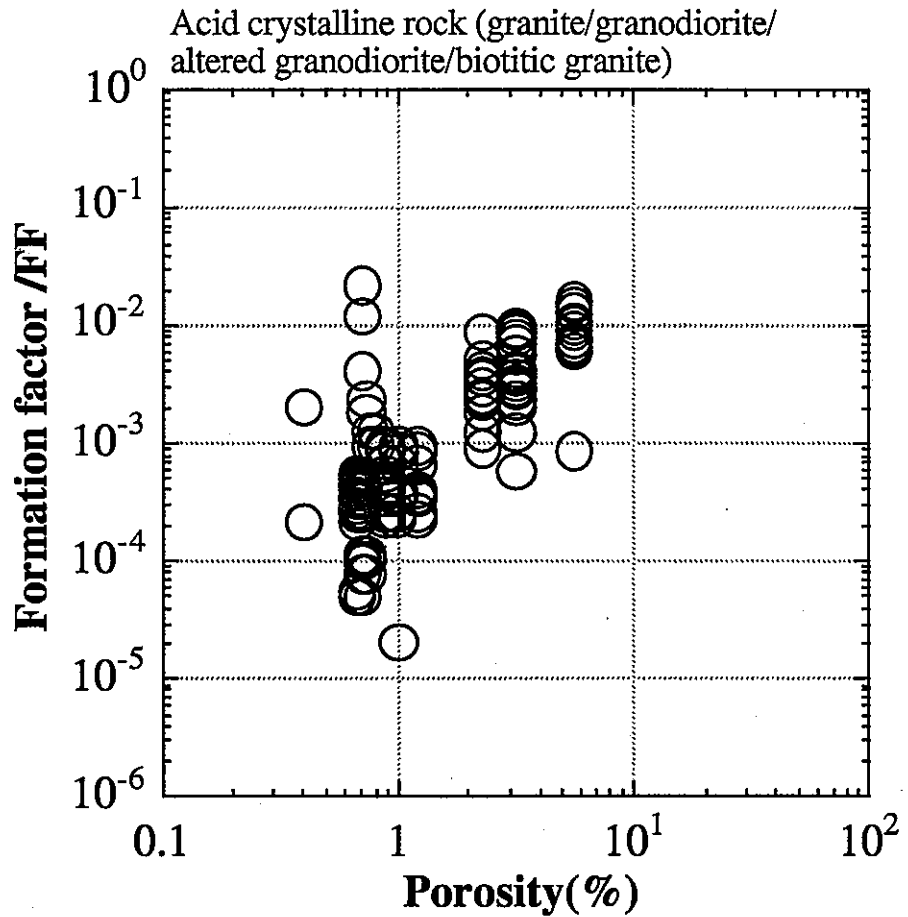


Figure 2.4.20-8 FF values as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite)  
Relatively a wide range of variation in FF is found even though it is the same porosity.

Figure 2.4.20-9 shows a correlation of geometric factors (Gf) as a function of rock porosity. As shown in Figure 2.4.20-9, no significant porosity dependency is found for Gf. This is considered to ascribe to the grain sizes of minerals composing the rock.

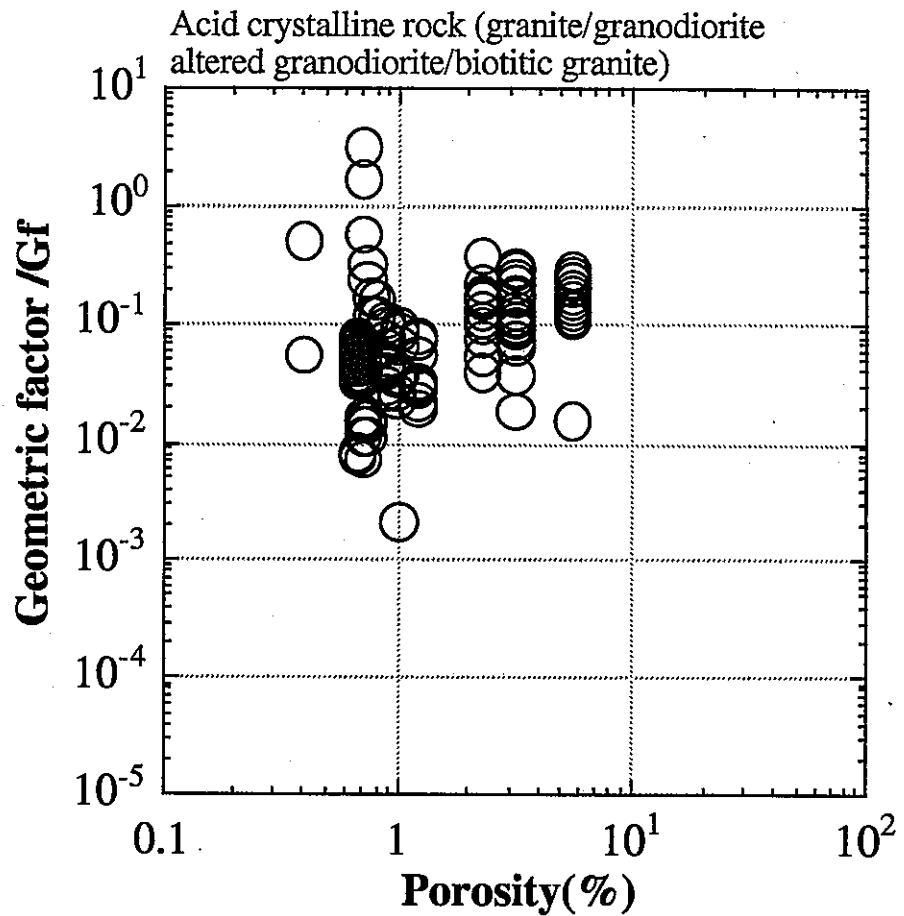


Figure 2.4.20-9 Gf values as a function of rock porosity for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite)  
No porosity dependency is found for Gf.

Figure 2.4.20-10 shows a plot of De values for individual elements in acid crystalline rock. No tendency between De values and elements is particularly found. Even the same element has a wide range of De values (for example, Cl, Na, Sr). However, the variation in rock porosity is also included in the variation of De. Furthermore, the tortuosity of rock is also different depending on the kind of rock, because the grain sizes are different depending on the kind of rock. Among these elements, a wide range of variation is found particularly for De values of Cl, Na and Sr. This may be due to the variation of porosity.

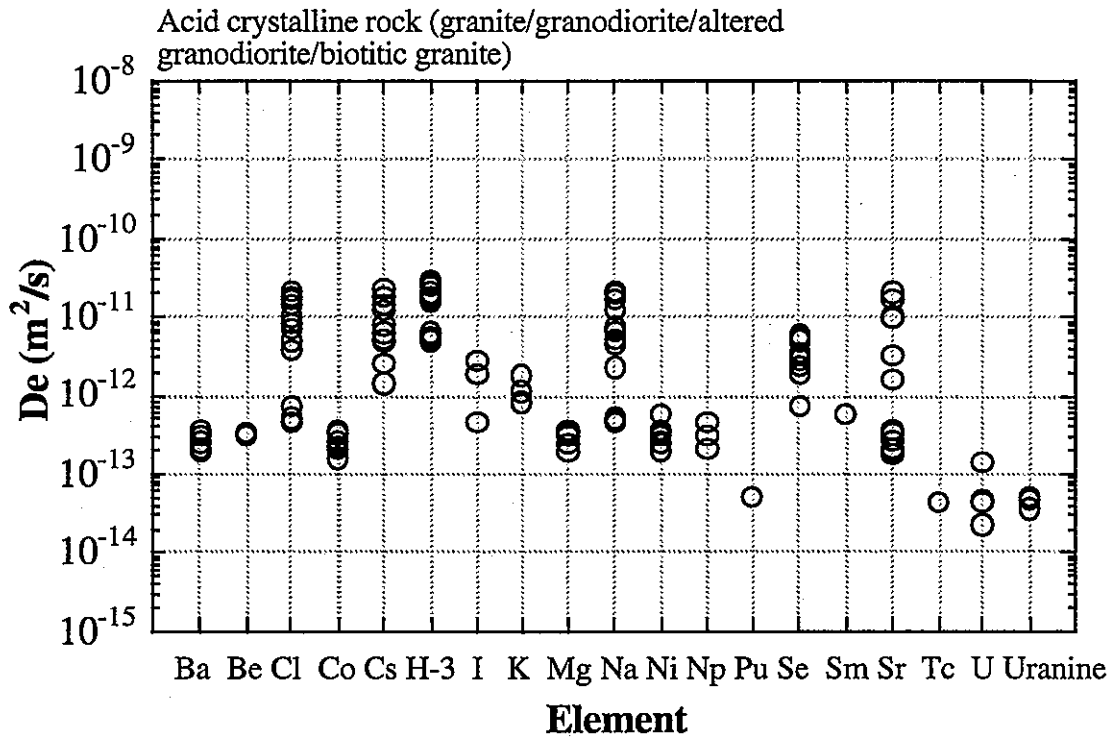


Figure 2.4.20-10 A plot of De values for individual elements in acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite)

No particular tendency between De values and individual elements is found, showing a wide range of De values even though the same element.

Figure 2.4.20-11 shows a correlation between  $D_e$  values and  $D_o$  values for acid crystalline rock. It seems that  $D_e$  also increases with increasing  $D_o$ . However, no clear correlation is found. A wide range of variation in  $D_e$  is found from around  $D_o=8 \times 10^{-10}$  to  $2 \times 10^{-10}$  m<sup>2</sup>/s (25 °C). This reason may be because the width of variation in rock porosity is large and pore structural factors are also different depending on the rock porosity.

The elements which take near  $D_o=2 \times 10^{-9}$  m<sup>2</sup>/s (25 °C) are Cl, Cs and I. This wide variation in  $D_e$  may be due to the differences of rock porosity and of pore structural factors by the difference of the kind of rock.

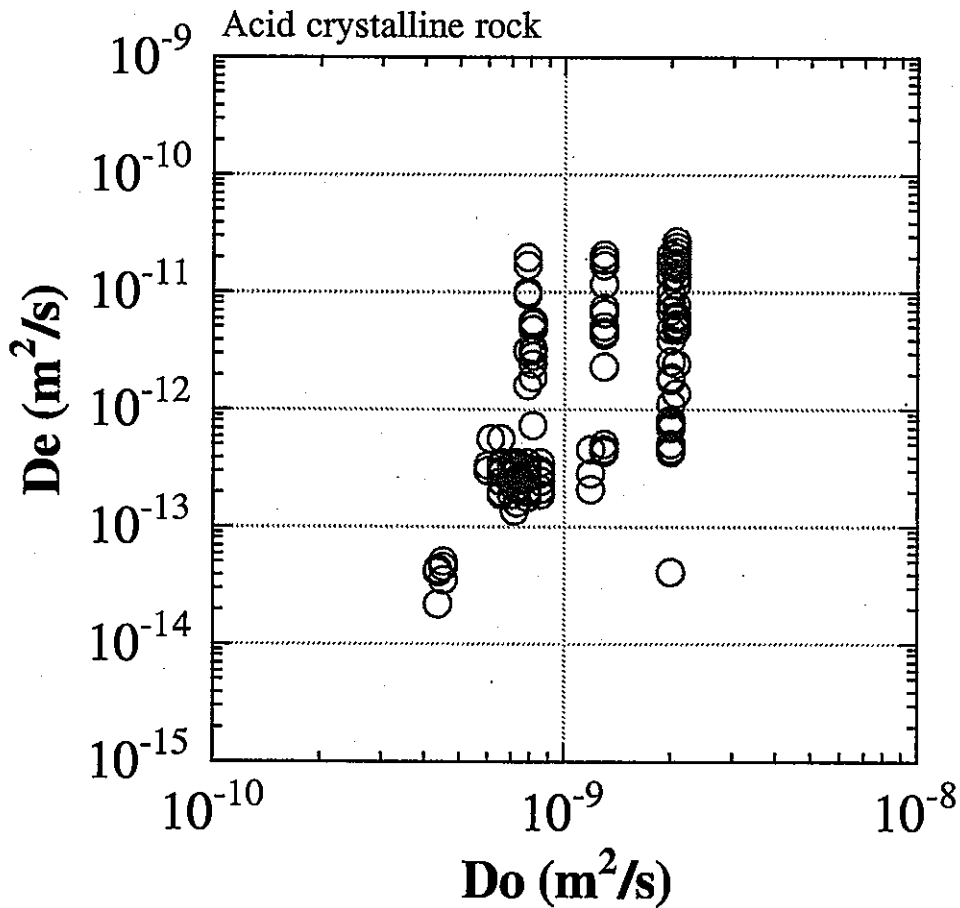


Figure 2.4.20-11 A correlation between  $D_e$  values and  $D_o$  values for acid crystalline rock (granite, granodiorite, altered granodiorite, biotitic granite)  
No systematic tendency is found between  $D_e$  and  $D_o$ .



# Chapter 3 Conclusions

## 3.1 Summary

A database for diffusivity for a data setting of  $D_e$  values in rock matrices in the second progress report, was developed. In this database, 3 kinds of diffusion coefficients:  $D_e$ ,  $D_a$  and  $D_o$  were treated. The conclusion is summarized as follows.

In **Chapter 1**, theoretical background for mass transport, methodology for diffusivity data acquisition and the basic structure of the database were explained.

In **Chapter 2**, rock to be treated in this database was categorized into 4 kinds of rocks: acid crystalline rock, alkaline crystalline rock, sedimentary rock (argillaceous/tuffaceous rock) and sedimentary rock (psammitic rock/sandy stone) from the viewpoint of geology and mass transport, and  $D_e$  and  $D_a$  data reported between 1980 and 1998 were explained for each element, including also experimental conditions, method for diffusivity data acquisition and discussion for correlations between various parameters.

The database was developed considering the following points.

- (1) Since Japanese geological environment is focused in the second progress report, data for diffusion are collected focused on Japanese major rocks.
- (2) Although 22 elements are considered to be important in performance assessment for geological disposal, all elements and aquatic tracers are treated in this database development considering general purpose.
- (3) Since limestone, which belongs to sedimentary rock, can become one of the natural resources and is inappropriate as a host rock, it is omitted in this database development.

Rock was categorized into 4 kinds of rocks, as described above. In addition, rocks around neutrality among crystalline rock were categorized into the alkaline crystalline rock in this database. The database is composed of sub-databases for 4 kinds of rocks. Furthermore, the sub-databases for 4 kinds of the rocks are composed of databases to individual element, in which totally, 24 items: element (nuclide), element No., species, charge, solid (rock name), dry density ( $\rho$ ), porosity ( $\phi$ ),  $D_e$ ,  $\log D_e$ ,  $D_o$ , formation factor (FF), geometric factor (Gf), rock capacity factor ( $\alpha$ ),  $D_a$ ,  $\log D_a$ , literature No., method, porewater, pH, tracer, contact time, temperature, Eh, and others, are input.

As a result of literature survey, for  $D_e$  values for acid crystalline rock, totally, 207 data for 18 elements and one tracer (hydrocarbon: uranine) have been reported and all data were for granitic rocks such as granite, granodiorite and biotitic granite. For alkaline crystalline rock, totally, 32  $D_e$  data for 6 elements have been reported for basalt, andesite and schist. For

sedimentary rock (argillaceous/tuffaceous rock), totally, 54 De data for 8 elements have been reported for mudstone, pelitic schist and tuff, and for sedimentary rock (psammitic rock/sandy stone), 11 De data for 1 element have been reported for siliceous sedimentary rock. As is clear, much De data are found in granitic rocks. Whilst, De data for psammitic rocks are short comparing to those for the other kinds of rocks. **Table 3.1-1** shows a summary of De and Da values for each element and each kind of rock treated in this database. The De values for totally 18 elements and one aquatic tracer (uranine) and Da values for totally 14 elements have been reported for acid crystalline rock. The De values for totally 6 elements and Da values for totally 4 elements have been reported for alkaline crystalline rock. The De and Da values for one element (I) have been reported for sedimentary rock (psammitic rock/sandy stone). The De and Da values for totally 8 elements have been reported for sedimentary rock (argillaceous/tuffaceous rock).

Correlativities have been approximately found between De values and rock porosities for all kinds of the rocks, but significant porosity dependencies have not been found for Da values. In addition, correlations between various parameters such as that between formation factors and rock porosities, that between geometric factors and rock porosities, that between De values and ionic charge, that between De values and Do values, and that between De values and element, were also discussed.

### **3.2 Future Work**

In this database, only domestic rocks have been treated, and data for rocks of foreign countries are not included. And since data collected in this database are based on literatures published between 1980 and 1998, much diffusivity data reported after this period are not included yet. This database will be periodically renewed incorporating new data. And diffusivity data for rocks of foreign countries will be also included in the database.

**Table 3.1-1** Summary of De and Da values for each element and each kind of rock treated in this database

Element (tracer)	Acid crystalline rock		Alkaline crystalline rock		Sedimentary rock(psammitic rock/sandy stone)		Sedimentary rock(argillaceous /tuffaceous rock)	
	De	Da	De	Da	De	Da	De	Da
Ba	○							
Be	○							
Cl	○	○	○	○			○	○
Co	○	○						
Cs	○	○	○	○			○	○
HTO	○	○						
I	○	○	○	○	○	○	○	○
K	○		○				○	
Mg	○							
Na	○	○	○				○	○
Ni	○	○	○	○			○	○
Np	○	○						
Pu	○							
Se	○	○					○	○
Sm	○	○					○	○
Sr	○	○						
Tc	○	○						
U	○	○						○
Uranine	○	○						
Number of ○	19	14	6	4	1	1	8	8

## **Chapter 4 Data Sheet**

In **Chapter 4**, data concerning diffusion, collected based on literature survey, are shown in data sheet. In addition, the footnotes and references are also shown.

### **4.1 Data Sheet for Sedimentary Rock (argillaceous/tuffaceous rock)**

The diffusivity data for Cl, Cs, I, K, Na, Ni, Se, Sm and U in sedimentary rock (argillaceous/tuffaceous rock) are shown in turn.

ページ #1 - "泥質/凝灰岩De/Da(Cl)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Cl:De(m2/s)	Cl:log De	Do(m2/s) 10)	Formation factor(FF) 11)	Geomteric factor(Gf) 12)	Rock capacity factor
0	Cl	1	Cl-	-1.0	Mudstone 1)	2.78	1.80	3.20e-14	-13.495	2.00e-09	1.60e-05	0.000890	
1	Cl	1	Cl-	-1.0	Mudstone 1)	2.78	1.80	9.60e-14	-13.018	2.00e-09	4.80e-05	0.00270	
2	Cl	1	Cl-	-1.0	Mudstone 1)	2.78	1.80	8.80e-14	-13.051	2.00e-09	4.50e-05	0.00250	0.100
3	Cl	1	Cl-	-1.0	Pelitic schist		1.00	1.50e-13	-12.824	2.00e-09	7.50e-05	0.00750	
4	Cl	1	Cl-	-1.0	Pelitic schist		1.00	6.30e-13	-12.201	2.00e-09	0.000320	0.0320	
5	Cl	1	Cl-	-1.0	Mudstone		0.740	3.20e-14	-13.495	2.00e-09	1.60e-05	0.00220	
6	Cl	1	Cl-	-1.0	Mudstone		0.740	9.60e-14	-13.018	2.00e-09	4.80e-05	0.00650	
7	Cl	1	Cl-	-1.0	Mudstone			8.80e-14	-13.051	2.00e-09			0.100
8	Cl	1	Cl-	-1.0	Mudstone		0.740	3.20e-14	-13.495	2.00e-09	1.60e-05	0.00220	
9	Cl	1	Cl-	-1.0	Mudstone		0.740	9.60e-14	-13.018	2.00e-09	4.80e-05	0.00650	
10	Cl	1	Cl-	-1.0	Tuff 8)	1.53	37.4	9.60e-12	-11.018	2.00e-09	0.00480	0.0130	0.0690
11	Cl	1	Cl-	-1.0	Tuff 8)	1.53	37.4	1.60e-11	-10.796	2.90e-09	0.00800	0.0210	0.00590
12	Cl	1	Cl-	-1.0	Tuff 8)	1.53	37.4	3.40e-11	-10.469	2.00e-09	0.0170	0.0450	0.374

ページ #2 - "泥質/凝灰岩Da/Da(Cl)data"

	Cl:Da(m <sup>2</sup> /s)	Cl:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0			1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 3)
1			1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 3)
2	8.80e-13	-12.058	1	Through-diffusion	Distilled water	6.00	0.6M Cl 2)	596days	25.0		aerobic 3)
3	2.60e-11	-10.585	2	Through-diffusion	Distilled water		NaCl 4)	11days	room		aerobic 5)
4	3.80e-11	-10.420	2	Through-diffusion	Distilled water		NaCl 4)	11days	room		aerobic 5)
5			3	Through-diffusion	Distilled water	5.50	60000ppmCl				aerobic 6)
6			3	Through-diffusion	Distilled water	5.50	60000ppmCl				aerobic 6)
7	8.80e-13	-12.056	4	Through-diffusion	Distilled water	6.00	0.6M Cl 2)	596days	25.0		aerobic
8			5	Through-diffusion	Distilled water		60000ppmCl	90days	room		aerobic 7)
9			5	Through-diffusion	Distilled water		60000ppmCl	90days	room		aerobic 7)
10	2.60e-11	-10.585	6	Through-diffusion	Distilled water	9.48	0.1M NaCl	18days	25.0		aerobic 9)
11	4.30e-11	-10.367	6	Through-diffusion	Distilled water	9.54	0.1M NaCl	18days	25.0		aerobic 9)
12	9.10e-11	-10.041	6	Through-diffusion	Distilled water	9.27	0.1M NaCl	18days	25.0		aerobic 9)

## ページ #1 - "泥質/凝灰岩De/Da(Cl)data"

## REFERENCE AND FOOTNOTE

## Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V.(1997).
- 2 佐藤治夫：緩衝材及び岩体中での核種の拡散・収着データ及びその機構、動燃技術資料「性能評価研究情報交換会報告書」、PNC TN1410 91-079、pp.275-286、動燃事業団(1991.10)(1991).  
H. Sato: PNC Technical Report "Report of Information Exchange Meeting on the Performance Assessment Research", PNC TN1410 91-079, pp.275-286, Power Reactor and Nuclear Fuel Development Corporation (1991. 10)(1991).
- 3 佐藤治夫、油井三和：岩石中のイオンの拡散係数の測定、日本原子力学会1992年秋の大会、F15、p.323(1992).  
Sato, H., Yui, M.: Measurement of Ionic Diffusion Coefficients in Rocks, 1992 Fall Meeting of the Atomic Energy Society of Japan, F15, p.323 (1992).
- 4 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 5 佐藤治夫、油井三和、石川博久：岩石の間隙構造に関する研究－岩石の間隙率およびNaClを用いたThrough-diffusion(透過拡散)試験による間隙因子の測定－、動燃技術資料、PNC TN8410 92-222、動燃事業団(1992. 8)(1992).  
Sato, H., Yui, M., Ishikawa, H.: A Study on Pore Structure of Rocks, PNC Technical Report, PNC TN8410 92-222, Power Reactor and Nuclear Fuel Development Corporation (1992. 8)(1992).
- 6 Sato, H.: Through-Diffusion Experiments on NaCl through Tuff -Measurement of Tortuosity-, PNC Technical Report, PNC TN8410 96-111, Power Reactor and Nuclear Fuel Corporation (1996. 7)(1996).

## Footnote:

- 1) The mudstone sample was sampled from Tome-gun, Miyagi-ken, Japan. This sample is approximately equivalent to a standard sample in the chemical composition of Japanese mudstone in the Geological Survey of Japan.
- 2) The tracer was used as a mixture of CsCl, NiCl and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water. The concentration of Cl<sup>-</sup> was estimated as a counter ion of Cs<sup>+</sup>, Ni<sup>2+</sup> and Sm<sup>3+</sup> in the tracer. Since the concentration of each cation was 0.1 M,

ページ #2 - “泥質/凝灰岩De/Da(Cl)data”

the concentration of Cl<sup>-</sup> is estimated to be 0.6 M.

- 3) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. The porosity and density shown are average values of respective measured values.  
The De values were originally obtained to derive formation factor and tortuosity of the rock.
- 4) The concentration of NaCl is not reported in the literature.
- 5) The De values were originally obtained to derive formation factor and tortuosity of the rock.
- 6) The porosity was determined by water saturation method (water intrusion method). The porosity shown is an average value of the measured ones.  
The De values were originally obtained to derive formation factor and tortuosity of the rock, because Cl<sup>-</sup> ion is non-sorbing.
- 7) The porosity was determined by two kinds of water saturation methods (water intrusion methods) and the porosity shown is an average value of the measured ones.  
The De values were originally obtained to derive formation factor and tortuosity of the rock, because Cl<sup>-</sup> ion is non-sorbing.
- 8) The tuff sample was sampled from 72.3m deep (AN-4 bore hole), Tono mine, Toki-shi, Gifu-ken, Japan.
- 9) The porosity and density were determined by mercury porosimetry.  
The De values were originally obtained to derive formation factor and tortuosity of the rock, because Cl<sup>-</sup> ion is non-sorbing.
- 10) The Do of Cl<sup>-</sup> is estimated 2.0e-9 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169 (1997).].
- 11) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 12) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.



	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Cs:De(m2/s)	Cs:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor
0	Cs	2	Cs+	1.0	Mudstone 1)	2.78	1.80	4.80e-13	-12.319	2.10e-09	0.000230	0.0130	1.23
1	Cs	2	Cs+	1.0	Mudstone 1)			4.80e-13	-12.319	2.10e-09	0.000230	0.0130	1.23

ページ #2 - "泥質/凝灰岩Da/Da(Cs)data"

	Cs:Da(m <sup>2</sup> /s)	Cs:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	3.90e-13	-12.409	1	Through-diffusion	Distilled water	6.00	0.1M CsCl 2)		25.0		aerobic 3)
1	3.90e-13	-12.409	2	Through-diffusion	Distilled water	6.00	0.1M CsCl 2)		25.0		aerobic

ページ #1 - “泥質/凝灰岩De/Da(Cs)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V.(1997).
- 2 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

Footnote:

- 1) The mudstone sample was sampled from Tome-gun, Miyagi-ken, Japan. This sample is approximately equivalent to a standard sample in the chemical composition of Japanese mudstone in the Geological Survey of Japan.
- 2) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water.
- 3) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. The porosity and density shown are average values of respective measured values.  
The De values for Cl<sup>-</sup> which is a counter ion of Cs<sup>+</sup> were originally obtained to derive formation factor and tortuosity of the rock.
- 4) The Do of Cs<sup>+</sup> is estimated 2.1e-9 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168 (1997).].
- 5) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\Phi \cdot Do)=FF/\Phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\Phi$  is the porosity and FF is the formation factor.

ページ #1 - "泥質/凝灰岩De/Da(I)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	I:De(m2/s)	I:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor	I:Da(m2/s)
0	I	3	I-	-1.0	Tuff 1)		24.8	5.30e-11	-10.276	2.00e-09	0.0270	0.110	0.20	2.65e-10
1	I	3	I-	-1.0	Tuff 1)		24.6	9.30e-11	-10.032	2.00e-09	0.0470	0.190	0.33	2.82e-10
2	I	3	I-	-1.0	Tuff 1)		24.6	5.30e-11	-10.276	2.00e-09	0.0270	0.110	0.20	2.65e-10
3	I	3	I-	-1.0	Tuff 1)		24.6	9.30e-11	-10.032	2.00e-09	0.0470	0.190	0.33	2.82e-10

ページ #2 - "泥質/凝灰岩De/Da(I)data"

	Elog Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	-9.5770	1	Through-diffusion	Deionized water		1M(l:127000ppm)KI		room		aerobic 2)
1	-9.5500	1	Through-diffusion	Deionized water		1M(l:127000ppm)KI		room		aerobic 2)
2	-9.5770	2	Through-diffusion	Distilled water		1M(l:127000ppm)KI		room		aerobic 3)
3	-9.5500	2	Through-diffusion	Distilled water		1M(l:127000ppm)KI		room		aerobic 3)

ページ #1 - “泥質/凝灰岩De/Da(l)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 西山勝栄、中嶋悟、多田隆治、内田隆：岩石間隙水中のイオンの拡散係数と間隙の性質、鉱山地質、40 (5)、323~336 (1990).  
Nishiyama, K., Nakashima, S., Tada, R., Uchida, T.: Diffusion of an Ion in Rock Pore Water and Its Relation to Pore Characteristics, Mining Geology, 40 (5), pp.323-336 (1990).
- 2 喜多治之、岩井孝幸、中嶋悟：花崗岩および凝灰岩間隙水中のイオンの拡散係数の測定、応用地質、30 (2)、26~32 (1989).  
Kita, H., Iwai, T., Nakashima, S.: Diffusion Coefficient Measurement of an Ion in Pore Water of Granite and Tuff, Journal of the Japan Society of Engineering Geology, 30-2, pp.26-32 (1989).

Footnote:

- 1) The tuff sample was sampled from Izu, Shizuoka-ken, Japan.
- 2) The rock porosity was measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry to discuss effective porosity.  
The Da values were calculated from De values and rock capacity factors.
- 3) The rock porosity was determined by water saturation method (water intrusion method). The error of the porosity is evaluated to be  $24 \pm 0.3\%$  in the literature.  
The Da values were calculated from De values and rock capacity factors.
- 4) The Do of I- is estimated  $2.0 \times 10^{-9}$  m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169 (1997).].
- 5) The formation factor (FF) was calculated based on the equation:  $FF = De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf = Dp/Do = De/(\Phi \cdot Do) = FF/\Phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\Phi$  is the porosity and FF is the formation factor.

ページ #1 - "泥質/凝灰岩De/Da(K)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	K:De(m2/s)	K:log De	Do(m2/s) 6)	Formation factor(FF) 7)	Geometric factor(Gf) 8)	Rock capacity factor	K:Da(m2/s)
0	K	4	K+	1.0	Mudstone 1)	2.72	0.740	4.30e-13	-12.367	2.00e-09	0.000220	0.0290		
1	K	4	K+	1.0	Mudstone 1)	2.72	0.740	5.50e-13	-12.250	2.00e-09	0.000280	0.0370		
2	K	4	K+	1.0	Tuff 2)	1.34	55.9	5.80e-10	-9.2370	2.00e-09	0.290	0.520		
3	K	4	K+	1.0	Tuff 2)	1.34	55.9	7.50e-10	-9.1250	2.00e-09	0.380	0.670		
4	K	4	K+	1.0	Mudstone 1)	2.72	0.740	4.31e-13	-12.368	2.00e-09	0.000220	0.0290		
5	K	4	K+	1.0	Mudstone 1)	2.72	0.740	4.23e-13	-12.374	2.00e-09	0.000210	0.0290		
6	K	4	K+	1.0	Mudstone 1)	2.72	0.740	5.54e-13	-12.256	2.00e-09	0.000280	0.0370		
7	K	4	K+	1.0	Tuff 2)	1.34	55.9	7.48e-10	-9.1280	2.00e-09	0.370	0.670		
8	K	4	K+	1.0	Tuff 2)	1.34	55.9	6.17e-10	-9.2100	2.00e-09	0.310	0.550		
9	K	4	K+	1.0	Tuff 2)	1.34	55.9	5.77e-10	-9.2390	2.00e-09	0.290	0.520		

ページ #2 - "泥質/凝灰岩De/Da(K)data"

	K.log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0		1	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
1		1	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
2		1	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
3		1	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
4		2	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
5		2	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
6		2	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
7		2	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
8		2	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)
9		2	conductivity 3)	0.1M KCl 4)		0.1M KCl 4)		room		aerobic 5)



ページ #1 - “泥質/凝灰岩De/Da(K)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Conca, J. L., Apted, M. J., Arthur, R. C.: Direct Determinations of Transport Parameters in Repository Materials, Scientific Basis for Nuclear Waste Management XVI (Interrante, C. G. and Pabalan, R. T., eds.), Materials Research Society Symposium Proceedings, Vol. 294, pp.839-844, Materials Research Society, Pittsburgh, Pennsylvania (1993).
- 2 Conca, J. L., Wright, J.: Direct Determinations of Unsaturated Flow and Transport, Proceedings of the Twelfth Annual American Geophysical Union Hydrology Days Meeting, Fort Collins, March 1992, pp.103-116 (1992).

Footnote:

- 1) The mudstone sample was sampled from Tome-gun, Miyagi-ken, Japan. This sample is approximately equivalent to a standard sample in the chemical composition of Japanese mudstone in the Geological Survey of Japan. What described above is not shown in the literature, but PNC supplied this sample and simultaneously measured the physical properties [H. Sato and T. Shibutani: PNC Technical Report, PNC TN8410 94-284, 71-89 (1994).].
- 2) The tuff sample was sampled from Tono mine, Toki-shi, Gifu-ken, Japan. The physical properties except for density are not reported in the literature, but PNC supplied this sample and simultaneously measured them [H. Sato and T. Shibutani: PNC Technical Report, PNC TN8410 94-284, 71-89 (1994).].
- 3) The De values were determined by electrical conductivity measurements, based on the Nernst-Einstein equation. The De values were determined using the unsaturated flow apparatus (UFA). Hydraulic conductivity can also be determined by this method.
- 4) The rock sample was firstly saturated with a 0.1 M KCl solution and then electrical conductivity was measured. The porewater and tracer are therefore the same.
- 5) Apparent diffusion coefficient (Da) cannot be obtained from the electrical conductivity measurements.
- 6) The Do of K<sup>+</sup> is estimated 2.0e-9 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168 (1997).].
- 7) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 8) The geometric factor (Gf) is also called tortuosity factor (Tf), and

ページ #2 - “泥質/凝灰岩De/Da(K)data”

was calculated based on the equation:  $Gf = Dp/Do = De / (\phi \cdot Do) = FF / \phi$ .  
Where  $Dp$  is the porewater diffusion coefficient,  $Do$  is the diffusion coefficient in free water,  $De$  is the effective diffusion coefficient,  $\phi$  is the porosity and  $FF$  is the formation factor.

ページ #1 - "泥質/凝灰岩De/Da(Na)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Na:De(m2/s)	Na:log De	Do(m2/s) 5)	Formation factor(FF) 6)	Geometric factor(Gf) 7)	Rock capacity factor
0	Na	5	Na+	1.0	Mudstone 1)	2.78	1.80	6.50e-14	-13.187	1.30e-09	6.00e-05	0.00280	
1	Na	5	Na+	1.0	Mudstone 1)	2.78	1.80	1.20e-13	-12.921	1.30e-09	9.20e-05	0.00510	
2	Na	5	Na+	1.0	Tuff 3)	1.53	37.4	1.50e-11	-10.824	1.30e-09	0.0120	0.0320	0.374
3	Na	5	Na+	1.0	Tuff 3)	1.53	37.4	2.10e-11	-10.678	1.30e-09	0.0160	0.0430	0.374
4	Na	5	Na+	1.0	Tuff 3)	1.53	37.4	3.80e-11	-10.420	1.30e-09	0.0290	0.0780	0.374

ページ #2 - "泥質/凝灰岩De/Da(Na)data"

	Na:Da(m <sup>2</sup> /s)	Na:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0			1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 2)
1			1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 2)
2	4.00e-11	-10.398	2	Through-diffusion	Distilled water	9.48	0.1M NaCl	18days	25.0		aerobic 4)
3	5.60e-11	-10.252	2	Through-diffusion	Distilled water	9.54	0.1M NaCl	18days	25.0		aerobic 4)
4	1.00e-10	-10.000	2	Through-diffusion	Distilled water	9.27	0.1M NaCl	18days	25.0		aerobic 4)

ページ #1 - “泥質/凝灰岩De/Da(Na)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V.(1997).
- 2 Sato, H.: Through-Diffusion Experiments on NaCl through Tuff -Measurement of Tortuosity-, PNC Technical Report, PNC TN8410 96-111, Power Reactor and Nuclear Fuel Development Corporation (1996.7)(1996).

Footnote:

- 1) The mudstone sample was sampled from Tome-gun, Miyagi-ken, Japan. This sample is approximately equivalent to a standard sample in the chemical composition of Japanese mudstone in the Geological Survey of Japan.
- 2) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. The porosity and density shown are average values of respective measured values.  
The De values were originally obtained to derive formation factor and tortuosity of the rock.
- 3) The tuff sample was sampled from 72.3m deep (AN-4 bore hole), Tono mine, Toki-shi, Gifu-ken, Japan.
- 4) The porosity and density were determined by mercury porosimetry.  
The De values were originally obtained to derive formation factor and tortuosity of the rock.
- 5) The Do of Na<sup>+</sup> is estimated 1.3e-9 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168 (1997).].
- 6) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 7) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "泥質/凝灰岩De/Da(Ni)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Ni:De(m2/s)	Ni:log De	Do(m2/s) 7)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor
0	Ni	8	Ni2+	2.0	Mudstone 1)	2.78	1.80	1.00e-13	-13.000	6.60e-10	0.000150	0.00840	0.33
1	Ni	8	Ni2+	2.0	Mudstone 1)			1.00e-13	-13.000	6.60e-10	0.000150	0.00840	0.33

ページ #2 - "泥質/凝灰岩De/Da(Ni)data"

	Ni:Da(m <sup>2</sup> /s)	Ni:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	3.00e-13	-12.523	1	Through-diffusion	Distilled water	6.00	0.1M NiCl <sub>2</sub> 2)		25.0		aerobic 3)
1	3.00e-13	-12.523	2	Through-diffusion	Distilled water	6.00	0.1M NiCl <sub>2</sub> 2)		25.0		aerobic 4)

ページ #1 - “泥質/凝灰岩De/Da(Ni)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V.(1997).
- 2 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

Footnote:

- 1) The mudstone sample was sampled from Tome-gun, Miyagi-ken, Japan. This sample is approximately equivalent to a standard sample in the chemical composition of Japanese mudstone in the Geological Survey of Japan.
- 2) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water.
- 3) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. The porosity and density shown are average values of respective measured values.  
The De values for Cl<sup>-</sup> which is a counter ion of Ni<sup>2+</sup> were originally obtained to derive formation factor and tortuosity of the rock.
- 4) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. However, neither the porosity nor density is reported in the literature.
- 5) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.
- 7) The Do of Ni<sup>2+</sup> is estimated 6.6e-10 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168 (1997).].



ページ #1 - "泥質/凝灰岩De/Da(Se)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Se:De(m2/s)	Se:log De	Do(m2/s) 8)	Formation factor(FF) 6)	Geometric factor(Gf) 7)	Rock capacity factor	Se:Da(m2/s)
0	Se	7	SeO32-	-2.0	Tuff 1)	1.50	37.4	8.40e-12	-11.194	8.10e-10	0.00780	0.0210	1.79	3.58e-12
1	Se	7	SeO32-	-2.0	Tuff 1)	1.50	37.4	2.70e-12	-11.569	8.10e-10	0.00330	0.00890	0.520	5.19e-12
2	Se	7	SeO32-	-2.0	Tuff 1)	1.50	37.4	5.90e-12	-11.229	8.10e-10	0.00730	0.0180	1.66	3.51e-12
3	Se	7	SeO32-	-2.0	Tuff 1)	1.50	37.4	5.60e-12	-11.252	8.10e-10	0.00690	0.0180	0.590	9.49e-12
4	Se	7	SeO32-	-2.0	Tuff 1)	1.50	37.4	6.00e-12	-11.222	8.10e-10	0.00740	0.0200	0.570	1.05e-11
5	Se	7	SeO32-	-2.0	Tuff 1)	1.50	37.4	6.90e-12	-11.181	8.10e-10	0.00850	0.0230	0.940	7.34e-12
6	Se	7	SeO32-	-2.0	Tuff 1)	1.53	37.3	8.42e-12	-11.192	8.10e-10	0.00790	0.0210	1.79	3.59e-12
7	Se	7	SeO32-	-2.0	Tuff 1)	1.53	37.3	5.57e-12	-11.254	8.10e-10	0.00680	0.0180	0.590	9.44e-12
8	Se	7	SeO32-	-2.0	Tuff 1)	1.53	37.3	6.20e-12	-11.208	8.10e-10	0.00770	0.0210	1.70	3.65e-12
9	Se	7	SeO32-	-2.0	Tuff 1)	1.53	37.3	6.20e-12	-11.208	8.10e-10	0.00770	0.0210	0.700	8.86e-12
10	Se	7	SeO32-	-2.0	Tuff 4)	1.53	37.3	8.80e-12	-11.056	8.10e-10	0.0110	0.0290	2.30	3.83e-12
11	Se	7	SeO32-	-2.0	Tuff 4)	1.53	37.3	3.40e-12	-11.489	8.10e-10	0.00420	0.0110	1.50	2.27e-12
12	Se	7	SeO32-	-2.0	Tuff 4)	1.53	37.3	7.90e-12	-11.102	8.10e-10	0.00980	0.0260	2.10	3.76e-12
13	Se	7	SeO32-	-2.0	Tuff 4)	1.53	37.3	5.90e-12	-11.229	8.10e-10	0.00730	0.0200	0.620	9.52e-12
14	Se	7	SeO32-	-2.0	Tuff 4)	1.53	37.3	6.40e-12	-11.194	8.10e-10	0.00790	0.0210	0.610	1.05e-11
15	Se	7	SeO32-	-2.0	Tuff 4)	1.53	37.3	7.40e-12	-11.131	8.10e-10	0.00910	0.0240	0.960	7.71e-12

ページ #2 - "泥質/凝灰岩De/Da(Se)data"

	Se/log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	-11.446	1	Through-diffusion	0.1M NaCl	7.90	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 2)
1	-11.285	1	Through-diffusion	0.1M NaCl	7.80	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 2)
2	-11.455	1	Through-diffusion	0.1M NaCl	7.70	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 2)
3	-11.003	1	Through-diffusion	0.1M NaCl	11.0	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 2)
4	-10.979	1	Through-diffusion	0.1M NaCl	11.0	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 2)
5	-11.134	1	Through-diffusion	0.1M NaCl	11.0	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 2)
6	-11.445	2	Through-diffusion	0.1M NaCl	8.00			22.0		anaerobic 3)
7	-11.025	2	Through-diffusion	0.1M NaCl	11.0			22.0		anaerobic 3)
8	-11.438	2	Through-diffusion	0.1M NaCl	8.00			22.0		anaerobic 3)
9	-11.053	2	Through-diffusion	0.1M NaCl	11.0			22.0		anaerobic 3)
10	-11.417	3	Through-diffusion	0.1M NaCl	7.90	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 5)
11	-11.644	3	Through-diffusion	0.1M NaCl	7.80	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 5)
12	-11.425	3	Through-diffusion	0.1M NaCl	7.70	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 5)
13	-11.021	3	Through-diffusion	0.1M NaCl	11.0	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 5)
14	-10.979	3	Through-diffusion	0.1M NaCl	11.0	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 5)
15	-11.113	3	Through-diffusion	0.1M NaCl	11.0	e-3M SeO2	about 100days	Temp. in G.B.		anaerobic 5)

ページ #1 - “泥質/凝灰岩De/Da(Se)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 佐藤治夫、澁谷朝紀：岩盤中における吸着・拡散係数、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010、pp.150-155、動燃事業団(1996. 12)(1996).  
Sato, H., Shibutani, T.: PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.150-155, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 2 動燃事業団：地層処分研究開発の現状(平成8年度、動燃技術資料、PNC TN1410 96-071、pp.208-210、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.208-210, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 3 Tachi, Y., Shibutani, T., Sato, H., Yui, M.: Sorption and Diffusion Behavior of Selenium in Tuff, Journal of Contaminant Hydrology 35, pp.77-89 (1998).

Footnote:

- 1) The tuff sample was sampled from Tono mine, Toki-shi, Gifu-ken, Japan. The mineralogy of the tuff is quartz, plagioclase, potassium feldspar, smectite, clinoptilolite (斜プチロル沸石), stilbite, amphibole, calcite, pyrite and lignite.
- 2) The density and porosity were determined by mercury porosimetry. Pore-size distribution and specific surface area of the pores were also evaluated by the same method. The former was evaluated that the pores in the range of 10nm~10 $\mu$ m were dominant, and the latter was estimated to be 17.8 m<sup>2</sup>/g on the average.  
The Da values were calculated from dry densities, porosities and distribution coefficients (Kd). The rock capacity factors are in same.  
The experiments were carried out in an N<sub>2</sub>-atmosphere glove-box, in which the concentration of oxygen was kept less than 1ppm.
- 3) The Da values were calculated from De values and rock capacity factors given in the literature.  
The experiments were carried out in an N<sub>2</sub>-atmosphere glove-box, in which the concentration of oxygen was kept less than 1ppm.
- 4) The tuff sample was sampled from the Toki Lignite-Bearing Formation (AN-4 bore hole, 72m below ground level), Tono area, Toki-shi, Gifu-ken, Japan.  
The mineralogy of the tuff is plagioclase, smectite, clinoptilite, heulandite, quartz, kaolinite, biotite, gypsum and pyrite, and these were identified by X-ray diffraction analysis and section observation.

ページ #2 - “泥質/凝灰岩De/Da(Se)data”

- 5) All the experiments were carried out in a N<sub>2</sub>-atmosphere glove-box, in which the concentration oxygen was kept less than 1ppm.  
The Da values were calculated from De values and rock capacity factors.  
The tracer was prepared by dissolving a SeO<sub>2</sub> powder in 0.1 M NaCl.  
Therefore, the ionic strength of the porewater is 0.1.
- 6) The formation factor (FF) was calculated based on the equation:  
 $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 7) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ .  
Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.
- 8) The Do of SeO<sub>3</sub><sup>2-</sup> is estimated  $8.1e-10$  m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.170 (1997).].

ページ #1 - "泥質/凝灰岩De/Da(Sm)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Sm:De(m2/s)	Sm:log De	Do(m2/s) 7)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor
0	Sm	8	Sm3+	2.0	Mudstone 1)	2.78	1.80	5.10e-14	-13.276	6.10e-10	0.000840	0.00460	0.20
1	Sm	8	Sm3+	2.0	Mudstone 1)			5.10e-14	-13.276	6.10e-10	0.000840	0.00460	0.20

ページ #2 - "泥質/凝灰岩De/Da(Sm)data"

	Sm:Da(m <sup>2</sup> /s)	Sm:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	2.65e-13	-12.577	1	Through-diffusion	Distilled water	6.00	0.1M SmCl <sub>3</sub> 2)		25.0		aerobic 3)
1	2.60e-13	-12.585	2	Through-diffusion	Distilled water	6.00	0.1M SmCl <sub>3</sub> 2)		25.0		aerobic 4)

ページ #1 - “泥質/凝灰岩De/Da(Sm)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V.(1997).
- 2 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

Footnote:

- 1) The mudstone sample was sampled from Tome-gun, Miyagi-ken, Japan. This sample is approximately equivalent to a standard sample in the chemical composition of Japanese mudstone in the Geological Survey of Japan.
- 2) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water.
- 3) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. The porosity and density shown are average values of respective measured values.  
The De values for Cl<sup>-</sup> which is a counter ion of Sm<sup>3+</sup> were originally obtained to derive formation factor and tortuosity of the rock.  
The Da was calculated from De and rock capacity factor in the literature.
- 4) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. However, neither the porosity nor density is reported in the literature.
- 5) The formation factor (FF) was calculated based on the equation:  
 $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ .  
Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.
- 7) The Do of Sm<sup>3+</sup> is estimated 6.1e-10 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion

ページ #2 - “泥質/凝灰岩De/Da(Sm)data”

Properties, Marcel Dekker, Inc., New York, U. S. A., p.169 (1997).].



ページ #1 - "泥質/凝灰岩De/Da(U)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	U:De(mZ/s)	U:log De	Do(mZ/s) 10)	Formation factor(FF) 8)	Geometric factor(GF) 9)	Rock capacity factor
0	U	9	carbonate complex 1)	-4.0	Tuff					7.20e-10			
1	U	9	carbonate complex 1)	-4.0	Tuff 5)					7.20e-10			
2	U	9	carbonate complex 1)	-4.0	Tuff 5)					7.20e-10			
3	U	9	carbonate complex 1)	-4.0	Tuff 5)					7.20e-10			

ページ #2 - "泥質/凝灰岩De/Da(U)data"

	U:Da(m2/s)	U:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	5.00e-12	-11.301	1	In-diffusion 2)	Deionized water		Uranyl nitrate 3)	30days	20~50		aerobic 4)
1	1.00e-12	-12.000	2	In-diffusion 2)	groundwater		1450ppm U-233 6)	14days	room		aerobic 7)
2	8.00e-13	-12.097	2	In-diffusion 2)	groundwater		1450ppm U-233 6)	14days	room		aerobic 7)
3	6.00e-11	-10.222	2	In-diffusion 2)	groundwater		1450ppm U-233 6)	14days	room		aerobic 7)

ページ #1 - “泥質/凝灰岩De/Da(U)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 小坂哲生、佐藤正知、古屋広高、新谷貞夫、松尾多盛：岩石ブロック中のウランの移動挙動、日本原子力学会「昭59分科会」、G82 (1984).  
Kosaka, T., Sato, S., Furuya, H., Araya, S., Matsuo, K.: Diffusion Behavior of Uranium in a Rock Block, 1984 Fall Meeting of the Atomic Energy Society of Japan, G82 (1984).
- 2 Ota, K., Yoshida, H.: Influence Microscopic Heterogeneity on Diffusion for Sedimentary Rocks, Proceedings of an NEA/SKI Symposium, Paris, France, 11-14, October, 1994, pp.237-243 (1994).

Footnote:

- 1) The dominant chemical species in the porewater is probably  $UO_2(CO_3)_3^{4-}$  judging from the chemical and redox conditions, but the speciation is not reported.
- 2) This method is sometimes called back-to-back method, but both are basically the same method.
- 3) A block sample with the size of 10x10x20mm, sealed with resin except for only one plane, was saturated with deionized water before the experiment. The sample was then contacted with a  $9.2 \times 10^{-3}$  M  $Na_2CO_3$  solution containing  $5.1 \times 10^{-4}$  M uranyl nitrate for a maximum of 30 days in a thermobath at 20~50°C. Then the sample was scraped at the end of the experiment and the rock powder collected was contacted with 3 N  $HNO_3$  solution to remove U(VI) from the rock. The extracted solution was centrifuged with 4000rpm for 20min and counted for  $\alpha$  activity with a liquid scintillation counter.
- 4) The sampling point is not reported. Neither the porosity nor density of the rock is reported in this literature.
- 5) The rock sample, tuffaceous sandstone, was sampled from Tono mine (KNA-3 bore hole), Toki-shi, Gifu-ken, Japan.
- 6) This is the initial concentration to contact with the rock sample. The uranium solution was prepared by leaching the uranium concentrated sedimentary rocks.
- 7) Neither the porosity nor density of the rock is reported in this literature.
- 8) The formation factor (FF) was calculated based on the equation:  
 $FF = D_e / D_o$ . Where  $D_e$  and  $D_o$  is the effective diffusion coefficient and

ページ #2 - “泥質/凝灰岩De/Da(U)data”

diffusion coefficient in free water, respectively.

- 9) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf = D_p / D_o = D_e / (\Phi \cdot D_o) = FF / \Phi$ . Where  $D_p$  is the porewater diffusion coefficient,  $D_o$  is the diffusion coefficient in free water,  $D_e$  is the effective diffusion coefficient,  $\Phi$  is the porosity and FF is the formation factor.
- 10) The  $D_o$  of  $UO_2(CO_3)_3^{4-}$  is estimated  $7.2 \pm 0.5 \times 10^{-10}$  m<sup>2</sup>/s at 25°C [山口徹治：稲田花崗岩中におけるアクチニド元素の拡散、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010、pp.156-160、動燃事業団(1996.12)(1996). Yamaguchi, T.: Diffusivity of Actinides in a Granite from Inada, Ibaraki, Japan, PNC Technical Report, PNC TN1100 96-010, pp.156-160, Power Reactor and Nuclear Fuel Development Corporation (1996.12)(1996).]. The  $D_o$  is not directly measured value, but was estimated based on the Stokes equation by the ionic radius calculated from the molecular structure.

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
0	Cl	1	Cl-	-1.0	Mudstone	2.78	1.80	3.20e-14	-13.495	2.00e-09
1	Cl	1	Cl-	-1.0	Mudstone	2.78	1.80	9.60e-14	-13.018	2.00e-09
2	Cl	1	Cl-	-1.0	Mudstone	2.78	1.80	8.90e-14	-13.051	2.00e-09
3	Cl	1	Cl-	-1.0	Pelitic schist		1.00	1.50e-13	-12.824	2.00e-09
4	Cl	1	Cl-	-1.0	Pelitic schist		1.00	6.30e-13	-12.201	2.00e-09
5	Cl	1	Cl-	-1.0	Mudstone		0.740	3.20e-14	-13.495	2.00e-09
6	Cl	1	Cl-	-1.0	Mudstone		0.740	9.60e-14	-13.018	2.00e-09
7	Cl	1	Cl-	-1.0	Mudstone			8.90e-14	-13.051	2.00e-09
8	Cl	1	Cl-	-1.0	Mudstone		0.740	3.20e-14	-13.495	2.00e-09
9	Cl	1	Cl-	-1.0	Mudstone		0.740	9.60e-14	-13.018	2.00e-09
10	Cl	1	Cl-	-1.0	Tuff	1.53	37.4	9.60e-12	-11.018	2.00e-09
11	Cl	1	Cl-	-1.0	Tuff	1.53	37.4	1.60e-11	-10.796	2.00e-09
12	Cl	1	Cl-	-1.0	Tuff	1.53	37.4	3.40e-11	-10.469	2.00e-09
13	Cs	2	Cs+	1.0	Mudstone	2.78	1.80	4.80e-13	-12.319	2.10e-09
14	Cs	2	Cs+	1.0	Mudstone			4.80e-13	-12.319	2.10e-09
15	I	3	I-	-1.0	Tuff		24.6	5.30e-11	-10.276	2.00e-09
16	I	3	I-	-1.0	Tuff		24.6	9.30e-11	-10.032	2.00e-09
17	I	3	I-	-1.0	Tuff		24.6	5.30e-11	-10.276	2.00e-09
18	I	3	I-	-1.0	Tuff		24.6	9.30e-11	-10.032	2.00e-09
19	K	4	K+	1.0	Mudstone	2.72	0.740	4.30e-13	-12.367	2.00e-09
20	K	4	K+	1.0	Mudstone	2.72	0.740	5.50e-13	-12.260	2.00e-09
21	K	4	K+	1.0	Tuff	1.34	55.9	5.80e-10	-9.2370	2.00e-09
22	K	4	K+	1.0	Tuff	1.34	55.9	7.50e-10	-9.1250	2.00e-09
23	K	4	K+	1.0	Mudstone	2.72	0.740	4.31e-13	-12.366	2.00e-09
24	K	4	K+	1.0	Mudstone	2.72	0.740	4.23e-13	-12.374	2.00e-09
25	K	4	K+	1.0	Mudstone	2.72	0.740	5.54e-13	-12.256	2.00e-09
26	K	4	K+	1.0	Tuff	1.34	55.9	7.48e-10	-9.1260	2.00e-09
27	K	4	K+	1.0	Tuff	1.34	55.9	6.17e-10	-9.2100	2.00e-09
28	K	4	K+	1.0	Tuff	1.34	55.9	5.77e-10	-9.2390	2.00e-09
29	Na	5	Na+	1.0	Mudstone	2.78	1.80	6.50e-14	-13.187	1.30e-09
30	Na	5	Na+	1.0	Mudstone	2.78	1.80	1.20e-13	-12.921	1.30e-09
31	Na	5	Na+	1.0	Tuff	1.53	37.4	1.50e-11	-10.824	1.30e-09
32	Na	5	Na+	1.0	Tuff	1.53	37.4	2.10e-11	-10.678	1.30e-09
33	Na	5	Na+	1.0	Tuff	1.53	37.4	3.80e-11	-10.420	1.30e-09

	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH	Tracer
0	1.60e-05	0.000890				Through-diffusion	Distilled water	5.50	10%NaCl
1	4.80e-05	0.00270				Through-diffusion	Distilled water	5.50	10%NaCl
2	4.50e-05	0.00250	0.10	8.80e-13	-12.056	Through-diffusion	Distilled water	6.00	0.6M Cl
3	7.50e-05	0.00750		2.60e-11	-10.585	Through-diffusion	Distilled water		NaCl
4	0.000320	0.0320		3.80e-11	-10.420	Through-diffusion	Distilled water		NaCl
5	1.60e-05	0.00220				Through-diffusion	Distilled water	5.50	60000ppmCl
6	4.80e-05	0.00650				Through-diffusion	Distilled water	5.50	60000ppmCl
7			0.10	8.80e-13	-12.056	Through-diffusion	Distilled water	6.00	0.6M Cl
8	1.60e-05	0.00220				Through-diffusion	Distilled water		60000ppmCl
9	4.80e-05	0.00650				Through-diffusion	Distilled water		60000ppmCl
10	0.00480	0.0130	0.069	2.60e-11	-10.585	Through-diffusion	Distilled water	9.48	0.1M NaCl
11	0.00800	0.0210	0.0059	4.30e-11	-10.367	Through-diffusion	Distilled water	9.54	0.1M NaCl
12	0.0170	0.0450	0.37	9.10e-11	-10.041	Through-diffusion	Distilled water	9.27	0.1M NaCl
13	0.000230	0.0130	1.2	3.90e-13	-12.409	Through-diffusion	Distilled water	6.00	0.1M CsCl
14	0.000230	0.0130	1.2	3.90e-13	-12.409	Through-diffusion	Distilled water	6.00	0.1M CsCl
15	0.0270	0.110	0.20	2.65e-10	-9.5770	Through-diffusion	Deionized water		1M(l:127000ppm)KI
16	0.0470	0.190	0.33	2.82e-10	-9.5500	Through-diffusion	Deionized water		1M(l:127000ppm)KI
17	0.0270	0.110	0.20	2.65e-10	-9.5770	Through-diffusion	Distilled water		1M(l:127000ppm)KI
18	0.0470	0.190	0.33	2.82e-10	-9.5500	Through-diffusion	Distilled water		1M(l:127000ppm)KI
19	0.000220	0.0290				conductivity	0.1M KCl		0.1M KCl
20	0.000280	0.0370				conductivity	0.1M KCl		0.1M KCl
21	0.290	0.520				conductivity	0.1M KCl		0.1M KCl
22	0.380	0.670				conductivity	0.1M KCl		0.1M KCl
23	0.000220	0.0290				conductivity	0.1M KCl		0.1M KCl
24	0.000210	0.0290				conductivity	0.1M KCl		0.1M KCl
25	0.000280	0.0370				conductivity	0.1M KCl		0.1M KCl
26	0.370	0.670				conductivity	0.1M KCl		0.1M KCl
27	0.310	0.550				conductivity	0.1M KCl		0.1M KCl
28	0.290	0.520				conductivity	0.1M KCl		0.1M KCl
29	5.00e-05	0.00280				Through-diffusion	Distilled water	5.50	10%NaCl
30	9.20e-05	0.00510				Through-diffusion	Distilled water	5.50	10%NaCl
31	0.0120	0.0320	0.37	4.00e-11	-10.398	Through-diffusion	Distilled water	9.48	0.1M NaCl
32	0.0160	0.0430	0.37	5.60e-11	-10.252	Through-diffusion	Distilled water	9.54	0.1M NaCl
33	0.0290	0.0780	0.37	1.00e-10	-10.000	Through-diffusion	Distilled water	9.27	0.1M NaCl

	Contact time	Temperature(°C)	Eh(mV)	Others
0		25.0		aerobic
1		25.0		aerobic
2	596days	25.0		aerobic
3	11days	room		aerobic
4	11days	room		aerobic
5				aerobic
6				aerobic
7	596days	25.0		aerobic
8	90days	room		aerobic
9	90days	room		aerobic
10	18days	25.0		aerobic
11	18days	25.0		aerobic
12	18days	25.0		aerobic
13		25.0		aerobic
14		25.0		aerobic
15		room		aerobic
16		room		aerobic
17		room		aerobic
18		room		aerobic
19		room		aerobic
20		room		aerobic
21		room		aerobic
22		room		aerobic
23		room		aerobic
24		room		aerobic
25		room		aerobic
26		room		aerobic
27		room		aerobic
28		room		aerobic
29		25.0		aerobic
30		25.0		aerobic
31	18days	25.0		aerobic
32	18days	25.0		aerobic
33	18days	25.0		aerobic

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
34	Ni	6	Ni2+	2.0	Mudstone	2.78	1.80	1.00e-13	-13.000	6.60e-10
35	Ni	6	Ni2+	2.0	Mudstone			1.00e-13	-13.000	6.60e-10
36	Se	7	SeO32-	-2.0	Tuff	1.50	37.4	6.40e-12	-11.194	8.10e-10
37	Se	7	SeO32-	-2.0	Tuff	1.50	37.4	2.70e-12	-11.569	8.10e-10
38	Se	7	SeO32-	-2.0	Tuff	1.50	37.4	5.90e-12	-11.229	8.10e-10
39	Se	7	SeO32-	-2.0	Tuff	1.50	37.4	5.60e-12	-11.252	8.10e-10
40	Se	7	SeO32-	-2.0	Tuff	1.50	37.4	6.00e-12	-11.222	8.10e-10
41	Se	7	SeO32-	-2.0	Tuff	1.50	37.4	6.90e-12	-11.161	8.10e-10
42	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	6.42e-12	-11.192	8.10e-10
43	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	5.57e-12	-11.254	8.10e-10
44	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	6.20e-12	-11.208	8.10e-10
45	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	6.20e-12	-11.208	8.10e-10
46	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	8.80e-12	-11.056	8.10e-10
47	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	3.40e-12	-11.469	8.10e-10
48	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	7.90e-12	-11.102	8.10e-10
49	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	5.90e-12	-11.229	8.10e-10
50	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	6.40e-12	-11.194	8.10e-10
51	Se	7	SeO32-	-2.0	Tuff	1.53	37.3	7.40e-12	-11.131	8.10e-10
52	Sm	8	Sm3+	2.0	Mudstone	2.78	1.80	5.10e-14	-13.276	6.10e-10
53	Sm	8	Sm3+	2.0	Mudstone			5.10e-14	-13.276	6.10e-10
54	U	9	carbonate complex	-4.0	Tuff					7.20e-10
55	U	9	carbonate complex	-4.0	Tuff					7.20e-10
56	U	9	carbonate complex	-4.0	Tuff					7.20e-10
57	U	9	carbonate complex	-4.0	Tuff					7.20e-10



	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH	Tracer
34	0.000150	0.00840	0.33	3.00e-13	-12.523	Through-diffusion	Distilled water	6.00	0.1M NiCl <sub>2</sub>
35	0.000150	0.00840	0.33	3.00e-13	-12.523	Through-diffusion	Distilled water	6.00	0.1M NiCl <sub>2</sub>
36	0.00790	0.0210	1.8	3.58e-12	-11.446	Through-diffusion	0.1M NaCl	7.90	e-3M SeO <sub>2</sub>
37	0.00330	0.00890	0.52	5.19e-12	-11.285	Through-diffusion	0.1M NaCl	7.80	e-3M SeO <sub>2</sub>
38	0.00730	0.0190	1.7	3.51e-12	-11.455	Through-diffusion	0.1M NaCl	7.70	e-3M SeO <sub>2</sub>
39	0.00690	0.0180	0.59	9.49e-12	-11.003	Through-diffusion	0.1M NaCl	11.0	e-3M SeO <sub>2</sub>
40	0.00740	0.0200	0.57	1.05e-11	-10.979	Through-diffusion	0.1M NaCl	11.0	e-3M SeO <sub>2</sub>
41	0.00850	0.0230	0.94	7.34e-12	-11.134	Through-diffusion	0.1M NaCl	11.0	e-3M SeO <sub>2</sub>
42	0.00790	0.0210	1.8	3.59e-12	-11.445	Through-diffusion	0.1M NaCl	8.00	
43	0.00680	0.0180	0.59	9.44e-12	-11.025	Through-diffusion	0.1M NaCl	11.0	
44	0.00770	0.0210	1.7	3.65e-12	-11.438	Through-diffusion	0.1M NaCl	8.00	
45	0.00770	0.0210	0.70	8.86e-12	-11.053	Through-diffusion	0.1M NaCl	11.0	
46	0.0110	0.0290	2.3	3.83e-12	-11.417	Through-diffusion	0.1M NaCl	7.90	e-3M SeO <sub>2</sub>
47	0.00420	0.0110	1.5	2.27e-12	-11.644	Through-diffusion	0.1M NaCl	7.80	e-3M SeO <sub>2</sub>
48	0.00980	0.0260	2.1	3.76e-12	-11.425	Through-diffusion	0.1M NaCl	7.70	e-3M SeO <sub>2</sub>
49	0.00730	0.0200	0.62	9.52e-12	-11.021	Through-diffusion	0.1M NaCl	11.0	e-3M SeO <sub>2</sub>
50	0.00790	0.0210	0.61	1.05e-11	-10.979	Through-diffusion	0.1M NaCl	11.0	e-3M SeO <sub>2</sub>
51	0.00910	0.0240	0.96	7.71e-12	-11.113	Through-diffusion	0.1M NaCl	11.0	e-3M SeO <sub>2</sub>
52	0.000840	0.00460	0.20	2.65e-13	-12.577	Through-diffusion	Distilled water	6.00	0.1M SmCl <sub>3</sub>
53	0.000840	0.00460	0.20	2.60e-13	-12.585	Through-diffusion	Distilled water	6.00	0.1M SmCl <sub>3</sub>
54				5.00e-12	-11.301	In-diffusion	Deionized water		Uranyl nitrate
55				1.00e-12	-12.000	In-diffusion	groundwater		1450ppm U-233
56				8.00e-13	-12.097	In-diffusion	groundwater		1450ppm U-233
57				6.00e-11	-10.222	In-diffusion	groundwater		1450ppm U-233

	Contact time	Temperature(°C)	Eh(mV)	Others
34		25.0		aerobic
35		25.0		aerobic
36	about 100days	Temp. in G.B.		anaerobic
37	about 100days	Temp. in G.B.		anaerobic
38	about 100days	Temp. in G.B.		anaerobic
39	about 100days	Temp. in G.B.		anaerobic
40	about 100days	Temp. in G.B.		anaerobic
41	about 100days	Temp. in G.B.		anaerobic
42		22.0		anaerobic
43		22.0		anaerobic
44		22.0		anaerobic
45		22.0		anaerobic
46	about 100days	Temp. in G.B.		anaerobic
47	about 100days	Temp. in G.B.		anaerobic
48	about 100days	Temp. in G.B.		anaerobic
49	about 100days	Temp. in G.B.		anaerobic
50	about 100days	Temp. in G.B.		anaerobic
51	about 100days	Temp. in G.B.		anaerobic
52		25.0		aerobic
53		25.0		aerobic
54	30days	20~50		aerobic
55	14days	room		aerobic
56	14days	room		aerobic
57	14days	room		aerobic

## **4.2 Data Sheet for Sedimentary Rock (psammitic rock/sandy stone)**

The diffusivity data for I in sedimentary rock (psammitic rock/sandy stone) are shown.

ページ #1 - "砂質岩De/Da(l)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	l:De(m2/s)	llog De	Do(m2/s) 7)	Formation factor(FF) 8)	Geometric factor(Gf) 9)	Rock capacity factor
0		1	-	-1.0	siliceous sed. rock		4.30	6.10e-12	-11.215	2.00e-09	0.00310	0.0720	0.042
1		1	-	-1.0	siliceous sed. rock		10.4	1.20e-11	-10.921	2.00e-09	0.00600	0.0580	0.13
2		1	-	-1.0	siliceous sed. rock	2.39	10.4	1.20e-11	-10.921	2.00e-09	0.00600	0.0580	0.12
3		1	-	-1.0	siliceous sed. rock	1.60	30.7	6.00e-11	-10.222	2.00e-09	0.0300	0.0980	0.40
4		1	-	-1.0	siliceous sed. rock	1.59	30.7	6.10e-11	-10.215	2.00e-09	0.0310	0.100	0.42
5		1	-	-1.0	siliceous sed. rock	1.82	30.8	1.70e-10	-9.7700	2.00e-09	0.0850	0.280	0.37
6		1	-	-1.0	siliceous sed. rock	1.80	30.8	1.60e-10	-9.7960	2.00e-09	0.0800	0.260	0.45
7		1	-	-1.0	siliceous sed. rock	1.52	35.3	9.90e-11	-10.004	2.00e-09	0.0500	0.140	0.40
8		1	-	-1.0	siliceous sed. rock	1.52	35.3	6.60e-11	-10.180	2.00e-09	0.0330	0.0930	0.40
9		1	-	-1.0	siliceous sed. rock	1.25	47.5	2.40e-10	-9.6200	2.00e-09	0.120	0.250	0.60
10		1	-	-1.0	siliceous sed. rock	1.21	47.5	3.10e-10	-9.5090	2.00e-09	0.160	0.340	0.62

ページ #2 - "砂質岩De/Da(t)data"

	I:Da(m <sup>2</sup> /s)	I:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	1.45e-10	-9.8390	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 1)
1	9.23e-11	-10.035	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
2	1.00e-10	-10.000	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
3	1.50e-10	-9.8240	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 3)
4	1.45e-10	-9.8390	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 3)
5	4.59e-10	-9.3382	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 4)
6	3.56e-10	-9.4490	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 4)
7	2.48e-10	-9.8060	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 5)
8	1.65e-10	-9.7830	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 5)
9	4.00e-10	-9.3980	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 6)
10	5.00e-10	-9.3010	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 6)

ページ #1 - “砂質岩De/Da(l)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 西山勝栄、中嶋悟、多田隆治、内田隆：岩石間隙水中のイオンの拡散係数と間隙の性質、鉱山地質、40 (5)、323~336 (1990).  
Nishiyama, K., Nakashima, S., Tada, R., Uchida, T.: Diffusion of an ion in Rock Pore Water and Its Relation to Pore Characteristics, Mining Geology, 40 (5), pp.323-336 (1990).

Footnote:

- 1) The rock sample was sampled from Onnagawa, Akita-ken, Japan.  
This sedimentary rock (珪質堆積岩) is mainly composed of quartz and the composition of constituent quartz in the rock is quite high, what is called chert.  
The dry density was calculated based on the rock porosity, rock capacity factor and distribution coefficient reported in the literature.  
The Da was calculated from De and rock capacity factor reported in the literature.
- 2) The rock sample was sampled from Onnagawa, Akita-ken, Japan.  
This sedimentary rock (珪質堆積岩) is mainly composed of quartz, but is cemented with dolomite, what is called dolomatic siliceous rock.  
The dry density was calculated based on the rock porosity, rock capacity factor and distribution coefficient reported in the literature.  
The Da was calculated from De and rock capacity factor reported in the literature.
- 3) The rock sample was sampled from Onnagawa, Akita-ken, Japan.  
This sedimentary rock (珪質堆積岩) is mainly composed of opal CT.  
The dry density was calculated based on the rock porosity, rock capacity factor and distribution coefficient reported in the literature.  
The Da was calculated from De and rock capacity factor reported in the literature.
- 4) The rock sample was sampled from Onnagawa, Akita-ken, Japan.  
This sedimentary rock (珪質堆積岩) is mainly composed of quartz.  
The dry density was calculated based on the rock porosity, rock capacity factor and distribution coefficient reported in the literature.  
The Da was calculated from De and rock capacity factor reported in the literature.
- 5) The rock sample was sampled from Kamanosawa, Aomori-ken, Japan.  
This sedimentary rock (珪質堆積岩) is mainly composed of opal CT.  
The dry density was calculated based on the rock porosity, rock capacity factor and distribution coefficient reported in the literature.  
The Da was calculated from De and rock capacity factor reported in the literature.
- 6) The rock sample was sampled from Kamanosawa, Aomori-ken, Japan.  
This sedimentary rock (珪質堆積岩) is mainly composed of opal CT.

ページ #2 - “砂質岩De/Da(I)data”

The dry density was calculated based on the rock porosity, rock capacity factor and distribution coefficient reported in the literature.

The Da was calculated from De and rock capacity factor reported in the literature.

- 7) The  $D_o$  of I- is estimated  $2.0e-9$  m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169 (1997).].
- 8) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where  $De$  and  $Do$  is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 9) The geometric factor ( $G_f$ ) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f=D_p/Do=De/(\phi \cdot Do)=FF/\phi$ . Where  $D_p$  is the porewater diffusion coefficient,  $Do$  is the diffusion coefficient in free water,  $De$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

## ページ #1 - "砂質岩De/Da data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
0		1	-	-1.0	siliceous sed. rock		4.30	6.10e-12	-11.215	2.00e-09
1		1	-	-1.0	siliceous sed. rock		10.4	1.20e-11	-10.921	2.00e-09
2		1	-	-1.0	siliceous sed. rock	2.39	10.4	1.20e-11	-10.921	2.00e-09
3		1	-	-1.0	siliceous sed. rock	1.60	30.7	6.00e-11	-10.222	2.00e-09
4		1	-	-1.0	siliceous sed. rock	1.59	30.7	6.10e-11	-10.215	2.00e-09
5		1	-	-1.0	siliceous sed. rock	1.82	30.8	1.70e-10	-9.7700	2.00e-09
6		1	-	-1.0	siliceous sed. rock	1.80	30.8	1.60e-10	-9.7960	2.00e-09
7		1	-	-1.0	siliceous sed. rock	1.52	35.3	9.90e-11	-10.004	2.00e-09
8		1	-	-1.0	siliceous sed. rock	1.52	35.3	6.60e-11	-10.180	2.00e-09
9		1	-	-1.0	siliceous sed. rock	1.25	47.5	2.40e-10	-9.6200	2.00e-09
10		1	-	-1.0	siliceous sed. rock	1.21	47.5	3.10e-10	-9.5090	2.00e-09



## ページ #2 - "砂質岩De/Da data"

	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH	Tracer
0	0.00310	0.0720	0.042	1.45e-10	-9.8390	Through-diffusion	Deionized water		1M(l:127000ppm)KI
1	0.00600	0.0580	0.13	9.23e-11	-10.035	Through-diffusion	Deionized water		1M(l:127000ppm)KI
2	0.00600	0.0580	0.12	1.00e-10	-10.000	Through-diffusion	Deionized water		1M(l:127000ppm)KI
3	0.0300	0.0980	0.40	1.50e-10	-9.8240	Through-diffusion	Deionized water		1M(l:127000ppm)KI
4	0.0310	0.100	0.42	1.45e-10	-9.8390	Through-diffusion	Deionized water		1M(l:127000ppm)KI
5	0.0850	0.280	0.37	4.59e-10	-9.3382	Through-diffusion	Deionized water		1M(l:127000ppm)KI
6	0.0800	0.260	0.45	3.56e-10	-9.4490	Through-diffusion	Deionized water		1M(l:127000ppm)KI
7	0.0500	0.140	0.40	2.48e-10	-9.6060	Through-diffusion	Deionized water		1M(l:127000ppm)KI
8	0.0330	0.0930	0.40	1.65e-10	-9.7830	Through-diffusion	Deionized water		1M(l:127000ppm)KI
9	0.120	0.250	0.60	4.00e-10	-9.3980	Through-diffusion	Deionized water		1M(l:127000ppm)KI
10	0.160	0.340	0.62	5.00e-10	-9.3010	Through-diffusion	Deionized water		1M(l:127000ppm)KI

## ページ #3 - "砂質岩De/Da data"

	Contact time	Temperature(°C)	Eh(mV)	Others
0		room		aerobic
1		room		aerobic
2		room		aerobic
3		room		aerobic
4		room		aerobic
5		room		aerobic
6		room		aerobic
7		room		aerobic
8		room		aerobic
9		room		aerobic
10		room		aerobic

### **4.3 Data Sheet for Alkaline Crystalline Rock**

The diffusivity data for Cl, Cs, I, K, Na and Ni in alkaline crystalline rock are shown in turn.

ページ #1 - "塩基性結晶質De/Da(Cl)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Cl:De(m2/s)	Cl:log De	Do(m2/s) 7)	Formation factor(FF) 8)	Geometric factor(Gf) 9)	Rock capacity factor	Cl:Da(m2/s)
0	Cl	1	Cl-	-1.0	Basalt 1)	2.75	2.80	4.10e-14	-13.387	2.00e-09	2.10e-05	0.000730	0.0550	7.50e-14
1	Cl	1	Cl-	-1.0	Basalt 1)	2.75	2.80	1.20e-14	-13.921	2.00e-09	6.00e-08	0.000210		
2	Cl	1	Cl-	-1.0	Basalt 1)	2.75	2.80	5.70e-15	-14.244	2.00e-09	2.90e-06	1.00e-04		
3	Cl	1	Cl-	-1.0	Basalt 1)	2.75	2.80	7.10e-15	-14.149	2.00e-09	3.60e-06	0.000130		
4	Cl	1	Cl-	-1.0	Basalt		2.56	1.20e-14	-13.921	2.00e-09	6.00e-06	0.000230		
5	Cl	1	Cl-	-1.0	Basalt		2.56	5.70e-15	-14.244	2.00e-09	2.90e-06	0.000110		
6	Cl	1	Cl-	-1.0	Basalt		2.56	7.10e-15	-14.149	2.00e-09	3.60e-06	0.000140		
7	Cl	1	Cl-	-1.0	Basalt 2)			4.10e-14	-13.387	2.00e-09	2.10e-05		0.0550	7.50e-14
8	Cl	1	Cl-	-1.0	Basalt 1)		2.08	1.20e-14	-13.921	2.00e-09	6.00e-06	0.000290		
9	Cl	1	Cl-	-1.0	Basalt 1)		2.08	5.70e-15	-14.244	2.00e-09	2.90e-06	0.000140		
10	Cl	1	Cl-	-1.0	Basalt 1)		2.08	7.10e-15	-14.149	2.00e-09	3.60e-06	0.000170		

ページ #2 - "塩基性結晶質De/Da(Cl)data"

	Cl:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	-13.125	1	Through-diffusion	Distilled water	7.10	0.6M Cl 3)	596days	25.0		aerobic 4)
1		1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 4)
2		1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 4)
3		1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 4)
4		2	Through-diffusion	Distilled water	5.50	60000ppmCl		probably room		aerobic 5)
5		2	Through-diffusion	Distilled water	5.50	60000ppmCl		probably room		aerobic 5)
6		2	Through-diffusion	Distilled water	5.50	60000ppmCl		probably room		aerobic 5)
7	-13.125	3	Through-diffusion	Distilled water	7.10	0.6M Cl 3)	about 600days	25.0		aerobic
8		4	Through-diffusion	Distilled water		60000ppmCl	90days	room		aerobic 6)
9		4	Through-diffusion	Distilled water		60000ppmCl	90days	room		aerobic 6)
10		4	Through-diffusion	Distilled water		60000ppmCl	90days	room		aerobic 6)

ページ #1 - “塩基性結晶質De/Da(Cl)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V. (1997).
- 2 佐藤治夫、油井三和：岩石中のイオンの拡散係数の測定、日本原子力学会1992年秋の大会、F15、p.323 (1992).  
Sato, H., Yui, M.: Measurement of Ionic Diffusion Coefficients in Rocks, 1992 Fall Meeting of the Atomic Energy Society of Japan, F15, p.323 (1992).
- 3 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor Nuclear Fuel Development Corporation (1996. 12)(1996).
- 4 佐藤治夫、油井三和、石川博久：岩石の間隙構造に関する研究－岩石の間隙率およびNaClを用いたThrough-diffusion(透過拡散)試験による間隙因子の測定－、動燃技術資料、PNC TN8410 92-222、動燃事業団(1992. 8)(1992).  
Sato, H., Yui, M., Ishikawa, H.: A Study on Pore Structure of Rocks, PNC Technical Report, PNC TN8410 92-222, Power Reactor Nuclear Fuel Development Corporation (1992. 8)(1992).

Footnote:

- 1) The rock sample was sampled from Kitamatsuura-gun, Nagasaki-ken, Japan. This rock is approximately equivalent to a standard sample in the chemical composition of Japanese basalt in the Geological Survey of Japan.
- 2) The rock sample was sampled from Kitamatsuura-gun, Nagasaki-ken, Japan.
- 3) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water. The concentration of Cl<sup>-</sup> was calculated as a counter ion of Cs<sup>+</sup>, Ni<sup>2+</sup> and Sm<sup>3+</sup> in the tracer. Since the concentration of each cation was 0.1 M, the concentration of Cl<sup>-</sup> is estimated to be 0.6 M.
- 4) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. The porosity and density shown are average values of respective measured values.
- 5) The De values were originally obtained to derive formation factor and

ページ #2 - “塩基性結晶質De/Da(Cl) data”

tortuosity of the rock.

The porosity was determined by water saturation method (water intrusion method). The porosity shown is an average value of the measured ones.

- 6) The De values were originally obtained to derive formation factor and tortuosity of the rock.

The porosity was determined by two kinds of water saturation methods (water intrusion methods). The porosity shown is an average value of the measured ones.

- 7) The Do of Cl<sup>-</sup> is estimated 2.0e-9 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169 (1997).].

- 8) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.

- 9) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\Phi \cdot Do)=FF/\Phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\Phi$  is the porosity and FF is the formation factor.

ページ #1 - "塩基性結晶質De/Da(Cs)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Cs:De(mZ/s)	Cs:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor	Cs:Da(mZ/s)
0	Cs	2	Cs+	1.0	Basalt 1)	2.75	2.80	1.50e-13	-12.824	2.10e-09	7.10e-05	0.00260	0.710	2.10e-13
1	Cs	2	Cs+	1.0	Basalt			1.50e-13	-12.824	2.10e-09	7.10e-05	0.00260	0.710	2.10e-13



ページ #2 - "塩基性結晶質De/Da(Cs)data"

	Cs:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	-12.678	1	Through-diffusion	Distilled water	7.10	0.1M CsCl 2)	596days	25.0		aerobic 3)
1	-12.678	2	Through-diffusion	Distilled water	7.10	0.1M CsCl 2)		25.0		aerobic 3)

ページ #1 - “塩基性結晶質De/Da(Cs)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V.(1997).
- 2 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN 1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

Footnote:

- 1) The rock sample was sampled from Kitamatsuura-gun, Nagasaki-ken, Japan. This rock is approximately equivalent to a standard sample in the chemical composition of Japanese basalt in the Geological Survey of Japan.
- 2) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water.
- 3) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry. The porosity and density shown are average values of respective measured ones.
- 4) The Do of Cs<sup>+</sup> is estimated 2.1e-9 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168 (1997).].
- 5) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\Phi \cdot Do)=FF/\Phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\Phi$  is the porosity and FF is the formation factor.

ページ #1 - "塩基性結晶質De/Da(I)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm <sup>3</sup> )	Porosity(%)	I:De(m <sup>2</sup> /s)	I:log De	Do(m <sup>2</sup> /s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor	I:Da(m <sup>2</sup> /s)
0		3	-	-1.0	Andesite 1)		3.00	3.60e-14	-13.444	2.00e-09	1.80e-05	0.000800	0.073	4.90e-13
1		3	-	-1.0	Andesite 1)		3.00	4.00e-14	-13.398	2.00e-09	2.00e-05	0.000870	0.13	3.10e-13
2		3	-	-1.0	Andesite 1)		12.1	1.50e-12	-11.824	2.00e-09	0.000750	0.00820	0.13	1.20e-11
3		3	-	-1.0	Andesite 1)		12.1	7.10e-12	-11.148	2.00e-09	0.00360	0.0300	0.016	4.40e-10
4		3	-	-1.0	Andesite 1)		14.5	3.70e-12	-11.432	2.00e-09	0.00190	0.0130	0.16	2.30e-11
5		3	-	-1.0	Andesite 1)		14.5	8.70e-12	-11.060	2.00e-09	0.00440	0.0300	0.12	7.30e-11
6		3	-	-1.0	Schist 3)		0.500	3.70e-14	-13.432	2.00e-09	1.90e-05	0.00370	0.050	7.40e-13
7		3	-	-1.0	Schist 3)		0.500	5.80e-13	-12.237	2.00e-09	0.000290	0.0580	0.050	1.20e-11

ページ #2 - "塩基性結晶質De/Da(I)data"

	I/log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	-12.310	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
1	-12.509	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
2	-10.921	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
3	-9.3570	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
4	-10.638	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
5	-10.137	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
6	-12.131	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)
7	-10.921	1	Through-diffusion	Deionized water		1M(I:127000ppm)KI		room		aerobic 2)

ページ #1 - “塩基性結晶質De/Da(l) data”

REFERENCE AND FOOTNOTE

Reference:

- 1 西山勝栄、中嶋悟、多田隆治、内田隆：岩石間隙水中のイオンの拡散係数と間隙の性質、鉱山地質、40(5)、323~336(1990).  
Nishiyama, K., Nakashima, S., Tada, R., Uchida, T.: Diffusion of an Ion in Rock Pore Water and Its Relation to Pore Characteristics, Mining Geology, 40 (5), pp.323-336 (1990).

Footnote:

- 1) The andesite sample was sampled from 3 points (places) in Kushikino, Kagoshima-ken, Japan.
- 2) The rock porosity was measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry to discuss effective porosity for transport.
- 3) The schist sample was sampled from Iwaki, Fukushima-ken, Japan.
- 4) The  $D_0$  of I- is estimated  $2.0 \times 10^{-9}$  m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169 (1997).].
- 5) The formation factor (FF) was calculated based on the equation:  $FF = D_e / D_0$ . Where  $D_e$  and  $D_0$  is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 6) The geometric factor ( $G_f$ ) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f = D_p / D_0 = D_e / (\phi \cdot D_0) = FF / \phi$ . Where  $D_p$  is the porewater diffusion coefficient,  $D_0$  is the diffusion coefficient in free water,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "塩基性結晶質De/Da(K)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	K:De(m2/s)	K:log De	Do(m2/s) 5)	Formation factor(FF) 6)	Geometric factor(Gf) 7)	Rock capacity factor	K:Da(m2/s)
0	K	4	K+	1.0	Basalt 1)	2.90	2.08	4.60e-13	-12.337	2.00e-09	0.000230	0.0110		
1	K	4	K+	1.0	Basalt 1)	2.90	2.08	2.50e-12	-11.602	2.00e-09	0.00130	0.0600		
2	K	4	K+	1.0	Basalt 1)	2.90	2.08	2.50e-12	-11.602	2.00e-09	0.00130	0.0600		
3	K	4	K+	1.0	Basalt 1)	2.90	2.08	4.60e-13	-12.337	2.00e-09	0.000230	0.0110		
4	K	4	K+	1.0	Basalt 1)	2.90	2.08	1.50e-12	-11.824	2.00e-09	0.000750	0.0360		
5	K	4	K+	1.0	Basalt 1)	2.90	2.08	1.90e-12	-11.721	2.00e-09	0.000950	0.0460		

ページ #2 - "塩基性結晶質De/Da(K)data"

	Klog Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0		1	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
1		1	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
2		2	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
3		2	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
4		2	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
5		2	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)

ページ #1 - “塩基性結晶質De/Da(K)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Conca, J. L., Apted, M. J., Arthur, R. C.: Direct Determinations of Transport Parameters in Repository Materials, Scientific Basis for Nuclear Waste Management XVI (Interrante, C. G. and Pabalan, R. T., eds.), Materials Research Society Symposium Proceedings, Vol. 294, pp.839-844, Materials Research Society, Pittsburgh, Pennsylvania (1993).
- 2 Conca, J. L., Wright, J.: Direct Determinations of Unsaturated Flow and Transport, Proceedings of the Twelfth Annual American Geophysical Union Hydrology Days Meeting, Fort Collins, March 1992, pp.103-116 (1992).

Footnote:

- 1) The basalt sample was sampled from Kitamatsuura-gun, Nagasaki-ken, Japan. This rock is a standard sample in the chemical composition of Japanese basalt in the Geological Survey of Japan.
- 2) The De values were determined by electrical conductivity measurements, based on the Nernst-Einstein equation. The De values were determined using the unsaturated flow apparatus (UFA). Hydraulic conductivity can also be determined by this method.
- 3) The rock sample was saturated with a 0.1 M KCl solution and then electrical conductivity was measured. The porewater and tracer are therefore the same.
- 4) Apparent diffusion coefficient (Da) can not be obtained from the electrical conductivity measurements. Since the porosity was not reported in these literatures, it was quoted from another literature which was published by PNC (H. Sato and T. Shibutani, PNC Technical Report, PNC TN8410 94-284, pp.71-89 (1994).). Because PNC supplied the sample and measured the porosity by water saturation method (water intrusion method).
- 5) The Do of K<sup>+</sup> is estimated 2.0e-9 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168 (1997).].
- 6) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 7) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\Phi \cdot Do)=FF/\Phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\Phi$  is the porosity and FF is the formation factor.



ページ #1 - "塩基性結晶質De/Da(Na)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Na:De(m2/s)	Na:log De	Do(m2/s) 3)	Formation factor(FF) 4)	Geometric factor(Gf) 5)	Rock capacity factor	Na:Da(m2/s)
0	Na	5	Na+	1.0	Basalt 1)	2.75	2.80	2.20e-14	-13.658	1.30e-09	1.70e-05	0.000610		
1	Na	5	Na+	1.0	Basalt 1)	2.75	2.80	1.80e-14	-13.745	1.30e-09	9.00e-06	0.000320		
2	Na	5	Na+	1.0	Basalt 1)	2.75	2.80	2.20e-14	-13.658	1.30e-09	1.70e-05	0.000610		

ページ #2 - "塩溶性結晶質De/Da(Na)data"

	Na:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0		1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 2)
1		1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 2)
2		1	Through-diffusion	Distilled water	5.50	10%NaCl		25.0		aerobic 2)

ページ #1 - “塩基性結晶質De/Da(Na)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V.(1997).

Footnote:

- 1) The rock sample was sampled from Kitamatsuura-gun, Nagasaki-ken, Japan. This rock is a standard sample in the chemical composition of Japanese basalt in the Geological Survey of Japan.
- 2) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry to discuss effective porosity for transport. The porosity and density shown are average values of respective measured values.  
The De values were originally obtained to derive formation factor and tortuosity of the rock.
- 3) The Do of Na+ is estimated  $1.3 \times 10^{-9}$  m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168 (1997).].
- 4) The formation factor (FF) was calculated based on the equation:  $FF = De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 5) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf = Dp/Do = De/(\phi \cdot Do) = FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "塩基性結晶質De/Da(Ni)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Ni:De(m2/s)	Ni:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor	Ni:Da(m2/s)
0	Ni	8	Ni2+	2.0	Basalt 1)	2.75	2.80	2.80e-14	-13.553	6.60e-10	4.20e-05	0.00150	0.220	1.30e-14
1	Ni	8	Ni2+	2.0	Basalt			2.80e-14	-13.553	6.60e-10	4.20e-05		0.220	1.30e-14

ページ #2 - "塩基性結晶質De/Da(Ni)data"

	Ni:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	-13.886	1	Through-diffusion	Distilled water	7.10	0.1M NiCl <sub>2</sub> 2)	596days	25.0		aerobic 3)
1	-13.886	2	Through-diffusion	Distilled water	7.10	0.1M NiCl <sub>2</sub> 2)		25.0		aerobic 3)

ページ #1 - “塩基性結晶質De/Da(Ni)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V.(1997).
- 2 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN 1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

Footnote:

- 1) The rock sample was sampled from Kitamatsuura-gun, Nagasaki-ken, Japan. This rock is a standard sample in the chemical composition of Japanese basalt in the Geological Survey of Japan.
- 2) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water.
- 3) The porosity and density were measured by water saturation method (water intrusion method) and mercury porosimetry. Pore-size distribution was also determined by mercury porosimetry to discuss effective porosity for transport. The porosity and density shown are average values of respective measured ones.
- 4) The Do of Ni<sup>2+</sup> is estimated 6.6e-10 m<sup>2</sup>/s at 25°C [Marcus, Y.: Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168 (1997).].
- 5) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and diffusion coefficient in free water, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the diffusion coefficient in free water, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "塩基性結晶質岩De/Da data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)	Formation factor(FF)
0	Cl	1	Cl-	-1.0	Basalt	2.75	2.80	4.10e-14	-13.387	2.00e-09	2.10e-05
1	Cl	1	Cl-	-1.0	Basalt	2.75	2.80	1.20e-14	-13.921	2.00e-09	6.00e-06
2	Cl	1	Cl-	-1.0	Basalt	2.75	2.80	5.70e-15	-14.244	2.00e-09	2.90e-06
3	Cl	1	Cl-	-1.0	Basalt	2.75	2.80	7.10e-15	-14.149	2.00e-09	3.60e-06
4	Cl	1	Cl-	-1.0	Basalt		2.56	1.20e-14	-13.921	2.00e-09	6.00e-06
5	Cl	1	Cl-	-1.0	Basalt		2.56	5.70e-15	-14.244	2.00e-09	2.90e-06
6	Cl	1	Cl-	-1.0	Basalt		2.56	7.10e-15	-14.149	2.00e-09	3.60e-06
7	Cl	1	Cl-	-1.0	Basalt			4.10e-14	-13.387	2.00e-09	2.10e-05
8	Cl	1	Cl-	-1.0	Basalt		2.08	1.20e-14	-13.921	2.00e-09	6.00e-06
9	Cl	1	Cl-	-1.0	Basalt		2.08	5.70e-15	-14.244	2.00e-09	2.90e-06
10	Cl	1	Cl-	-1.0	Basalt		2.08	7.10e-15	-14.149	2.00e-09	3.60e-06
11	Cs	2	Cs+	1.0	Basalt	2.75	2.80	1.50e-13	-12.824	2.10e-09	7.10e-05
12	Cs	2	Cs+	1.0	Basalt			1.50e-13	-12.824	2.10e-09	7.10e-05
13	I	3	I-	-1.0	Andesite		3.00	3.60e-14	-13.444	2.00e-09	1.80e-05
14	I	3	I-	-1.0	Andesite		3.00	4.00e-14	-13.398	2.00e-09	2.00e-05
15	I	3	I-	-1.0	Andesite		12.1	1.50e-12	-11.824	2.00e-09	0.000750
16	I	3	I-	-1.0	Andesite		12.1	7.10e-12	-11.149	2.00e-09	0.00360
17	I	3	I-	-1.0	Andesite		14.5	3.70e-12	-11.432	2.00e-09	0.00190
18	I	3	I-	-1.0	Andesite		14.5	8.70e-12	-11.060	2.00e-09	0.00440
19	I	3	I-	-1.0	Schist		0.500	3.70e-14	-13.432	2.00e-09	1.90e-05
20	I	3	I-	-1.0	Schist		0.500	5.80e-13	-12.237	2.00e-09	0.000290
21	K	4	K+	1.0	Basalt	2.90	2.08	4.60e-13	-12.337	2.00e-09	0.000230
22	K	4	K+	1.0	Basalt	2.90	2.08	2.50e-12	-11.602	2.00e-09	0.00130
23	K	4	K+	1.0	Basalt	2.90	2.08	2.50e-12	-11.602	2.00e-09	0.00130
24	K	4	K+	1.0	Basalt	2.90	2.08	4.60e-13	-12.337	2.00e-09	0.000230
25	K	4	K+	1.0	Basalt	2.90	2.08	1.50e-12	-11.824	2.00e-09	0.000750
26	K	4	K+	1.0	Basalt	2.90	2.08	1.90e-12	-11.721	2.00e-09	0.000950
27	Na	5	Na+	1.0	Basalt	2.75	2.80	2.20e-14	-13.658	1.30e-09	1.70e-05
28	Na	5	Na+	1.0	Basalt	2.75	2.80	1.80e-14	-13.745	1.30e-09	9.00e-06
29	Na	5	Na+	1.0	Basalt	2.75	2.80	2.20e-14	-13.658	1.30e-09	1.70e-05
30	Ni	6	Ni2+	2.0	Basalt	2.75	2.80	2.80e-14	-13.553	6.60e-10	4.20e-05
31	Ni	6	Ni2+	2.0	Basalt			2.80e-14	-13.553	6.60e-10	4.20e-05

ページ #2 - "塩基性結晶質岩De/Da data"

	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH	Tracer	Contact time
0	0.000730	0.0550	7.50e-14	-13.125	Through-diffusion	Distilled water	7.10	0.6M Cl	596days
1	0.000210				Through-diffusion	Distilled water	5.50	10%NaCl	
2	1.00e-04				Through-diffusion	Distilled water	5.50	10%NaCl	
3	0.000130				Through-diffusion	Distilled water	5.50	10%NaCl	
4	0.000230				Through-diffusion	Distilled water	5.50	60000ppmNaCl	
5	0.000110				Through-diffusion	Distilled water	5.50	60000ppmNaCl	
6	0.000140				Through-diffusion	Distilled water	5.50	60000ppmNaCl	
7		0.0550	7.50e-14	-13.125	Through-diffusion	Distilled water	7.10	0.6M Cl	about 600days
8	0.000290				Through-diffusion	Distilled water		60000ppmNaCl	90days
9	0.000140				Through-diffusion	Distilled water		60000ppmNaCl	90days
10	0.000170				Through-diffusion	Distilled water		60000ppmNaCl	90days
11	0.00260	0.710	2.10e-13	-12.678	Through-diffusion	Distilled water	7.10	0.1M CsCl	596days
12	0.00260	0.710	2.10e-13	-12.678	Through-diffusion	Distilled water	7.10	0.1M CsCl	
13	0.000600	0.0730	4.90e-13	-12.310	Through-diffusion	Deionized water		1M(I:127000ppm)KI	
14	0.000670	0.130	3.10e-13	-12.509	Through-diffusion	Deionized water		1M(I:127000ppm)KI	
15	0.00620	0.130	1.20e-11	-10.921	Through-diffusion	Deionized water		1M(I:127000ppm)KI	
16	0.0300	0.0160	4.40e-10	-9.3570	Through-diffusion	Deionized water		1M(I:127000ppm)KI	
17	0.0130	0.160	2.30e-11	-10.638	Through-diffusion	Deionized water		1M(I:127000ppm)KI	
18	0.0300	0.120	7.30e-11	-10.137	Through-diffusion	Deionized water		1M(I:127000ppm)KI	
19	0.00370	0.0500	7.40e-13	-12.131	Through-diffusion	Deionized water		1M(I:127000ppm)KI	
20	0.0580	0.0500	1.20e-11	-10.921	Through-diffusion	Deionized water		1M(I:127000ppm)KI	
21	0.0110				conductivity	0.1M KCl		0.1M KCl	
22	0.0600				conductivity	0.1M KCl		0.1M KCl	
23	0.0600				conductivity	0.1M KCl		0.1M KCl	
24	0.0110				conductivity	0.1M KCl		0.1M KCl	
25	0.0360				conductivity	0.1M KCl		0.1M KCl	
26	0.0460				conductivity	0.1M KCl		0.1M KCl	
27	0.000610				Through-diffusion	Distilled water	5.50	10%NaCl	
28	0.000320				Through-diffusion	Distilled water	5.50	10%NaCl	
29	0.000610				Through-diffusion	Distilled water	5.50	10%NaCl	
30	0.00150	0.220	1.30e-14	-13.886	Through-diffusion	Distilled water	7.10	0.1M NiCl <sub>2</sub>	596days
31		0.220	1.30e-14	-13.886	Through-diffusion	Distilled water	7.10	0.1M NiCl <sub>2</sub>	



ページ #3 - "塩基性結晶質岩De/Da data"

	Temperature(°C)	Eh(mV)	Others
0	25.0		aerobic
1	25.0		aerobic
2	25.0		aerobic
3	25.0		aerobic
4	probably room		aerobic
5	probably room		aerobic
6	probably room		aerobic
7	25.0		aerobic
8	room		aerobic
9	room		aerobic
10	room		aerobic
11	25.0		aerobic
12	25.0		aerobic
13	room		aerobic
14	room		aerobic
15	room		aerobic
16	room		aerobic
17	room		aerobic
18	room		aerobic
19	room		aerobic
20	room		aerobic
21	probably room		aerobic
22	probably room		aerobic
23	probably room		aerobic
24	probably room		aerobic
25	probably room		aerobic
26	probably room		aerobic
27	25.0		aerobic
28	25.0		aerobic
29	25.0		aerobic
30	25.0		aerobic
31	25.0		aerobic

#### **4.4 Data Sheet for Acid Crystalline Rock**

The diffusivity data for Ba, Be, Cl, Co, Cs, HTO, I, K, Mg, Na, Ni, Np, Pu, Se, Sm, Sr, Tc, U and uranium in acid crystalline rock are shown in turn.

ページ #1 - "酸性結晶質岩De/Da(Ba)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Ba:De(m2/s)	Ba:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor	Ba:Da(m2/s)
0	Ba	1	Ba2+	2.0	Granite 1)		0.680	2.10e-13	-12.878	8.50e-10	0.000250	0.0360		
1	Ba	1	Ba2+	2.0	Granite 1)		0.680	3.50e-13	-12.456	8.50e-10	0.000410	0.0610		
2	Ba	1	Ba2+	2.0	Granite 1)		0.680	3.00e-13	-12.523	8.50e-10	0.000350	0.0520		
3	Ba	1	Ba2+	2.0	Granite 1)		0.680	2.50e-13	-12.602	8.50e-10	0.000290	0.0430		
4	Ba	1	Ba2+	2.0	Granite 1)		0.680	2.90e-13	-12.538	8.50e-10	0.000340	0.0500		
5	Ba	1	Ba2+	2.0	Granite 1)		0.680	1.90e-13	-12.721	8.50e-10	0.000220	0.0330		

ページ #2 - "酸性結晶質岩De/Da(Ba)data"

	Balog Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0		1	Through-diffusion	0.1M KCl 3)	4.00	Ba(NO3)2 2)		25.0		aerobic
1		1	Through-diffusion	0.1M KCl 3)	4.00	Ba(NO3)2 2)		25.0		aerobic
2		1	Through-diffusion	0.1M KCl 3)	4.00	Ba(NO3)2 2)		25.0		aerobic
3		1	Through-diffusion	0.1M KCl 3)	4.00	Ba(NO3)2 2)		25.0		aerobic
4		1	Through-diffusion	0.1M KCl 3)	4.00	Ba(NO3)2 2)		25.0		aerobic
5		1	Through-diffusion	0.1M KCl 3)	4.00	Ba(NO3)2 2)		25.0		aerobic

ページ #1 - “酸性結晶質岩De/Da(Ba)data”

---

REFERENCE AND FOOTNOTE

Reference:

- 1 Yamaguchi, T., Sakamoto, Y., Senoo, M.: Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796-803, the Atomic Energy Society of Japan (August 1993) (1993).

Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. This kind of rock, regarded as one of the candidate host rocks for repository, is major host rock in Japan.
- 2) The tracer was used as a mixture of Ba(NO<sub>3</sub>)<sub>2</sub>, BeSO<sub>4</sub>, CoCl<sub>2</sub>, MgCl<sub>2</sub>, NiCl<sub>2</sub> and Sr(NO<sub>3</sub>)<sub>2</sub>, what we call as a multi-tracer. Individual concentrations in the tracer are not reported.
- 3) This solution (0.1 M KCl) was used to keep the ionic strength of the porewater in both cells.
- 4) The Do of Ba<sup>2+</sup> is estimated 8.5e-10 m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 5) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "酸性結晶質岩De/Da(Be)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Be:De(m2/s)	Be:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor	Be:Da(m2/s)
0	Be	2	Be2+	2.0	Granite 1)		0.680	3.00e-13	-12.523	6.00e-10	5.00e-05	0.00740		
1	Be	2	Be2+	2.0	Granite 1)		0.680	3.30e-13	-12.481	6.00e-10	5.50e-05	0.00810		

ページ #2 - "酸性結晶質岩De/Da(Be)data"

	Be:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0		1	Through-diffusion	0.1M KCl 3)	4.00	BeSO <sub>4</sub> 2)		25.0		aerobic
1		1	Through-diffusion	0.1M KCl 3)	4.00	BeSO <sub>4</sub> 2)		25.0		aerobic

ページ #1 - “酸性結晶質岩De/Da(Be)data”

---

REFERENCE AND FOOTNOTE

Reference:

- 1 Yamaguchi, T., Sakamoto, Y., Senoo, M.: Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796-803, the Atomic Energy Society of Japan (August 1993) (1993).

Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. This kind of rock, regarded as one of the candidate host rocks for repository, is major host rock in Japan.
- 2) The tracer was used as a mixture of Ba(NO<sub>3</sub>)<sub>2</sub>, BeSO<sub>4</sub>, CoCl<sub>2</sub>, MgCl<sub>2</sub>, NiCl<sub>2</sub> and Sr(NO<sub>3</sub>)<sub>2</sub>, what we call as a multi-tracer. Individual concentrations in the tracer are not reported.
- 3) This solution (0.1 M KCl) was used to keep the ionic strength of the porewater in both cells.
- 4) The Do of Be<sup>2+</sup> is estimated 6.0e-10 m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 5) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.



ページ #1 - "融性結晶質岩De/Da(Cl)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Cl:De(m2/s)	Cl:log De	Do(m2/s) 7)	Formation factor(FF) 8)	Geometric factor(Gf) 9)	Rock capacity factor
0	Cl 1)	3	Cl-	-1.0	Granodiorite		0.880	4.50e-13	-12.347	2.00e-09	0.000230	0.0260	
1	Cl 1)	3	Cl-	-1.0	Granodiorite		0.880	7.10e-13	-12.149	2.00e-09	0.000360	0.0410	
2	Cl 1)	3	Cl-	-1.0	Granodiorite		0.880	5.10e-13	-12.292	2.00e-09	0.000260	0.0300	
3	Cl 1)	3	Cl-	-1.0	Granodiorite		0.990	4.50e-13	-12.347	2.00e-09	0.000230	0.0230	
4	Cl 1)	3	Cl-	-1.0	Granodiorite		0.990	7.10e-13	-12.149	2.00e-09	0.000360	0.0360	
5	Cl 1)	3	Cl-	-1.0	Granodiorite		0.990	5.10e-13	-12.292	2.00e-09	0.000260	0.0260	
6	Cl 1)	3	Cl-	-1.0	Granodiorite	2.64	1.20	4.50e-13	-12.347	2.00e-09	0.000230	0.0190	
7	Cl 1)	3	Cl-	-1.0	Granodiorite	2.64	1.20	7.10e-13	-12.149	2.00e-09	0.000360	0.0300	
8	Cl 1)	3	Cl-	-1.0	Granodiorite	2.64	1.20	5.10e-13	-12.292	2.00e-09	0.000260	0.0220	
9	Cl	3	Cl-	-1.0	Granodiorite	2.64	1.20	7.40e-13	-12.131	2.00e-09	0.000370	0.0310	0.0370
10	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	1.70e-11	-10.770	2.00e-09	0.00850	0.370	3.00
11	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	8.10e-12	-11.092	2.00e-09	0.00410	0.180	1.20
12	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	4.80e-12	-11.319	2.00e-09	0.00240	0.100	0.280
13	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	1.40e-11	-10.854	2.00e-09	0.00700	0.220	2.50
14	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	6.90e-12	-11.161	2.00e-09	0.00350	0.110	1.30
15	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	3.90e-12	-11.409	2.00e-09	0.00200	0.0630	0.200
16	Cl	3	Cl-	-1.0	Fracture fillings	2.43	5.60	2.10e-11	-10.678	2.00e-09	0.0110	0.200	1.20
17	Cl	3	Cl-	-1.0	Fracture fillings	2.43	5.60	1.40e-11	-10.854	2.00e-09	0.00700	0.130	0.410
18	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	1.00e-11	-11.000	2.00e-09	0.00500	0.220	
19	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	8.30e-12	-11.081	2.00e-09	0.00420	0.130	
20	Cl	3	Cl-	-1.0	Fracture fillings	2.43	5.60	1.80e-11	-10.745	2.00e-09	0.00900	0.160	
21	Cl	3	Cl-	-1.0	Granodiorite		2.30	1.00e-11	-11.000	2.00e-09	0.00500	0.220	
22	Cl	3	Cl-	-1.0	Altered granodiorite		3.20	8.30e-12	-11.081	2.00e-09	0.00420	0.130	
23	Cl	3	Cl-	-1.0	Fracture fillings		5.60	1.80e-11	-10.745	2.00e-09	0.00900	0.160	
24	Cl	3	Cl-	-1.0	Granodiorite		1.00	7.41e-13	-12.130	2.00e-09	0.000370	0.0370	
25	Cl	3	Cl-	-1.0	Granite		0.990	4.50e-13	-12.347	2.00e-09	0.000230	0.0230	
26	Cl	3	Cl-	-1.0	Granite		0.990	7.10e-13	-12.149	2.00e-09	0.000360	0.0360	
27	Cl	3	Cl-	-1.0	Granite		0.990	5.10e-13	-12.292	2.00e-09	0.000260	0.0260	
28	Cl	3	Cl-	-1.0	Granodiorite 6)			7.40e-13	-12.131	2.00e-09	0.000370		0.140
29	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	1.00e-11	-11.000	2.00e-09	0.00500	0.220	
30	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	8.30e-12	-11.081	2.00e-09	0.00420	0.130	
31	Cl	3	Cl-	-1.0	Fracture fillings	2.43	5.60	1.80e-11	-10.745	2.00e-09	0.00900	0.160	

ページ #2 - "放射性結晶質岩De/Da(Cl)data"

	Cl:Da(m2/s)	Cl:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0			1	Through-diffusion	Distilled water		60000ppmCl(NaCl)	90days	room		aerobic
1			1	Through-diffusion	Distilled water		60000ppmCl(NaCl)	90days	room		aerobic
2			1	Through-diffusion	Distilled water		60000ppmCl(NaCl)	90days	room		aerobic
3			1	Through-diffusion	Distilled water		60000ppmCl(NaCl)	90days	room		aerobic
4			1	Through-diffusion	Distilled water		60000ppmCl(NaCl)	90days	room		aerobic
5			1	Through-diffusion	Distilled water		60000ppmCl(NaCl)	90days	room		aerobic
6			2	Through-diffusion	Distilled water	5.50	10%NaCl	90days	25.0		aerobic
7			2	Through-diffusion	Distilled water	5.50	10%NaCl	90days	25.0		aerobic
8			2	Through-diffusion	Distilled water	5.50	10%NaCl	90days	25.0		aerobic
9	2.00e-11	-10.700	2	Through-diffusion	Distilled water	5.80	0.6M Cl 3)		25.0		aerobic
10	5.60e-12	-11.252	3	Through-diffusion	groundwater 4)	8.70	0.06M(2127ppm) 5)	30days	25.0		aerobic 2)
11	6.70e-12	-11.174	3	Through-diffusion	groundwater 4)	8.70	0.06M(2127ppm) 5)	30days	25.0		aerobic 2)
12	1.70e-11	-10.770	3	Through-diffusion	groundwater 4)	8.70	0.06M(2127ppm) 5)	30days	25.0		aerobic 2)
13	5.70e-12	-11.244	3	Through-diffusion	groundwater 4)	8.77	0.06M(2127ppm) 5)	30days	25.0		aerobic 2)
14	5.50e-12	-11.260	3	Through-diffusion	groundwater 4)	8.77	0.06M(2127ppm) 5)	30days	25.0		aerobic 2)
15	2.00e-11	-10.699	3	Through-diffusion	groundwater 4)	8.77	0.06M(2127ppm) 5)	30days	25.0		aerobic 2)
16	1.80e-11	-10.745	3	Through-diffusion	groundwater 4)	8.92	0.06M(2127ppm) 5)	30days	25.0		aerobic 2)
17	3.40e-11	-10.469	3	Through-diffusion	groundwater 4)	8.92	0.06M(2127ppm) 5)	30days	25.0		aerobic 2)
18	9.80e-12	-11.009	4	Through-diffusion	groundwater	8.70	0.06M(2127ppm) 5)	30days	25.0		aerobic
19	1.00e-11	-11.000	4	Through-diffusion	groundwater	8.77	0.06M(2127ppm) 5)	30days	25.0		aerobic
20	2.60e-11	-10.585	4	Through-diffusion	groundwater	8.92	0.06M(2127ppm) 5)	30days	25.0		aerobic
21	9.80e-12	-11.009	5	Through-diffusion	groundwater	8.70			25.0		aerobic
22	1.00e-11	-11.000	5	Through-diffusion	groundwater	8.77			25.0		aerobic
23	2.60e-11	-10.585	5	Through-diffusion	groundwater	8.92			25.0		aerobic
24	2.00e-11	-10.699	6	Through-diffusion	Distilled water	6.00	0.6M Cl 3)		20.0		aerobic
25			7	Through-diffusion	Distilled water	5.50	60000ppmCl(NaCl)		probably room		aerobic
26			7	Through-diffusion	Distilled water	5.50	60000ppmCl(NaCl)		probably room		aerobic
27			7	Through-diffusion	Distilled water	5.50	60000ppmCl(NaCl)		probably room		aerobic
28	2.00e-11	-10.699	8	Through-diffusion	Distilled water	5.80	0.6M Cl 3)		25.0		aerobic
29	9.80e-12	-11.009	9	Through-diffusion	groundwater	8.70	0.06M(2127ppm) 5)	30days	25.0		aerobic
30	1.00e-11	-11.000	9	Through-diffusion	groundwater	8.77	0.06M(2127ppm) 5)	30days	25.0		aerobic
31	2.60e-11	-10.585	9	Through-diffusion	groundwater	8.92	0.06M(2127ppm) 5)	30days	25.0		aerobic

## ページ #1 - “酸性結晶質岩De/Da(Cl)data”

## REFERENCE AND FOOTNOTE

## Reference:

- 1 佐藤治夫、油井三和、石川博久：岩石の間隙構造に関する研究－岩石の間隙率およびNaClを用いたThrough-diffusion(透過拡散)試験による間隙因子の測定－、動燃技術資料、PNC TN8410 92-222、動燃事業団(1992. 8)(1992).  
Sato, H., Yui, M., Ishikawa, H.: A Study on Pore Structure of Rocks, PNC Technical Report, PNC TN8410 92-222, Power Reactor and Nuclear Fuel Development Corporation (1992. 8)(1992).
- 2 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V. (1997).
- 3 Sato, H., Shibutani, T., Tachi, Y., Ota, K., Kenji, A., Yui, M.: Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127, Power Reactor and Nuclear Fuel Development Corporation (1997. 8)(1997).  
佐藤治夫、澁谷朝紀、館幸男、太田久仁雄、天野憲治、油井三和：物質移行経路を考慮した割れ目を含む結晶質岩中の核種の拡散挙動、動燃技術資料、PNC TN 1410 97-127、動燃事業団(1997. 8)(1997).
- 4 佐藤治夫、澁谷朝紀：亀裂性岩盤中における吸着・拡散係数、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010、pp.139-144、動燃事業団(1996.12)(1996).  
Sato, H., Shibutani, T.: PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.139-144, Power Reactor and Nuclear Fuel Development Corporation (1996. 12) (1996).
- 5 佐藤治夫、館幸男、澁谷朝紀、油井三和：物質移行経路を考慮した核種の拡散挙動、日本原子力学会1997年春の年会、L12、p.561(1997).  
Sato, H., Tachi, Y., Shibutani, T., Yui, M.: Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan, L12, p.561 (1997).
- 6 佐藤治夫、澁谷朝紀、吉川英樹、油井三和：花崗閃緑岩中のCs, Ni, Smの拡散機構に関する研究、日本原子力学会1993年秋の大会、M29、p.667(1993).  
Sato, H., Shibutani, T., Yoshikawa, H., Yui, M.: A Study on Diffusion Mechanism for Cs, Ni and Sm in Granodiorite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M29, p.667 (1993).
- 7 佐藤治夫、油井三和：岩石中のイオンの拡散係数の測定、日本原子力学会1992年秋の大会、F15、p.323(1992).  
Sato, H., Yui, M.: Measurement of Ionic Diffusion Coefficients in Rocks, 1992 Fall Meeting of the Atomic Energy Society of Japan, F15, p.323 (1992).
- 8 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).

ページ #2 - “酸性結晶質岩De/Da(Cl)data”

Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

9 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.205-207、動燃事業団(1996. 12)(1996)。

Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.205-207, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

Footnote:

- 1) These data were originally obtained to evaluate formation factor and tortuosity of the rock.
- 2) The major mineral composition of the fracture fillings is calcite and stilbite, and those of the altered and intact granodiorite are quartz and plagioclase. The content of constituent chlorite in the altered granodiorite is a little higher than that in the intact granodiorite.
- 3) The concentration of Cl<sup>-</sup> was calculated as a counter ion of Cs<sup>+</sup>, Ni<sup>2+</sup> and Sm<sup>3+</sup>. In this study, a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub> was used as a tracer solution. Since the concentration of each cation was 0.1 M, the concentration of Cl<sup>-</sup> is estimated to be 0.6 M.
- 4) The in-situ groundwater sampled from in-situ test site was used for the experiments. The chemical composition of the groundwater is described in more detail in the literature. Basically, the concentrations of Na, K, Ca, Si, F<sup>-</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup> are shown in it.
- 5) The tracer solution was prepared as a mixture of 0.01 M CsCl and 0.05 M NaCl. The concentration of Cl<sup>-</sup> is therefore estimated to be 0.06 M as a counter ion of Cs<sup>+</sup> and Na<sup>+</sup>.
- 6) The rock sample was sampled from Sori, Seta-gun, Gunma-ken. The granodiorite is approximately equivalent to a standard sample in the chemical composition of Japanese granite in the Geological Survey of Japan. The granodiorite, regarded as one of the candidate host rocks for repository, is major host rock in Japan.
- 7) The Do of Cl<sup>-</sup> is estimated 2.0e-9 m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169.].
- 8) The formation factor (FF) was calculated based on the equation: FF=De/Do. Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 9) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation: Gf=Dp/Do=De/(Φ·Do)=FF/Φ. Where Dp is the porewater diffusion coefficient, Do is the free water

ページ #3 - “酸性結晶質岩 $D_e/D_a(Cl)$  data”

diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "酸性結晶質岩De/Da(Co)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Co:De(m2/s)	Co:log De	Do(m2/s) 5)	Formation factor(FF) 6)	Geometric factor(Gf) 7)	Rock capacity factor
0	Co	4	Co2+	2.0	Granite 1)		0.680	2.10e-13	-12.878	7.30e-10	0.000290	0.0420	
1	Co	4	Co2+	2.0	Granite 1)		0.680	3.40e-13	-12.469	7.30e-10	0.000470	0.0680	
2	Co	4	Co2+	2.0	Granite 1)		0.680	3.10e-13	-12.509	7.30e-10	0.000420	0.0620	
3	Co	4	Co2+	2.0	Granite 1)		0.680	2.60e-13	-12.585	7.30e-10	0.000360	0.0520	
4	Co	4	Co2+	2.0	Granite 1)		0.680	2.20e-13	-12.658	7.30e-10	0.000300	0.0440	
5	Co	4	Co2+	2.0	Granite 1)		0.680	3.50e-13	-12.456	7.30e-10	0.000480	0.0710	
6	Co	4	Co2+	2.0	Granite 1)		0.680	2.50e-13	-12.602	7.30e-10	0.000340	0.0500	
7	Co	4	Co2+	2.0	Granite 1)		0.680	1.60e-13	-12.798	7.30e-10	0.000220	0.0320	
8	Co-60	4	Probably Co2+	2.0	Granite 1)		0.800			7.30e-10			
9	Co-60	4	Probably Co2+	2.0	Granite 1)		0.800			7.30e-10			
10	Co-60	4	Probably Co2+	2.0	Granite 1)		0.800			7.30e-10			
11	Co-60	4	Probably Co2+	2.0	Granite 1)		0.800			7.30e-10			

ページ #2 - "酸性結晶質岩De/Da(Co)data"

	Co:Da(m2/s)	Co:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0			1	Through-diffusion	0.1M KCl 3)	4.00	CoCl2 2)		25.0		aerobic 4)
1			1	Through-diffusion	0.1M KCl 3)	4.00	CoCl2 2)		25.0		aerobic 4)
2			1	Through-diffusion	0.1M KCl 3)	4.00	CoCl2 2)		25.0		aerobic 4)
3			1	Through-diffusion	0.1M KCl 3)	4.00	CoCl2 2)		25.0		aerobic 4)
4			1	Through-diffusion	0.1M KCl 3)	4.00	CoCl2 2)		25.0		aerobic 4)
5			1	Through-diffusion	0.1M KCl 3)	4.00	CoCl2 2)		25.0		aerobic 4)
6			1	Through-diffusion	0.1M KCl 3)	4.00	CoCl2 2)		25.0		aerobic 4)
7			1	Through-diffusion	0.1M KCl 3)	4.00	CoCl2 2)		25.0		aerobic 4)
8	2.00e-11	-10.699	2	In-diffusion	Deionized water	4.30	2KBq/ml 8)	7 days	30.0		aerobic 9)
9	8.00e-12	-11.697	2	In-diffusion	Deionized water	6.00	2KBq/ml 8)	7 days	30.0		aerobic 9)
10	2.00e-11	-10.699	2	In-diffusion	Deionized water	7.70	2KBq/ml 8)	7 days	30.0		aerobic 9)
11	2.00e-11	-10.699	2	In-diffusion	Deionized water	9.40	2KBq/ml 8)	7 days	30.0		aerobic 9)

## ページ #1 - “酸性結晶質岩De/Da(Co)data”

## REFERENCE AND FOOTNOTE

## Reference:

- 1 Yamaguchi, T., Sakamoto, Y., Senoo, M.: Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796-803, the Atomic Energy Society of Japan (August 1993) (1993).
- 2 Idemitsu, K., Furuya, H., Hara, T., Inagaki, Y.: Migration of Cesium, Strontium and Cobalt in Water-Saturation Inada Granite, Journal of Nuclear Science and Technology, Vol.29, No.5, pp.454-460, the Atomic Energy Society of Japan (1992).

## Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The granite, regarded as one of the candidate host rocks for repository, is major host rock in Japan.
- 2) The tracer was used as a mixture of Ba(NO<sub>3</sub>)<sub>2</sub>, BeSO<sub>4</sub>, CoCl<sub>2</sub>, MgCl<sub>2</sub>, NiCl<sub>2</sub> and Sr(NO<sub>3</sub>)<sub>2</sub>, what we call as a multi-tracer. Individual concentrations in the tracer are not reported.
- 3) This solution (0.1 M KCl) was used to keep the ionic strength of the porewater in both cells.
- 4) Apparent diffusion coefficient (Da) is not reported in any literature.
- 5) The Do of Co<sup>2+</sup> is estimated 7.3e-10 m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 6) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 7) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\Phi \cdot Do)=FF/\Phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\Phi$  is the porosity and FF is the formation factor.
- 8) A mixture solution of Cs-134, Sr-85 and Co-60 was prepared as a tracer solution. The concentration of each tracer is approximately 2kBq/ml.
- 9) De can not be obtained from this method. In this literature, two kinds of Da values, Df and Dm, are reported based on the model of grain-boundary diffusion in a crystalline solid. The Df is the diffusion in fissures which are perpendicular to the specimen surface, and the other is the diffusion into matrix which is parallel to the specimen surface. Since diffusion in rock matrix is controlled by Df, the Df was adopted



ページ #2 - “酸性結晶質岩De/Da(Co)data”

in this database.

ページ #1 - "酸性結晶質岩De/Da(Cs)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm <sup>3</sup> )	Porosity(%)	Cs:De(m <sup>2</sup> /s)	Cs:log De	Do(m <sup>2</sup> /s) 10)	Formation factor(FF) 11)	Geometric factor(Gf) 12)	Rock capacity factor
0	Cs	5	Cs+	1.0	Granodiorite	2.64	1.20	1.40e-12	-11.854	2.10e-09	0.000670	0.0560	0.140
1	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	7.80e-12	-11.108	2.10e-09	0.00370	0.160	0.650
2	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	2.50e-12	-11.602	2.10e-09	0.00120	0.0520	0.190
3	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	5.00e-12	-11.301	2.10e-09	0.00240	0.100	0.700
4	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	2.50e-12	-11.602	2.10e-09	0.00120	0.0380	0.700
5	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	4.80e-12	-11.319	2.10e-09	0.00230	0.0720	0.910
6	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	6.00e-12	-11.222	2.10e-09	0.00290	0.0910	1.20
7	Cs	5	Cs+	1.0	Fracture fillings	2.43	5.60	1.40e-11	-10.854	2.10e-09	0.00670	0.120	1.30
8	Cs	5	Cs+	1.0	Fracture fillings	2.43	5.60	2.20e-11	-10.658	2.10e-09	0.01000	0.180	1.70
9	Cs	5	Cs+	1.0	Granodiorite		0.880	1.40e-12	-11.854	2.10e-09	0.000670	0.0760	
10	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	5.10e-12	-11.292	2.10e-09	0.00240	0.100	
11	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	1.20e-11	-10.921	2.10e-09	0.000570	0.0180	
12	Cs	5	Cs+	1.0	Fracture fillings	2.43	5.60	1.80e-11	-10.745	2.10e-09	0.000860	0.0150	
13	Cs	5	Cs+	1.0	Granodiorite		2.30	5.10e-12	-11.292	2.10e-09	0.00240	0.100	
14	Cs	5	Cs+	1.0	Altered granodiorite		3.20	1.20e-11	-10.921	2.10e-09	0.000570	0.0180	
15	Cs	5	Cs+	1.0	Fracture fillings		5.60	1.80e-11	-10.745	2.10e-09	0.000860	0.0160	
16	Cs	5	Cs+	1.0	Granodiorite		1.00	1.36e-12	-11.888	2.10e-09	0.000650	0.0650	
17	Cs	5	Cs+	1.0	Granodiorite 4)			1.40e-12	-11.854	2.10e-09	0.000670		0.140
18	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	5.10e-12	-11.292	2.10e-09	0.00240	0.100	
19	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	1.20e-11	-10.921	2.10e-09	0.000570	0.0180	
20	Cs	5	Cs+	1.0	Fracture fillings	2.43	5.60	1.80e-11	-10.745	2.10e-09	0.000860	0.0150	
21	Cs	5	Cs+	1.0	Granodiorite 4)		0.880	1.40e-12	-11.854	2.10e-09	0.000670	0.0760	
22	Cs-134	5	Cs+	1.0	Granite					2.10e-09			
23	Cs-134	5	Cs+	1.0	Biotitic granite 7)	2.70	0.374			2.10e-09			
24	Cs-134	5	Cs+	1.0	Granite 7)		0.800			2.10e-09			
25	Cs-134	5	Cs+	1.0	Granite 7)		0.800			2.10e-09			
26	Cs-134	5	Cs+	1.0	Granite 7)		0.800			2.10e-09			
27	Cs-134	5	Cs+	1.0	Granite 7)		0.800			2.10e-09			
28	Cs-134	5	Cs+	1.0	Granite 7)		0.800			2.10e-09			
29	Cs-134	5	Cs+	1.0	Granite 7)		0.800			2.10e-09			

ページ #2 - "酸性結晶質岩Da/Da(Cs)data"

	Cs:Da(m <sup>2</sup> /s)	Cs:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	1.00e-11	-11.000	1	Through-diffusion	Distilled water	5.80	0.1M CsCl		25.0		aerobic
1	1.20e-11	-10.921	2	Through-diffusion	groundwater 1)	8.70	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
2	1.30e-11	-10.886	2	Through-diffusion	groundwater 1)	8.70	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
3	7.10e-12	-11.149	2	Through-diffusion	groundwater 1)	8.70	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
4	3.20e-12	-11.485	2	Through-diffusion	groundwater 1)	8.77	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
5	5.30e-12	-11.276	2	Through-diffusion	groundwater 1)	8.77	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
6	5.10e-12	-11.292	2	Through-diffusion	groundwater 1)	8.77	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
7	1.10e-11	-10.959	2	Through-diffusion	groundwater 1)	8.92	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
8	1.30e-11	-10.886	2	Through-diffusion	groundwater 1)	8.92	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
9	1.00e-11	-11.000	3	Through-diffusion	Distilled water	6.00	0.1M CsCl 3)		25.0		aerobic
10	1.10e-11	-10.959	4	Through-diffusion	groundwater	8.70	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
11	4.50e-12	-11.347	4	Through-diffusion	groundwater	8.77	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
12	1.20e-11	-10.921	4	Through-diffusion	groundwater	8.92	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
13	1.10e-11	-10.959	5	Through-diffusion	groundwater	8.70			25.0		aerobic
14	4.50e-12	-11.347	5	Through-diffusion	groundwater	8.77			25.0		aerobic
15	1.20e-11	-10.921	5	Through-diffusion	groundwater	8.92			25.0		aerobic
16	1.00e-11	-11.000	6	Through-diffusion	Distilled water	6.00	0.1M CsCl		20.0		aerobic
17	1.00e-11	-11.000	7	Through-diffusion	Distilled water	5.80	0.1M CsCl 3)		25.0		aerobic
18	1.10e-11	-10.959	8	Through-diffusion	groundwater	8.70	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
19	4.60e-12	-11.347	8	Through-diffusion	groundwater	8.77	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
20	1.20e-11	-10.921	8	Through-diffusion	groundwater	8.92	0.01M(1330ppm)CsCl	30days	25.0		aerobic 2)
21	1.00e-11	-11.000	9	Through-diffusion	Distilled water	6.00	0.1M CsCl 3)		20.0		aerobic
22	3.10e-14	-13.509	10	Advection diffusion	Distilled water		CsCl(+Cs-134) 6)		probably room		aerobic 5)
23	8.00e-12	-11.097	11	In-diffusion 8)	Distilled water		Cs-134+Cs carrier 9)	51days	70.0		aerobic
24	1.00e-11	-11.000	12	In-diffusion	Deionized water	4.30	2kBq/ml 13)	7days	30.0		aerobic 14)
25	1.00e-11	-11.000	12	In-diffusion	Deionized water	6.00	2kBq/ml 13)	7days	30.0		aerobic 14)
26	1.00e-11	-11.000	12	In-diffusion	Deionized water	7.70	2kBq/ml 13)	7days	30.0		aerobic 14)
27	1.00e-11	-11.000	12	In-diffusion	Deionized water	9.40	2kBq/ml 13)	7days	30.0		aerobic 14)
28	1.50e-11	-10.824	12	In-diffusion	Deionized water	4.20	2kBq/ml 13)	7days	40.0		aerobic 14)
29	3.00e-11	-10.523	12	In-diffusion	Deionized water	4.20	2kBq/ml 13)	7days	56.0		aerobic 14)

## ページ #1 - “酸性結晶質岩De/Da(Cs)data”

## REFERENCE AND FOOTNOTE

## Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V. (1997).
- 2 Sato, H., Shibutani, T., Tachi, Y., Ota, K., Kenji, A., Yui, M.: Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127, Power Reactor and Nuclear Fuel Development Corporation (1997. 8)(1997).  
佐藤治夫、澁谷朝紀、館幸男、太田久仁雄、天野憲治、油井三和：物質移行経路を考慮した割れ目を含む結晶質岩中の核種の拡散挙動、動燃技術資料、PNC TN1410 97-127, 動燃事業団(1997. 8)(1997).
- 3 動燃事業団：地層処分研究開発の現状(平成5年度)、動燃技術資料、PNC TN1410 94-094, pp.131-133, 動燃事業団(1994. 11)(1994).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1993-, PNC Technical Report, PNC TN1410 94-094, pp.131-133, Power Reactor and Nuclear Fuel Development Corporation (1994. 11)(1994).
- 4 佐藤治夫、澁谷朝紀：亀裂性岩盤中における吸着・拡散係数、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010, pp.139-144, 動燃事業団(1996.12)(1996).  
Sato, H., Shibutani, T.: PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.139-144, Power Reactor and Nuclear Fuel Development Corporation (1996. 12) (1996).
- 5 佐藤治夫、館幸男、澁谷朝紀、油井三和：物質移行経路を考慮した核種の拡散挙動、日本原子力学会1997年春の年会、L12, p.561(1997).  
Sato, H., Tachi, Y., Shibutani, T., Yui, M.: Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan, L12, p.561 (1997).
- 6 佐藤治夫、澁谷朝紀、吉川英樹、油井三和：花崗閃緑岩中のCs, Ni, Smの拡散機構に関する研究、日本原子力学会1993年秋の大会、M29, p.667(1993).  
Sato, H., Shibutani, T., Yoshikawa, H., Yui, M.: A Study on Diffusion Mechanism for Cs, Ni and Sm in Granodiorite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M29, p.667 (1993).
- 7 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071, pp.202-204, 動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 8 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071, pp.205-207, 動燃事業団(1996. 12)(1996).

ページ #2 - “酸性結晶質岩De/Da(Cs)data”

- Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.205-207, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 9 佐藤治夫、澁谷朝紀：緩衝材および岩石への核種の吸着・拡散メカニズム、動燃技報、No.91、PNC TN8410 94-284、pp.71-89、動燃事業団(1994. 9)(1994).  
Sato, H., Shibutani, T.: Study on Adsorption and Diffusion of Nuclides in Buffer Material and Geosphere, PNC Technical Review, No.91, PNC TN 8410 94-284, pp.71-89, Power Reactor and Nuclear Fuel Development Corporation (1994. 9)(1994).
- 10 向井悟、北尾秀夫、片岡伸一：亀裂性岩石中のI, Se及びCsを用いた移流・拡散試験、日本原子力学会1998年秋の大会、M25、p.842(1998).  
Mukai, S., Kitao, H., Kataoka, S.: Advection Diffusion Experiment Using I, Se and Cs in Fractured Rocks, 1998 Fall Meeting of the Atomic Energy Society of Japan, M25, p.842 (1998).
- 11 塚本政樹、大江俊昭：花崗岩中への放射性セシウムの拡散挙動、日本原子力学会「昭和63年秋の大会」、J45、p.169(1988).  
Tsukamoto, M., Ohe, T.: Diffusion of Radioactive Cesium into Granitic Core Sample, 1988 Fall Meeting of the Atomic Energy Society of Japan, J45, p.169 (1988).
- 12 Idemitsu, K., Furuya, H., Hara, T., Inagaki, Y.: Migration of Cesium, Strontium and Cobalt in Water-Saturation Inada Granite, Journal of Nuclear Science and Technology, Vol.29, No.5, pp.454-460, the Atomic Energy Society of Japan (1992).

Footnote:

- 1) The in-situ groundwater sampled from in-situ test site was used for the experiments. The chemical composition of the groundwater is described in more detail in the literature. Basically, the concentrations of Na, K, Ca, Si, F<sup>-</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup> are shown in it.
- 2) The rock samples were sampled from a drift of 250m level in the Kamaishi mine, Iwate-ken, Japan. The 250m drift is positioned at 250m above sea level, but it is about 700m deep from ground surface because it is mountain area.  
The major mineral composition of the fracture fillings is calcite and stilbite, and those of the altered and intact granodiorite are quartz and plagioclase. The content of constituent chlorite in the altered granodiorite is a little higher than that in the intact granodiorite.
- 3) The tracer solution was prepared with a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water.
- 4) The rock sample was sampled from Sori, Seta-gun, Gunma-ken, Japan. This granodiorite approximately has the same chemical composition as a standard sample of Japanese rocks in the Geological Survey of Japan. This kind of rock, regarded as one of the candidate host rocks for

ページ #3 - “酸性結晶質岩De/Da(Cs)data”

repository, is major host rock in Japan.

- 5) The advection diffusion experiments were carried out using a granitic rock with an artificial single fracture, which aperture was prepared to produce 50-100  $\mu\text{m}$ . The length of the rocks is 17-18cm. The Da was calculated fitting based on an equation considered advection-dispersion-sorption (retardation) and matrix diffusion from the concentration profile from the surface of the fracture.
- 6) The activity of Cs-134 used in the experiment is 130Bq/ml.
- 7) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The granite, regarded as one of the candidate host rocks for repository, is major host rock in Japan.
- 8) The rock sample with the size of 50mm $\phi$ x70mm was used.
- 9) The tracer was prepared as a mixture of  $7\text{e-}3\ \mu\text{Ci/ml}$  Cs-134 and 1ppb cold Cs carrier, and the total initial concentration of Cs is 1ppb.
- 10) The Do of Cs+ is estimated  $2.1\text{e-}9\ \text{m}^2/\text{s}$  at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 11) The formation factor (FF) was calculated based on the equation:  $\text{FF}=\text{De}/\text{Do}$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 12) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $\text{Gf}=\text{Dp}/\text{Do}=\text{De}/(\phi\cdot\text{Do})=\text{FF}/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.
- 13) A mixture solution of Cs-134, Sr-85 and Co-60 was prepared as a tracer solution. The concentration of each tracer is approximately 2kBq/ml.
- 14) De can not be obtained from this method. In this literature, two kinds of Da values, Df and Dm, are reported based on the model of grain-boundary diffusion in a crystalline solid. The Df is the diffusion in fissures which are perpendicular to the specimen surface, and the other is the diffusion into matrix which is parallel to the specimen surface. Since diffusion in rock matrix is controlled by Df, the Df was adopted in this database.

ページ #1 - "酸性結晶質岩De/Da(H)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	HTO:De(m2/s)	HTO:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor
0	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	5.00e-12	-11.301	2.10e-09	0.00240	0.100	0.00
1	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	4.80e-12	-11.319	2.10e-09	0.00230	0.100	0.00
2	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	6.20e-12	-11.208	2.10e-09	0.00300	0.130	0.0330
3	H-3	6	H2O	0.0	Altered granodiorite	2.81	3.20	1.70e-11	-10.770	2.10e-09	0.00810	0.250	0.00
4	H-3	6	H2O	0.0	Altered granodiorite	2.81	3.20	2.00e-11	-10.698	2.10e-09	0.00950	0.300	0.00
5	H-3	6	H2O	0.0	Altered granodiorite	2.81	3.20	1.70e-11	-10.770	2.10e-09	0.00810	0.250	0.00
6	H-3	6	H2O	0.0	Fracture fillings	2.43	5.60	2.70e-11	-10.568	2.10e-09	0.0130	0.230	0.0560
7	H-3	6	H2O	0.0	Fracture fillings	2.43	5.60	2.80e-11	-10.553	2.10e-09	0.0130	0.240	0.0560
8	H-3	6	H2O	0.0	Fracture fillings	2.43	5.60	1.60e-11	-10.796	2.10e-09	0.00760	0.140	0.0560
9	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	5.30e-12	-11.276	2.10e-09	0.00250	0.110	
10	H-3	6	H2O	0.0	Altered granodiorite	2.81	3.20	1.80e-11	-10.745	2.10e-09	0.00860	0.180	
11	H-3	6	H2O	0.0	Granodiorite		2.30	5.30e-12	-11.276	2.10e-09	0.00250	0.110	
12	H-3	6	H2O	0.0	Altered granodiorite		3.20	1.80e-11	-10.745	2.10e-09	0.00860	0.180	
13	H-3	6	H2O	0.0	Fracture fillings		5.60	2.40e-11	-10.620	2.10e-09	0.0110	0.200	
14	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	6.30e-12	-11.276	2.10e-09	0.00250	0.110	
15	H-3	6	H2O	0.0	Altered granodiorite	2.81	3.20	1.80e-11	-10.745	2.10e-09	0.00860	0.180	

ページ #2 - "酸性結晶質岩De/Da(H)data"

	HTO:Da(m2/s)	HTO:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	2.20e-10	-9.6580	1	Through-diffusion	groundwater 1)	8.70	6000Bq/ml	23days	23.0		aerobic 2)
1	2.10e-10	-9.6780	1	Through-diffusion	groundwater 1)	8.70	6000Bq/ml	23days	23.0		aerobic 2)
2	1.90e-10	-9.7210	1	Through-diffusion	groundwater 1)	8.70	6000Bq/ml	23days	23.0		aerobic 2)
3	5.30e-10	-9.2760	1	Through-diffusion	groundwater 1)	8.77	6000Bq/ml	23days	23.0		aerobic 2)
4	6.30e-10	-9.2010	1	Through-diffusion	groundwater 1)	8.77	6000Bq/ml	23days	23.0		aerobic 2)
5	5.30e-10	-9.2760	1	Through-diffusion	groundwater 1)	8.77	6000Bq/ml	23days	23.0		aerobic 2)
6	4.80e-10	-9.3190	1	Through-diffusion	groundwater 1)	8.92	6000Bq/ml	23days	23.0		aerobic 2)
7	5.00e-10	-9.3010	1	Through-diffusion	groundwater 1)	8.92	6000Bq/ml	23days	23.0		aerobic 2)
8	2.90e-10	-9.5380	1	Through-diffusion	groundwater 1)	8.92	6000Bq/ml	23days	23.0		aerobic 2)
9	2.10e-10	-9.6780	2	Through-diffusion	groundwater	8.70	6000Bq/ml	23days	23.0		aerobic
10	5.80e-10	-9.2520	2	Through-diffusion	groundwater	8.77	6000Bq/ml	23days	23.0		aerobic
11	2.10e-10	-9.6780	3	Through-diffusion	groundwater	8.70	2MBq/5ml 3)		23.0		aerobic
12	5.80e-10	-9.2520	3	Through-diffusion	groundwater	8.77	2MBq/5ml 3)		23.0		aerobic
13	4.30e-10	-9.3670	3	Through-diffusion	groundwater	8.92	2MBq/5ml 3)		23.0		aerobic
14	2.10e-10	-9.6780	4	Through-diffusion	groundwater	8.70	6000Bq/ml	23days	23.0		aerobic
15	5.80e-10	-9.2520	4	Through-diffusion	groundwater	8.77	6000Bq/ml	23days	23.0		aerobic



## ページ #1 - “酸性結晶質岩De/Da(H)data”

## REFERENCE AND FOOTNOTE

## Reference:

- 1 Sato, H., Shibutani, T., Tachi, Y., Ota, K., Amano, K., Yui, M.: Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127, Power Reactor and Nuclear Fuel Development Corporation (1997. 8)(1997).  
佐藤治夫、澁谷朝紀、館幸男、太田久仁雄、天野憲治、油井三和：物質移行経路を考慮した割れ目を含む結晶質岩中の核種の拡散挙動、動燃技術資料、PNC TN 1410 97-127, 動燃事業団(1997. 8)(1997).
- 2 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN 1410 96-071、pp.205-207、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.205-207, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 3 佐藤治夫、館幸男、澁谷朝紀、油井三和：物質移行経路を考慮した核種の拡散挙動、日本原子力学会1997年春の年会、L12、p.561(1997).  
Sato, H., Tachi, Y., Shibutani, T., Yui, M.: Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan, L12, p.561 (1997).
- 4 佐藤治夫、澁谷朝紀：亀裂性岩盤中における吸着・拡散係数、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010、pp.139-144、動燃事業団(1996.12)(1996).  
Sato, H., Shibutani, T.: PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.139-144, Power Reactor and Nuclear Fuel Development Corporation (1996. 12) (1996).

## Footnote:

- 1) The in-situ groundwater sampled from in-situ test site was used for the experiments. The chemical composition of the groundwater is described in more detail in the literature. Basically, the concentrations of Na, K, Ca, Si, F-, Cl-, SO42-, CO32-, HCO3- are shown in it.
- 2) The major mineral composition of the fracture fillings is calcite and stilbite, and those of the altered and intact granodiorite are quartz and plagioclase. The content of constituent chlorite in the altered granodiorite is a little higher than that in the intact granodiorite.
- 3) This is a stock solution obtained from Amersham Japan. Whether the tracer solution with this concentration (2MBq/ml) was actually used or

ページ #2 - “酸性結晶質岩De/Da(H)data”

not for diffusion experiments, is unclear.

- 4) The  $D_0$  of heavy water was used as a value of HTO. The  $D_0$  has been reported  $2.14 \times 10^{-9}$  m<sup>2</sup>/s at 25°C [Chemical Handbook, Basic Version II, 2nd ed., Chemical Society of Japan, Maruzen, Tokyo, p.604 (1975) [In Japanese].].
- 5) The formation factor (FF) was calculated based on the equation:  
 $FF = D_e / D_0$ . Where  $D_e$  and  $D_0$  is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 6) The geometric factor ( $G_f$ ) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f = D_p / D_0 = D_e / (\phi \cdot D_0) = FF / \phi$ .  
Where  $D_p$  is the porewater diffusion coefficient,  $D_0$  is the free water diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "放射性結晶質岩De/Da(l)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	I:De(m2/s)	I:log De	Do(m2/s) 5)	Formation factor(FF) 6)	Geometric factor(Gf) 7)	Rock capacity factor	I:Da(m2/s)
0		7	I-	-1.0	Granite		0.800	2.60e-12	-11.585	2.00e-09	0.00130	0.180	0.0950	2.74e-11
1		7	I-	-1.0	Granite		0.800	1.90e-12	-11.721	2.00e-09	0.000950	0.120	0.140	1.36e-11
2		7	I-	-1.0	Granite		0.770	2.60e-12	-11.585	2.00e-09	0.00130	0.170	0.0900	2.89e-11
3		7	I-	-1.0	Granite		0.770	1.90e-12	-11.721	2.00e-09	0.000950	0.120	0.130	1.46e-11
4	I-125	7	I-	-1.0	Granite 2)		0.400	4.40e-13	-12.357	2.00e-09	0.000220	0.0550	0.340	1.30e-12

ページ #2 - "酸性結晶質岩De/Da(I)data"

	I/log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	-10.562	1	Through-diffusion	Deionized water		1M(127000ppm)KI		probably room		aerobic
1	-10.866	1	Through-diffusion	Deionized water		1M(127000ppm)KI		probably room		aerobic
2	-10.539	2	Through-diffusion	Distilled water		1M(127000ppm)KI		probably room		aerobic
3	-10.838	2	Through-diffusion	Distilled water		1M(127000ppm)KI		probably room		aerobic
4	-11.886	3	Through-diffusion	groundwater 1)		Sr12(3.4e-4M) 3)		probably room		aerobic 4)

ページ #1 - “酸性結晶質岩De/Da(l)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 西山勝栄、中嶋悟、多田隆治、内田隆：岩石間隙水中のイオンの拡散係数と間隙の性質、*鉱山地質*、40 (5)、323~336 (1990).  
Nishiyama, K., Nakashima, S., Tada, R., Uchida, T.: Diffusion of an Ion in Rock Pore Water and Its Relation to Pore Characteristics, *Mining Geology*, 40 (5), pp.323-336 (1990).
- 2 喜多治之、岩井孝幸、中嶋悟：花崗岩および凝灰岩間隙水中のイオンの拡散係数の測定、*応用地質*、30 (2)、26~32 (1989).  
Kita, H., Iwai, T., Nakashima, S.: Diffusion Coefficient Measurement of an Ion in Pore Water of Granite and Tuff, *Journal of the Japan Society of Engineering Geology*, 30-2, pp.26-32 (1989).
- 3 山口徹治、中山真一：微小間隙中における拡散係数に対する吸着の影響、日本原子力学会1997年春の年会、L10, p.559 (1997).  
Yamaguchi, T., Nakayama, S.: Effect of Sorption on Diffusivity in Micropores, 1997 Annual Meeting of the Atomic Energy Society of Japan, L10, p.559 (1997).

Footnote:

- 1) The groundwater was sampled from a fracture zone at 240m in depth. The ionic strength of the groundwater is  $\sim 0.01$  according to the literature.
- 2) The rock sample was sampled from a fracture zone at 240m in depth in the Canada Whiteshell Laboratories, Canada.
- 3) The tracer solution was prepared by dissolving a small amount of I-125 and Sr-85 in a non-radioactive SrI2 solution, which concentration is  $3.4e-4$  M.
- 4) The porosity was calculated from effective diffusion coefficient ( $D_e$ ), free water diffusion coefficient ( $D_o$ ) and geometric factor ( $G_f$ ) based on  $D_e = \phi \cdot G_f \cdot D_o$  described in the literature. Where  $\phi$  is the porosity.
- 5) The  $D_o$  of I- is estimated  $2.0e-9$  m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169.].
- 6) The formation factor (FF) was calculated based on the equation:  $FF = D_e / D_o$ . Where  $D_e$  and  $D_o$  is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 7) The geometric factor ( $G_f$ ) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f = D_p / D_o = D_e / (\phi \cdot D_o) = FF / \phi$ . Where  $D_p$  is the porewater diffusion coefficient,  $D_o$  is the free water diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "凝性結晶質岩De/Da(K)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	K:De(m2/s)	K:log De	Do(m2/s) 5)	Formation factor(FF) 6)	Geometric factor(Gf) 7)	Rock capacity factor	K:Da(m2/s)
0	K	8	K+	1.0	Granite 1)	2.60	0.880	7.70e-13	-12.114	2.00e-09	0.000390	0.0440		
1	K	8	K+	1.0	Granite 1)	2.60	0.880	1.10e-12	-11.959	2.00e-09	0.000550	0.0630		
2	K	8	K+	1.0	Granite 1)	2.60	0.880	8.30e-13	-12.081	2.00e-09	0.000410	0.0470		
3	K	8	K+	1.0	Granite 1)	2.60	0.880	1.80e-12	-11.745	2.00e-09	0.000900	0.100		
4	K	8	K+	1.0	Granite 1)	2.60	0.880	8.30e-13	-12.081	2.00e-09	0.000420	0.0470		
5	K	8	K+	1.0	Granite 1)	2.60	0.880	1.80e-12	-11.745	2.00e-09	0.000900	0.100		

ページ #2 - "酸性結晶質岩De/Da(K)data"

	K:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0		1	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
1		1	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
2		1	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
3		1	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
4		2	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)
5		2	conductivity 2)	0.1M KCl 3)		0.1M KCl 3)		probably room		aerobic 4)

ページ #1 - “酸性結晶質岩De/Da(K)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Conca, J. L., Wright, J.: Direct Determinations of Unsaturated Flow and Transport, Proceedings of the Twelfth Annual American Geophysical Union Hydrology Days Meeting, Fort Collins, March 1992, pp.103-116 (1992).
- 2 Conca, J. L., Apte, M. J., Arthur, R. C.: Direct Determinations of Transport Parameters in Repository Materials, Scientific Basis for Nuclear Waste Management XVI (Interrante, C. G. and Pabalan, R. T., eds.), Materials Research Society Symposium Proceedings, Vol. 294, pp.839-844, Materials Research Society, Pittsburgh, Pennsylvania (1993).

Footnote:

- 1) The rock sample used for the experiment was sampled from Sori, Setagun, Gunma-ken, Japan. The granodiorite, one of the candidate host rocks for repository, is major host rock in Japan.
- 2) The De values were determined by electrical conductivity measurements, based on the Nernst-Einstein equation. The De values were determined using the unsaturated flow apparatus (UFA). Hydraulic conductivity can also be determined by this method. The hydraulic conductivity is also reported in the literature.
- 3) The rock sample was firstly saturated with a 0.1 M KCl solution and then electrical conductivity was measured. The porewater and tracer are therefore the same.
- 4) Apparent diffusion coefficient (Da) can not be obtained from the electrical conductivity measurements. Since the porosity was not reported in these literatures, it was quoted from another literature (H. Sato and T. Shibutani, PNC Technical Report, PNC TN8410 94-284, pp.71-89.). Because PNC supplied the sample and determined the porosity by water saturation method (water intrusion method).
- 5) The Do of K<sup>+</sup> is estimated 2.0e-9 m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 6) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 7) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion



ページ #2 - “酸性結晶質岩De/Da(K)data”

coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and  $FF$  is the formation factor.

ページ #1 - "酸性結晶質岩De/Da(Mg)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Mg:De(m2/s)	Mg:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor	Mg:Da(m2/s)
0	Mg	9	Mg2+	2.0	Granite 1)		0.680	1.90e-13	-12.721	7.10e-10	0.000270	0.0390		
1	Mg	9	Mg2+	2.0	Granite 1)		0.680	3.30e-13	-12.481	7.10e-10	0.000460	0.0680		
2	Mg	9	Mg2+	2.0	Granite 1)		0.680	3.00e-13	-12.523	7.10e-10	0.000420	0.0620		
3	Mg	9	Mg2+	2.0	Granite 1)		0.680	2.90e-13	-12.538	7.10e-10	0.000410	0.0600		
4	Mg	9	Mg2+	2.0	Granite 1)		0.680	2.30e-13	-12.638	7.10e-10	0.000320	0.0480		
5	Mg	9	Mg2+	2.0	Granite 1)		0.680	3.60e-13	-12.444	7.10e-10	0.000510	0.0750		
6	Mg	9	Mg2+	2.0	Granite 1)		0.680	2.40e-13	-12.620	7.10e-10	0.000340	0.0500		
7	Mg	9	Mg2+	2.0	Granite 1)		0.680	1.90e-13	-12.721	7.10e-10	0.000270	0.0390		

ページ #2 - "礫性結晶質岩De/Da(Mg)data"

	Mg:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0		1	Through-diffusion	0.1M KCl 3)	4.00	MgCl2 2)		25.0		aerobic
1		1	Through-diffusion	0.1M KCl 3)	4.00	MgCl2 2)		25.0		aerobic
2		1	Through-diffusion	0.1M KCl 3)	4.00	MgCl2 2)		25.0		aerobic
3		1	Through-diffusion	0.1M KCl 3)	4.00	MgCl2 2)		25.0		aerobic
4		1	Through-diffusion	0.1M KCl 3)	4.00	MgCl2 2)		25.0		aerobic
5		1	Through-diffusion	0.1M KCl 3)	4.00	MgCl2 2)		25.0		aerobic
6		1	Through-diffusion	0.1M KCl 3)	4.00	MgCl2 2)		25.0		aerobic
7		1	Through-diffusion	0.1M KCl 3)	4.00	MgCl2 2)		25.0		aerobic

ページ #1 - “酸性結晶質岩De/Da(Mg)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Yamaguchi, T., Sakamoto, Y., Senoo, M.: Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796-803, the Atomic Energy Society of Japan (August 1993) (1993).

Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The kind of the rock, one of the candidate host rocks for repository, is major host rock in Japan.
- 2) The tracer was used as a mixture of Ba(NO<sub>3</sub>)<sub>2</sub>, BeSO<sub>4</sub>, CoCl<sub>2</sub>, MgCl<sub>2</sub>, NiCl<sub>2</sub> and Sr(NO<sub>3</sub>)<sub>2</sub>, what we call as a multi-tracer. Individual concentrations in the tracer are not reported.
- 3) This solution (0.1 M KCl) was used to keep the ionic strength of the porewater in both cells.
- 4) The Do of Mg<sup>2+</sup> is estimated 7.1e-10 m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 5) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Na:De(m2/s)	Na:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor
0	Na 1)	10	Na+	1.0	Granodiorite	2.64	1.20	4.40e-13	-12.357	1.30e-09	0.00340	0.0280	
1	Na 1)	10	Na+	1.0	Granodiorite	2.64	1.20	5.20e-13	-12.284	1.30e-09	0.00340	0.0330	
2	Na 1)	10	Na+	1.0	Granodiorite	2.64	1.20	4.70e-13	-12.328	1.30e-09	0.00360	0.0300	
3	Na	10	Na+	1.0	Granodiorite	2.73	2.30	6.50e-12	-11.187	1.30e-09	0.00500	0.220	0.0850
4	Na	10	Na+	1.0	Granodiorite	2.73	2.30	2.30e-12	-11.638	1.30e-09	0.00180	0.0770	
5	Na	10	Na+	1.0	Granodiorite	2.73	2.30	5.20e-12	-11.284	1.30e-09	0.00400	0.170	0.960
6	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	1.20e-11	-10.921	1.30e-09	0.00920	0.290	1.43
7	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	5.10e-12	-11.292	1.30e-09	0.00390	0.120	0.660
8	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	4.30e-12	-11.367	1.30e-09	0.00330	0.100	0.330
9	Na	10	Na+	1.0	Fracture fillings	2.43	5.60	2.10e-11	-10.678	1.30e-09	0.0160	0.290	1.11
10	Na	10	Na+	1.0	Fracture fillings	2.43	5.60	1.70e-11	-10.770	1.30e-09	0.0130	0.230	0.850
11	Na	10	Na+	1.0	Fracture fillings	2.43	5.60			1.30e-09			
12	Na	10	Na+	1.0	Granodiorite		2.30	4.70e-12	-11.328	1.30e-09	0.00360	0.160	
13	Na	10	Na+	1.0	Altered granodiorite		3.20	7.20e-12	-11.143	1.30e-09	0.00550	0.170	
14	Na	10	Na+	1.0	Fracture fillings		5.60	1.90e-11	-10.721	1.30e-09	0.0150	0.260	
15	Na	10	Na+	1.0	Granodiorite	2.73	2.30	4.70e-12	-11.328	1.30e-09	0.00360	0.160	
16	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	7.20e-12	-11.143	1.30e-09	0.00550	0.170	
17	Na	10	Na+	1.0	Fracture fillings	2.43	5.60	1.90e-11	-10.721	1.30e-09	0.0150	0.260	
18	Na	10	Na+	1.0	Granodiorite	2.73	2.30	4.70e-12	-11.328	1.30e-09	0.00360	0.160	
19	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	7.20e-12	-11.143	1.30e-09	0.00550	0.170	
20	Na	10	Na+	1.0	Fracture fillings	2.43	5.60	1.90e-11	-10.721	1.30e-09	0.0150	0.260	

ページ #2 - "酸性結晶質岩De/Da(Na)data"

	Na:Da(m2/s)	Na:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0			1	Through-diffusion	Distilled water	6.50	10%NaCl		25.0		aerobic
1			1	Through-diffusion	Distilled water	6.50	10%NaCl		25.0		aerobic
2			1	Through-diffusion	Distilled water	6.50	10%NaCl		25.0		aerobic
3	8.00e-11	-10.097	2	Through-diffusion	groundwater 3)	8.70	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
4			2	Through-diffusion	groundwater 3)	8.70	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
5	5.40e-12	-11.268	2	Through-diffusion	groundwater 3)	8.70	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
6	8.40e-12	-11.076	2	Through-diffusion	groundwater 3)	8.77	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
7	7.70e-12	-11.114	2	Through-diffusion	groundwater 3)	8.77	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
8	1.30e-11	-10.886	2	Through-diffusion	groundwater 3)	8.77	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
9	1.90e-11	-10.721	2	Through-diffusion	groundwater 3)	8.92	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
10	2.00e-11	-10.699	2	Through-diffusion	groundwater 3)	8.92	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
11			2	Through-diffusion	groundwater 3)	8.92	0.05M(1150ppmNa)	30days	25.0		aerobic 2)
12	4.30e-11	-10.367	3	Through-diffusion	groundwater	8.70			25.0		aerobic
13	9.70e-12	-11.013	3	Through-diffusion	groundwater	8.77			25.0		aerobic
14	2.00e-11	-10.699	3	Through-diffusion	groundwater	8.92			25.0		aerobic
15	4.30e-11	-10.367	4	Through-diffusion	groundwater	8.70	0.05M(1150ppmNa)	30days	25.0		aerobic
16	9.70e-12	-11.013	4	Through-diffusion	groundwater	8.77	0.05M(1150ppmNa)	30days	25.0		aerobic
17	2.00e-11	-10.699	4	Through-diffusion	groundwater	8.92	0.05M(1150ppmNa)	30days	25.0		aerobic
18	4.30e-11	-10.367	5	Through-diffusion	groundwater	8.70	0.05M(1150ppmNa)	30days	25.0		aerobic
19	9.70e-12	-11.013	5	Through-diffusion	groundwater	8.77	0.05M(1150ppmNa)	30days	25.0		aerobic
20	2.00e-11	-10.699	5	Through-diffusion	groundwater	8.92	0.05M(1150ppmNa)	30days	25.0		aerobic

## ページ #1 - “酸性結晶質岩De/Da(Na)data”

## REFERENCE AND FOOTNOTE

## Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V. (1997).
- 2 Sato, H., Shibutani, T., Tachi, Y., Ota, K., Amano, K., Yui, M.: Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127, Power Reactor and Nuclear Fuel Development Corporation (1997. 8)(1997).  
佐藤治夫、澁谷朝紀、館幸男、太田久仁雄、天野憲治、油井三和：物質移行経路を考慮した割れ目を含む結晶質岩中の核種の拡散挙動、動燃技術資料、PNC TN1410 97-127, 動燃事業団(1997. 8)(1997).
- 3 佐藤治夫、館幸男、澁谷朝紀、油井三和：物質移行経路を考慮した核種の拡散挙動、日本原子力学会1997年春の年会、L12, p.561(1997).  
Sato, H., Tachi, Y., Shibutani, T., Yui, M.: Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan, L12, p.561 (1997).
- 4 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071, pp.205-207、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.205-207 Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 5 佐藤治夫、澁谷朝紀：亀裂性岩盤中における吸着・拡散係数、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010, pp.139-144、動燃事業団(1996.12)(1996).  
Sato, H., Shibutani, T.: PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.139-144, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

## Footnote:

- 1) These data were originally obtained to evaluate formation factor and tortuosity of the rock.
- 2) The major mineral composition of the fracture fillings is calcite and stilbite, and those of the altered and intact granodiorite are quartz and plagioclase. The content of constituent chlorite in the altered granodiorite is a little higher than that in the intact granodiorite.
- 3) The in-situ groundwater sampled from in-situ test site was used for the experiments. The chemical composition of the groundwater is described in more detail in the literature. Basically, the concentrations of Na, K, Ca, Si, F<sup>-</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup> are shown in it.

ページ #2 - “酸性結晶質岩De/Da(Na)data”

- 4) The  $D_0$  of  $\text{Na}^+$  is estimated  $1.3 \times 10^{-9} \text{ m}^2/\text{s}$  at  $25^\circ\text{C}$  [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 5) The formation factor (FF) was calculated based on the equation:  
 $FF = D_e/D_0$ . Where  $D_e$  and  $D_0$  is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 6) The geometric factor ( $G_f$ ) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f = D_p/D_0 = D_e/(\phi \cdot D_0) = FF/\phi$ .  
Where  $D_p$  is the porewater diffusion coefficient,  $D_0$  is the free water diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.



ページ #1 - "放射性結晶質岩De/Da(Ni)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Ni:De(m2/s)	Ni:log De	Do(m2/s) 6)	Formation factor(FF) 7)	Geometric factor(Gf) 8)	Rock capacity factor
0	Ni	11	Ni2+	2.0	Granodiorite	2.64	1.20	5.70e-13	-12.244	6.60e-10	0.000860	0.0720	0.0570
1	Ni	11	Ni2+	2.0	Granite 2)		0.680	2.00e-13	-12.699	6.60e-10	0.000300	0.0450	
2	Ni	11	Ni2+	2.0	Granite 2)		0.680	3.40e-13	-12.469	6.60e-10	0.000520	0.0760	
3	Ni	11	Ni2+	2.0	Granite 2)		0.680	3.00e-13	-12.523	6.60e-10	0.000450	0.0670	
4	Ni	11	Ni2+	2.0	Granite 2)		0.680	2.90e-13	-12.538	6.60e-10	0.000440	0.0650	
5	Ni	11	Ni2+	2.0	Granite 2)		0.680	2.40e-13	-12.620	6.60e-10	0.000360	0.0530	
6	Ni	11	Ni2+	2.0	Granite 2)		0.680	3.60e-13	-12.444	6.60e-10	0.000550	0.0800	
7	Ni	11	Ni2+	2.0	Granite 2)		0.680	2.50e-13	-12.602	6.60e-10	0.000380	0.0560	
8	Ni	11	Ni2+	2.0	Granite 2)		0.680	1.90e-13	-12.721	6.60e-10	0.000280	0.0420	
9	Ni	11	Ni2+	2.0	Granodiorite		0.880	5.70e-13	-12.244	6.60e-10	0.000860	0.0980	
10	Ni	11	Ni2+	2.0	Granodiorite		1.00	5.72e-13	-12.243	6.60e-10	0.000870	0.0870	
11	Ni	11	Ni2+	2.0	Granodiorite 5)			5.70e-13	-12.244	6.60e-10			0.0570
12	Ni	11	Ni2+	2.0	Granodiorite 5)		0.880	5.70e-13	-12.244	6.60e-10	0.000860	0.0980	

ページ #2 - "酸性結晶質岩Da/Da(Ni)data"

	Ni:Da(m <sup>2</sup> /s)	Ni:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	1.00e-11	-11.000	1	Through-diffusion	Distilled water	5.80	0.1MNICl <sub>2</sub> 1)	137days	25.0		aerobic
1			2	Through-diffusion	0.1M KCl	4.00	NICl <sub>2</sub> 3)		25.0		aerobic
2			2	Through-diffusion	0.1M KCl	4.00	NICl <sub>2</sub> 3)		25.0		aerobic
3			2	Through-diffusion	0.1M KCl	4.00	NICl <sub>2</sub> 3)		25.0		aerobic
4			2	Through-diffusion	0.1M KCl	4.00	NICl <sub>2</sub> 3)		25.0		aerobic
5			2	Through-diffusion	0.1M KCl	4.00	NICl <sub>2</sub> 3)		25.0		aerobic
6			2	Through-diffusion	0.1M KCl	4.00	NICl <sub>2</sub> 3)		25.0		aerobic
7			2	Through-diffusion	0.1M KCl	4.00	NICl <sub>2</sub> 3)		25.0		aerobic
8			2	Through-diffusion	0.1M KCl	4.00	NICl <sub>2</sub> 3)		25.0		aerobic
9	1.00e-11	-11.000	3	Through-diffusion	Distilled water	6.00	0.1MNICl <sub>2</sub> 4)		20.0		aerobic
10	1.00e-11	-11.000	4	Through-diffusion	Distilled water	6.00	0.1MNICl <sub>2</sub> 4)		20.0		aerobic
11	1.00e-11	-11.000	5	Through-diffusion	Distilled water	5.80	0.1MNICl <sub>2</sub>		25.0		aerobic
12	1.00e-11	-11.000	6	Through-diffusion	Distilled water	5.80	0.1MNICl <sub>2</sub>		20.0		aerobic

ページ #1 - “酸性結晶質岩De/Da(Ni)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V. (1997).
- 2 Yamaguchi, T., Sakamoto, Y., Senoo, M.: Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796-803, the Atomic Energy Society of Japan (August 1993) (1993).
- 3 動燃事業団：地層処分研究開発の現状(平成5年度)、動燃技術資料、PNC TN1410 94-094、pp.131-133、動燃事業団(1994. 11)(1994).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1993-, PNC Technical Report, PNC TN1410 94-094, pp.131-133, Power Reactor and Nuclear Fuel Development Corporation (1994. 11)(1994).
- 4 佐藤治夫、澁谷朝紀、吉川英樹、油井三和：花崗閃緑岩中のCs, Ni, Smの拡散機構に関する研究、日本原子力学会1993年秋の大会、M29、p.667(1993).  
Sato, H., Shibutani, T., Yoshikawa, H., Yui, M.: A Study on Diffusion Mechanism for Cs, Ni and Sm in Granodiorite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M29, p.667 (1993).
- 5 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 6 佐藤治夫、澁谷朝紀：緩衝材および岩石への核種の吸着・拡散メカニズム、動燃技報、No.91、PNC TN8410 94-284、pp.71-89、動燃事業団(1994. 9)(1994).  
Sato, H., Shibutani, T.: Study on Adsorption and Diffusion of Nuclides in Buffer Material and Geosphere, PNC Technical Review, No.91, PNC TN8410 94-284, pp.71-89, Power Reactor and Nuclear Fuel Development Corporation (1994. 9)(1994).

Footnote:

- 1) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water.
- 2) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The kind of the rock, regarded as one of the candidate host rocks for repository, is major host rock in Japan.
- 3) The tracer was used as a mixture of Ba(NO<sub>3</sub>)<sub>2</sub>, BeSO<sub>4</sub>, CoCl<sub>2</sub>, MgCl<sub>2</sub>, NiCl<sub>2</sub> and Sr(NO<sub>3</sub>)<sub>2</sub>, what we call as a multi-tracer. Individual concentrations in the tracer are not reported.

ページ #2 - “酸性結晶質岩De/Da(Ni)data”

- 4) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>, which individual concentrations in the solutions are all 0.1 M.
- 5) The rock sample was sampled from Sori, Seta-gun, Gunma-ken, Japan. This rock is approximately equivalent to a standard sample in the chemical composition of Japanese granite in the Geological Survey of Japan.
- 6) The  $D_0$  of Ni<sup>2+</sup> is estimated 6.6e-10 m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 7) The formation factor (FF) was calculated based on the equation:  
 $FF = D_e / D_0$ . Where  $D_e$  and  $D_0$  is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 8) The geometric factor ( $G_f$ ) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f = D_p / D_0 = D_e / (\phi \cdot D_0) = FF / \phi$ .  
Where  $D_p$  is the porewater diffusion coefficient,  $D_0$  is the free water diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "酸性結晶質岩De/Da(Np)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Np:De(m2/s)	Np:log De	Do(m2/s) 8)	Formation factor(FF) 9)	Geometric factor(Gf) 10)	Rock capacity factor
0	Np-237	12	NpO2+	1.0	Granite 1)		0.740	2.10e-13	-12.678	1.20e-09	0.00180	0.240	0.0160
1	Np-237	12	NpO2+	1.0	Granite 1)		0.740	2.90e-13	-12.538	1.20e-09	0.00240	0.330	0.00400
2	Np-237	12			Granite 1)	2.64	0.490	4.60e-13	-12.337				0.240
3	Np-237	12			Granite 1)	2.64	0.490	4.60e-13	-12.337				0.240
4	Np-237	12			Granite 1)	2.64	0.490	4.60e-13	-12.337				0.240
5	Np-237	12	NpO2+	1.0	Granite 1)			2.10e-13	-12.678	1.20e-09	0.00180		0.0160
6	Np-237	12	NpO2+	1.0	Granite 1)			2.90e-13	-12.538	1.20e-09	0.00240		0.00400

ページ #2 - "放射性結晶質岩De/Da(Np)data"

	Np:Da(m2/s)	Np:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	1.30e-11	-10.886	1	Through-diffusion	groundwater 3)	7.40	1117Bq/ml 4)	about 180days	probably room		aerobic 2)
1	7.20e-11	-10.143	1	Through-diffusion	groundwater 3)	7.40	1117Bq/ml 4)	about 180days	probably room		aerobic 2)
2	1.92e-12	-11.717	2	Through-diffusion	1M NaHCO3	8.60	356Bq/ml	about 350days	26±1	-500~-400	reducing 5)
3	1.92e-12	-11.717	2	Through-diffusion	1M NaHCO3	8.60	356Bq/ml	about 350days	26±1	-500~-400	reducing 5)
4	1.92e-12	-11.717	2	Through-diffusion	1M NaHCO3	8.60	356Bq/ml	about 350days	26±1	-500~-400	reducing 5)
5	1.30e-11	-10.886	3	Through-diffusion	groundwater 6)	7.40			room		aerobic 7)
6	7.20e-11	-10.143	3	Through-diffusion	groundwater 6)	7.40			room		aerobic 7)

ページ #1 - “酸性結晶質岩De/Da(Np)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 熊田政弘、岩井孝幸、佐川民雄、鈴木隆次、西山勝栄：岩石コアを用いた放射性核種の拡散実験、JAERI-M 90-179、日本原子力研究所(1990. 10)(1990).  
Kumata, M., Iwai, T., Sagawa, T., Suzuki, T., Nishiyama, K.: Diffusion Experiment of a Radionuclide in Granitic Rock Cores, JAERI Report, JAERI-M 90-179, Japan Atomic Energy Research Institute (1990. 10) (1990).
- 2 山口徹治、中山真一、斎藤光男：低酸素条件下におけるNp, Puの花崗岩内拡散実験、日本原子力学会1998年秋の大会、M17, p.834 (1998).  
Yamaguchi, T., Nakayama, S., Saito, M.: Np and Pu Diffusion Experiments in Granite under Antoxic Conditions, 1998 Fall Meeting of the Atomic Energy Society of Japan, M17, p.834 (1998).
- 3 Yamaguchi, T., Sakamoto, Y., Nakayama, S., Vandergraaf, T. T.: Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, Journal of Contaminant Hydrology, 26, pp.109-117, Elsevier Science B. V. (1997).

Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The kind of the rock is granite, which is one of the candidate host rocks for repository and major host rock in Japan.
- 2) The porosity of the granite sample described is an average value of porosity data obtained by water saturation method (water intrusion method).  
The pH of the solution is an average value of reported pH range. The actual pH range is described to be between 7.2 and 7.5 in the literature.
- 3) The groundwater was sampled from Mt. Tsukuba, Ibaraki-ken, Japan, and the ionic strength of the groundwater was estimated to be  $I=0.077$  from the chemical composition in the literature. The groundwater was extracted from 250m deep from ground surface. The groundwater was filtered through a  $0.45\mu\text{m}$  filter before the experiment.
- 4) The stock solution of the tracer is a 3M HNO<sub>3</sub> solution, in which about 0.03mCi ( $1.11\text{e}6$  Bq) Np-237 is contained. The tracer was prepared by diluting the stock solution in groundwater.
- 5) Though the dominant chemical species of Np in the solution is not shown, it is reported that the TTA extraction showed Np(IV). The obtained De values were approximately the same degree as those of NpO<sub>2</sub><sup>+</sup>.  
The De described is an average value of data obtained from experiments in triplicate. The error of the De was evaluated to be  $4.6\pm 1.7\text{e-}13$  m<sup>2</sup>/s in the literature.  
The Da was calculated based on rock capacity factor and De.  
The Eh stands for redox potential vs. SHE (Standard Hydrogen Electrode)

ページ #2 - “酸性結晶質岩De/Da(Np)data”

or NHE (Normal Hydrogen Electrode). The experiments were carried out in a glove box purged with Ar gas, in which oxygen concentration is 1~ several ppm.

Sodium hydrosulfite ( $\text{Na}_2\text{S}_2\text{O}_4$ ) was added as a reductant to lower the Eh of the solution. This reagent was prepared to produce 0.05 M.

Distribution coefficient (Kd) was estimated to be  $9 \pm 7 \times 10^{-5}$  m<sup>3</sup>/kg from rock capacity factor, porosity and dry density in the literature.

- 6) The ionic strength of the groundwater was estimated to be  $I=0.077$  in the literature.
- 7) The pH of the solution is an average value of reported pH range. The actual pH range is described to be between 7.2 and 7.5 in the literature.
- 8) The  $D_0$  of  $\text{NpO}_2^{2+}$  is estimated  $1.2 \times 10^{-9}$  m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 9) The formation factor (FF) was calculated based on the equation:  
 $FF = D_e / D_0$ . Where  $D_e$  and  $D_0$  is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 10) The geometric factor ( $G_f$ ) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f = D_p / D_0 = D_e / (\phi \cdot D_0) = FF / \phi$ .  
Where  $D_p$  is the porewater diffusion coefficient,  $D_0$  is the free water diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.



ページ #1 - "酸性結晶質岩De/Da(Pu)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Pu:De(m2/s)	Pu:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor
0	Pu-239	13	Carbonate complex 2)	-2.0	Granite 1)			5.10e-14	-13.292				
1	Pu-239	13	Carbonate complex 2)	-2.0	Granite 1)			5.10e-14	-13.292				
2	Pu-239	13	Carbonate complex 2)	-2.0	Granite 1)			5.10e-14	-13.292				

ページ #2 - "酸性結晶質岩Da/Da(Pu)data"

	Pu:Da(m2/s)	Pu:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0			1	Through-diffusion	NaHCO3+NaNO2	9.30	5708q/ml(e-6M)		25.0		aerobic 3)
1			1	Through-diffusion	NaHCO3+NaNO2	9.30	5708q/ml(e-6M)		25.0		aerobic 3)
2			1	Through-diffusion	NaHCO3+NaNO2	9.30	5708q/ml(e-6M)		25.0		aerobic 3)

ページ #1 - “酸性結晶質岩De/Da(Pu)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 山口徹治：稲田花崗岩中におけるアクチニド元素の拡散、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010、pp.156-160、動燃事業団(1996.12)(1996).  
Yamaguchi, T.: Diffusivity of Actinides in a Granite from Inada, Ibaraki, Japan, PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.156-160, Power Reactor and Nuclear Fuel Development Corporation (1996. 10)(1996).

Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The kind of the rock is granite, which is one of the candidate host rocks for repository and major host rock in Japan.
- 2) The dominant chemical species of plutonium in the solution was estimated to be  $\text{Pu}(\text{OH})_2(\text{CO}_3)_2^{2-}$  in the literature from the pH of the porewater and the chemical conditions of the porewater.
- 3) The porewater used in this experiment is a mixture solution of  $\text{NaHCO}_3$  and  $\text{NaNO}_3$ , in which total concentrations of  $\text{CO}_3^{2-}$  and  $\text{NO}_2^-$  are respectively 0.1 and 0.01 M.  
The  $D_e$  described is an average value of data obtained from experiments in triplicate. The error of the  $D_e$  was evaluated to be  $5.1 \pm 2.2 \times 10^{-14} \text{ m}^2/\text{s}$  in the literature.
- 4) The  $D_o$  for Pu carbonate complex is not reported. The  $D_o$  for Pu is reported only for  $\text{PuO}_2^{2+}$ .
- 5) The formation factor (FF) was calculated based on the equation:  $\text{FF} = D_e/D_o$ . Where  $D_e$  and  $D_o$  is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 6) The geometric factor ( $G_f$ ) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f = D_p/D_o = D_e/(\phi \cdot D_o) = \text{FF}/\phi$ . Where  $D_p$  is the porewater diffusion coefficient,  $D_o$  is the free water diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "酸性結晶質岩De/Da(Se)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Se:De(m2/s)	Se:log De	Do(m2/s) 5)	Formation factor(FF) 6)	Geometric factor(Gf) 7)	Rock capacity factor
0	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	3.20e-12	-11.495	8.10e-10	0.00400	0.170	0.320
1	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	7.20e-13	-12.143	8.10e-10	0.000880	0.0390	0.170
2	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	1.90e-12	-11.721	8.10e-10	0.00230	0.100	0.360
3	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20			8.10e-10			
4	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20	2.40e-12	-11.620	8.10e-10	0.00300	0.0930	0.180
5	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20	3.30e-12	-11.481	8.10e-10	0.00410	0.130	0.350
6	Se	14	SeO32-	-2.0	Fracture fillings	2.43	5.60	5.70e-12	-11.244	8.10e-10	0.00700	0.130	0.700
7	Se	14	SeO32-	-2.0	Fracture fillings	2.43	5.60	4.80e-12	-11.310	8.10e-10	0.00600	0.110	1.31
8	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	1.90e-12	-11.721	8.10e-10	0.00230	0.100	
9	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20	2.90e-12	-11.538	8.10e-10	0.00360	0.110	
10	Se	14	SeO32-	-2.0	Granodiorite		2.30	1.90e-12	-11.721	8.10e-10	0.00230	0.100	
11	Se	14	SeO32-	-2.0	Altered granodiorite		3.20	2.90e-12	-11.538	8.10e-10	0.00360	0.110	
12	Se	14	SeO32-	-2.0	Fracture fillings		5.60	5.30e-12	-11.276	8.10e-10	0.00650	0.120	
13	Se	14	SeO42- 3)	-2.0	Granodiorite					1.00e-09			
14	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	1.90e-12	-11.721	8.10e-10	0.00230	0.100	
15	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20	2.90e-12	-11.538	8.10e-10	0.00360	0.100	

ページ #2 - "酸性結晶質岩De/Da(Se)data"

	Se:Da(m2/s)	Se:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	1.00e-10	-10.000	1	Through-diffusion	groundwater 1)	8.70	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
1	4.30e-12	-11.367	1	Through-diffusion	groundwater 1)	8.70	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
2	5.30e-12	-11.276	1	Through-diffusion	groundwater 1)	8.70	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
3	1.40e-12	-11.854	1	Through-diffusion	groundwater 1)	8.77	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
4	1.30e-11	-10.886	1	Through-diffusion	groundwater 1)	8.77	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
5	9.30e-12	-11.032	1	Through-diffusion	groundwater 1)	8.77	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
6	8.10e-12	-11.092	1	Through-diffusion	groundwater 1)	8.92	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
7	3.70e-12	-11.432	1	Through-diffusion	groundwater 1)	8.92	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
8	6.50e-12	-11.187	2	Through-diffusion	groundwater 1)	8.70	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
9	1.10e-11	-10.959	2	Through-diffusion	groundwater 1)	8.77	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
10	6.50e-12	-11.187	3	Through-diffusion	groundwater 1)	8.70			Temp. in G.B.		anaerobic 2)
11	1.10e-11	-10.959	3	Through-diffusion	groundwater 1)	8.77			Temp. in G.B.		anaerobic 2)
12	5.90e-12	-11.229	3	Through-diffusion	groundwater 1)	8.92			Temp. in G.B.		anaerobic 2)
13	4.30e-11	-10.367	4	Advection diffusion	Distilled water		SeO <sub>2</sub> (1000ppmSe)		probably room		aerobic 4)
14	6.50e-12	-11.187	5	Through-diffusion	groundwater 1)	8.70	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)
15	1.10e-11	-10.959	5	Through-diffusion	groundwater 1)	8.77	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic 2)

## ページ #1 - “酸性結晶質岩De/Da(Se)data”

## REFERENCE AND FOOTNOTE

## Reference:

- 1 Sato, H., Shibutani, T., Tachi, Y., Ota, K., Kenji, A., Yui, M.: Diffusion Behaviour of Nuclides Considering Pathways in Fractured Crystalline Rocks, PNC Technical Report, PNC TN1410 97-127, Power Reactor and Nuclear Fuel Corporation (1997. 8)(1997).  
佐藤治夫、澁谷朝紀、館幸男、太田久仁雄、天野憲治、油井三和：物質移行経路を考慮した割れ目を含む結晶質岩中の核種の拡散挙動、動燃技術資料、PNC TN 1410 97-127, 動燃事業団(1997. 8)(1997).
- 2 動燃事業団：地層処分研究開発の現状(平成8年度、動燃技術資料、PNC TN1410 96-071、pp.205-207、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.205-207, Power Reactor and Nuclear Fuel Corporation (1996. 12)(1996).
- 3 佐藤治夫、館幸男、澁谷朝紀、油井三和：物質移行経路を考慮した核種の拡散挙動、日本原子力学会1997年春の年会、L12、p.561(1997).  
Sato, H., Tachi, Y., Shibutani, T., Yui, M.: Diffusion Behavior of Nuclides Considered Pathways in Geological Materials, 1997 Annual Meeting of the Atomic Energy Society of Japan, L12, p.561 (1997).
- 4 向井悟、北尾秀夫、片岡伸一：亀裂性岩石中のI, Se及びCsを用いた移流・拡散試験、日本原子力学会1998年秋の大会、M25、p.842(1998).  
Mukai, S., Kitao, H., Kataoka, S.: Advection Diffusion Experiment Using I, Se and Cs in Fractured Rocks, 1998 Fall Meeting of the Atomic Energy Society of Japan, M25, p.842 (1998).
- 5 佐藤治夫、澁谷朝紀：亀裂性岩盤中における吸着・拡散係数、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010、pp.139-144、動燃事業団(1996.12)(1996).  
Sato, H., Shibutani, T.: PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.139-144, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).

## Footnote:

- 1) The in-situ groundwater sampled from in-situ test site was used for the experiments. The chemical composition of the groundwater is reported in more detail in the literature. Basically, the concentrations of Na, K, Ca, Si, F-, Cl-, SO42-, CO32-, HCO3- are shown in it.
- 2) The major mineral composition of the fracture fillings is calcite and stilbite, and those of the altered and intact granodiorite are quartz and plagioclase. The content of constituent chlorite in the altered granodiorite is a little higher than that in the intact granodiorite.  
The experiments only for Se were carried out under anaerobic conditions. Specifically, the experiments for Se were all performed in an N2-glove

ページ #2 - “酸性結晶質岩De/Da(Se)data”

- box kept less than 1ppm oxygen concentration.
- 3) The dominant species shown is predicted one, because the experiment was carried out under aerobic conditions. Selenium normally takes a valence of VI under aerobic conditions. In this study, it is presumed that the pH of the porewater was probably relatively high because the solution containing Se contacted with the rock.
  - 4) The advection diffusion experiments were carried out using granitic rocks with an artificial single fracture, which aperture was prepared to produce 50-100 $\mu$ m. The length of the rock samples is 17-18cm. The Da was calculated by fitting based on an equation considered advection-dispersion-sorption (retardation) and matrix diffusion from the concentration profile from the surface of the fracture.
  - 5) The Do values of SeO3<sup>2-</sup> and SeO4<sup>2-</sup> are estimated 8.1e-10 and 1.0e-9 m<sup>2</sup>/s at 25°C, respectively [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.170.].
  - 6) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
  - 7) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "酸性結晶質岩De/Da(Sm)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Sm:De(m2/s)	Sm:log De	Do(m2/s) 3)	Formation factor(FF) 4)	Geometric factor(Gf) 5)	Rock capacity factor
0	Sm	15	Sm3+	3.0	Granodiorite 2)	2.64	1.20	5.70e-13	-12.244	6.10e-10	0.000930	0.0780	0.0570
1	Sm	15	Sm3+	3.0	Granodiorite		0.880	5.70e-13	-12.244	6.10e-10	0.000930	0.110	
2	Sm	15	Sm3+	3.0	Granodiorite		1.00	5.74e-13	-12.241	6.10e-10	0.000940	0.0940	
3	Sm	15	Sm3+	3.0	Granodiorite			5.70e-13	-12.244	6.10e-10	0.000930		0.0570
4	Sm	15	Sm3+	3.0	Granodiorite 2)		0.880	5.70e-13	-12.244	6.10e-10	0.000930	0.110	



ページ #2 - "酸性結晶質岩Da/Da(Sm)data"

	Sm:Da(m2/s)	Sm:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	1.00e-11	-11.000	1	Through-diffusion	Distilled water	5.80	0.1M SmCl3 1)	137days	25.0		aerobic
1	1.00e-11	-11.000	2	Through-diffusion	Distilled water	6.00	0.1M SmCl3 1)		20.0		aerobic
2	1.00e-11	-11.000	3	Through-diffusion	Distilled water	6.00	0.1M SmCl3 1)		20.0		aerobic
3	1.00e-11	-11.000	4	Through-diffusion	Distilled water	5.80	0.1M SmCl3 1)		25.0		aerobic
4	1.00e-11	-11.000	5	Through-diffusion	Distilled water	6.00	0.1M SmCl3 1)		20.0		aerobic

ページ #1 - “酸性結晶質岩De/Da(Sm)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Sato, H., Shibutani, T., Yui, M.: Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone, Journal of Contaminant Hydrology, 26, pp.119-133, Elsevier Science B.V. (1997).
- 2 動燃事業団：地層処分研究開発の現状(平成5年度)、動燃技術資料、PNC TN1410 94-094、pp.131-133、動燃事業団(1994. 11)(1994).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1993-, PNC Technical Report, PNC TN1410 94-094, pp.131-133, Power Reactor and Nuclear Fuel Development Corporation (1994. 11)(1994).
- 3 佐藤治夫、澁谷朝紀、吉川英樹、油井三和：花崗閃緑岩中のCs, Ni, Smの拡散機構に関する研究、日本原子力学会1993年秋の大会、M29、p.667(1993).  
Sato, H., Shibutani, T., Yoshikawa, H., Yui, M.: A Study on Diffusion Mechanism for Cs, Ni and Sm in Granodiorite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M29, p.667 (1993).
- 4 動燃事業団：地層処分研究開発の現状(平成8年度)、動燃技術資料、PNC TN1410 96-071、pp.202-204、動燃事業団(1996. 12)(1996).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1996-, PNC Technical Report, PNC TN1410 96-071, pp.202-204, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 5 佐藤治夫、澁谷朝紀：緩衝材および岩石への核種の吸着・拡散メカニズム、動燃技術報、No.91、PNC TN8410 94-284、pp.71-89、動燃事業団(1994. 9)(1994).  
Sato, H., Shibutani, T.: Study on Adsorption and Diffusion of Nuclides in Buffer Material and Geosphere, PNC Technical Review, No.91, PNC TN 8410 94-284, pp.71-89, Power Reactor and Nuclear Fuel Development Corporation (1994. 9)(1994).

Footnote:

- 1) The tracer was used as a mixture of CsCl, NiCl<sub>2</sub> and SmCl<sub>3</sub>. Each reagent was dissolved to produce 0.1 M in distilled water.
- 2) The rock sample was sampled from Sori, Seta-gun, Gunma-ken, Japan. The rock is approximately equivalent to a standard sample in the chemical composition of Japanese granite in the Geological Survey of Japan. This kind of rock, one of the candidate host rocks for repository, is major rock in Japan.
- 3) The  $D_0$  of Sm<sup>3+</sup> is estimated 6.1e-10 m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169.].
- 4) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where  $D_e$  and  $D_0$  is the effective diffusion coefficient and free water diffusion coefficient, respectively.

ページ #2 - “酸性結晶質岩De/Da(Sm)data”

- 5) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf = D_p / D_o = D_e / (\phi \cdot D_o) = FF / \phi$ . Where  $D_p$  is the porewater diffusion coefficient,  $D_o$  is the free water diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "酸性結晶質岩De/Da(Sr)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Sr:De(m2/s)	Sr:log De	Do(m2/s) 11)	Formation factor(FF) 12)	Geometric factor(Gf) 13)	Rock capacity factor
0	Sr	18	Sr2+	2.0	Granite 1)		0.700	2.00e-13	-12.699	7.90e-10	0.000250	0.0360	0.0190
1	Sr	18	Sr2+	2.0	Granite 1)		0.700	2.10e-13	-12.678	7.90e-10	0.000270	0.0380	0.00200
2	Sr	18	Sr2+	2.0	Granite 1)		0.700	2.60e-13	-12.585	7.90e-10	0.000330	0.0470	0.0220
3	Sr	18	Sr2+	2.0	Granite 1)		0.700	2.60e-13	-12.585	7.90e-10	0.000330	0.0470	0.00300
4	Sr	18	Sr2+	2.0	Granite 1)		0.700	3.10e-13	-12.509	7.90e-10	0.000390	0.0560	0.0190
5	Sr	18	Sr2+	2.0	Granite 1)		0.700	3.60e-13	-12.444	7.90e-10	0.000460	0.0650	0.0140
6	Sr	18	Sr2+	2.0	Granite 1)		0.700	3.20e-12	-11.495	7.90e-10	0.00410	0.580	2.40
7	Sr	18	Sr2+	2.0	Granite 1)		0.700	9.50e-12	-11.022	7.90e-10	0.0120	1.70	1.50
8	Sr	18	Sr2+	2.0	Granite 1)		0.700	1.70e-11	-10.770	7.90e-10	0.0220	3.10	1.70
9	Sr	18	Sr2+	2.0	Granite 1)		0.680	2.10e-13	-12.678	7.90e-10	0.000270	0.0390	
10	Sr	18	Sr2+	2.0	Granite 1)		0.680	3.60e-13	-12.444	7.90e-10	0.000460	0.0670	
11	Sr	18	Sr2+	2.0	Granite 1)		0.680	3.10e-13	-12.509	7.90e-10	0.000390	0.0580	
12	Sr	18	Sr2+	2.0	Granite 1)		0.680	2.60e-13	-12.585	7.90e-10	0.000330	0.0480	
13	Sr	18	Sr2+	2.0	Granite 1)		0.680	2.60e-13	-12.585	7.90e-10	0.000330	0.0480	
14	Sr	18	Sr2+	2.0	Granite 1)		0.680	2.00e-13	-12.699	7.90e-10	0.000250	0.0370	
15	Sr	18	Sr2+	2.0	Granite 1)			2.70e-13	-12.589	7.90e-10	0.000340		
16	Sr	18	Sr2+	2.0	Granite 1)			1.00e-11	-11.000	7.90e-10	0.0130		
17	Sr	18	Sr2+	2.0	Granite 1)		0.700	2.70e-13	-12.589	7.90e-10	0.000340	0.0490	0.0130
18	Sr	18	Sr2+	2.0	Granite 7)		0.400	1.60e-12	-11.706	7.90e-10	0.00200	0.510	1.20
19	Sr	18	Sr2+	2.0	Granite 1)			1.00e-11	-11.000	7.90e-10	0.0130		2.00
20	Sr	18	Sr2+	2.0	Granite 1)			2.00e-11	-10.699	7.80e-10	0.0260		3.00
21	Sr	18	Sr2+	2.0	Granite 1)			1.80e-13	-12.745	7.90e-10	0.000180		0.0380
22	Sr	18	Sr2+	2.0	Granite 1)			3.20e-13	-12.495	7.90e-10	0.000410		
23	Sr	18	Sr2+	2.0	Granite 1)		0.800						
24	Sr	18	Sr2+	2.0	Granite 1)		0.800						
25	Sr	18	Sr2+	2.0	Granite 1)		0.800						
26	Sr	18	Sr2+	2.0	Granite 1)		0.800						

ページ #2 - "酸性結晶質岩Da/Da(Sr)data"

	Sr:Da(m2/s)	Sr:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	1.00e-11	-11.000	1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
1	1.00e-10	-10.000	1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
2	1.18e-11	-10.928	1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
3	8.67e-11	-10.062	1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
4	1.63e-11	-10.788	1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
5	2.57e-11	-10.590	1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
6	1.33e-12	-11.876	1	Through-diffusion	Deionized water	5.50	Sr(NO3)2 2)		25.0		aerobic
7	6.33e-12	-11.199	1	Through-diffusion	Deionized water	5.50	Sr(NO3)2 2)		25.0		aerobic
8	1.00e-11	-11.000	1	Through-diffusion	Deionized water	5.50	Sr(NO3)2 2)		25.0		aerobic
9			1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
10			1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
11			1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
12			1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
13			1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
14			1	Through-diffusion	0.1M KCl	4.00	Sr(NO3)2 2)		25.0		aerobic
15			2	Through-diffusion	0.1M KCl		1M Sr(NO3)2		probably room		aerobic 3)
16			2	Through-diffusion	Deionized water		1M Sr(NO3)2		probably room		aerobic 4)
17	2.08e-11	-10.682	3	Through-diffusion	0.1M KCl	4.00	5)		25.0		aerobic 6)
18	1.30e-11	-10.886	4	Through-diffusion	groundwater 8)		Sr2(3.4e-3M) 9)	300days	probably room		aerobic 10)
19	5.00e-12	-11.301	5	Through-diffusion	0.1M KCl	4.00			probably room		aerobic
20	6.67e-12	-11.176	5	Through-diffusion	0.1M KCl	4.00			probably room		aerobic
21	4.74e-12	-11.324	5	Through-diffusion	Deionized water	4.00					aerobic
22			5	Through-diffusion	Deionized water	4.00					aerobic
23	1.00e-11	-11.000	6	In-diffusion	Deionized water	4.30	2kBq/ml Sr 14)	7days	30.0		aerobic 15)
24	4.00e-12	-11.398	6	In-diffusion	Deionized water	6.00	2kBq/ml Sr 14)	7days	30.0		aerobic 15)
25	1.00e-11	-11.000	6	In-diffusion	Deionized water	7.70	2kBq/ml Sr 14)	7days	30.0		aerobic 15)
26	1.00e-11	-11.000	6	In-diffusion	Deionized water	9.40	2kBq/ml Sr 14)	7days	30.0		aerobic 15)

ページ #1 - “酸性結晶質岩De/Da(Sr)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 Yamaguchi, T., Sakamoto, Y., Senoo, M.: Consideration on Effective Diffusivity of Strontium in Granite, Journal of Nuclear Science and Technology, 30 (8), pp.796-803, the Atomic Energy Society of Japan (August 1993) (1993).
- 2 山口徹治：稲田花崗岩中におけるアクチニド元素の拡散、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010、pp.156-160、動燃事業団(1996.12)(1996).  
Yamaguchi, T.: Diffusivity of Actinides in a Granite from Inada, Ibaraki, Japan, PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.156-160, Power Reactor and Nuclear Fuel Development Corporation (1996. 12)(1996).
- 3 Yamaguchi, T., Sakamoto, Y., Nakayama, S., Vandergraaf, T. T.: Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, Journal of Contaminant Hydrology, 26, pp.109-117, Elsevier Science B.V.(1997).
- 4 山口徹治、中山真一：微小間隙中における拡散係数に対する吸着の影響、日本原子力学会1997年春の年会、L10、p.559(1997).  
Yamaguchi, T., Nakayama, S.: Effect of Sorption on Diffusivity in Micropores, 1997 Annual Meeting of the Atomic Energy Society of Japan, L10, p.559 (1997).
- 5 山口徹治、坂本義昭、妹尾宗明：岩石中におけるストロンチウムの表面拡散、日本原子力学会1992年春の年会、C19、p.119(1992).  
Yamaguchi, T., Sakamoto, Y., Senoo, M.: Surface Diffusion of Strontium in Rocks, 1992 Annual Meeting of the Atomic Energy Society of Japan, C19, p.119 (1992).
- 6 Idemitsu, K., Furuya, H., Hara, T., Inagaki, Y.: Migration of Cesium, Strontium and Cobalt in Water-Saturated Inada Granite, Journal of Nuclear Science and Technology, Vol.29, No.5, pp.454-460, the Atomic Energy Society of Japan (1992).

Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The rock sample was collected on the ground surface. The kind of the rock is granite, which is regarded as one of the candidate host rocks for repository and major rock in Japan.
- 2) The tracer was used as a mixture of Ba(NO<sub>3</sub>)<sub>2</sub>, BeSO<sub>4</sub>, CoCl<sub>2</sub>, MgCl<sub>2</sub>, NiCl<sub>2</sub> and Sr(NO<sub>3</sub>)<sub>2</sub>, what we call as a multi-tracer. Individual concentrations in the tracer are not reported.
- 3) The error of the De is  $2.7 \pm 0.6 \times 10^{-13}$  m<sup>2</sup>/s and distribution coefficient

ページ #2 - “酸性結晶質岩De/Da(Sr)data”

(Kd) in this case is reported to be 0.002 ml/g.

- 4) The error of the De is  $1.0 \pm 0.7e-11$  m<sup>2</sup>/s and the Kd in this case is reported to be 0.7 ml/g.
- 5) The tracer was used as a mixture of Ba, Co, Mg, Ni and Sr, what we call as a multi-tracer. Individual concentrations in the tracer are not reported.
- 6) The error of the De and rock capacity factor is  $2.7 \pm 0.6e-13$  m<sup>2</sup>/s and  $0.013 \pm 0.008$ , respectively.
- 7) The rock sample was sampled from a fracture zone at 240m in depth in the Canada Whiteshell Laboratories, Canada.
- 8) The groundwater was also sampled from a fracture zone at 240m in depth. The ionic strength of the porewater is  $\sim 0.01$ .
- 9) The tracer solution was prepared by adding a small amount of I-125 and Sr-85 in a non-radioactive SrI<sub>2</sub> solution, which concentration is  $3.4e-4$  M.
- 10) The porosity was calculated based on  $De = \phi \cdot Gf \cdot Do$  from De, free water diffusion coefficient (Do) and geometric factor (Gf). Where  $\phi$  is the porosity.
- 11) The Do of Sr<sup>2+</sup> is estimated  $7.9e-10$  m<sup>2</sup>/s at 25°C [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.168.].
- 12) The formation factor (FF) was calculated based on the equation:  $FF = De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 13) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf = Dp/Do = De/(\phi \cdot Do) = FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.
- 14) A mixture solution of Cs-134, Sr-85 and Co-60 was prepared as a tracer solution. The concentration of each tracer is approximately 2 kBq/ml.
- 15) De can not be obtained from this method. In this literature, two kinds of Da values, Df and Dm, are reported based on the model of grain-boundary diffusion in a crystalline solid. The Df is the diffusion in fissures which are perpendicular to the specimen surface, and the other is the diffusion into matrix which is parallel to the specimen surface. Since diffusion in rock matrix is controlled by Df, the Df was adopted in this database.

ページ #1 - "酸性結晶質岩De/Da(Tc)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Tc:De(m2/s)	Tc:log De	Do(m2/s) 3)	Formation factor(FF) 4)	Geometric factor(Gf) 5)	Rock capacity factor	Tc:Da(m2/s)
0	Tc-99	17	TcO4-	-1.0	Granite 1)		7.70	4.20e-14	-13.377	2.00e-09	2.10e-05	0.00210		
1	Tc-99	17	TcO4-	-1.0	Granite 1)		1.00	4.20e-14	-13.377	2.00e-09	2.10e-05	0.00210		4.20e-12



ページ #2 - "酸性結晶質岩De/Da(Tc)data"

	Tc:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0		1	Through-diffusion	Distilled water	8.55~8.60	1.4e4Bq/ml	20days	23.0		aerobic 2)
1	-11.377	2	Through-diffusion	Distilled water		1.4e4Bq/ml	20days	room		aerobic

ページ #1 - “酸性結晶質岩De/Da(Tc)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 動燃事業団：地層処分研究開発の現状(平成元年度)、動燃技術資料、PNC TN1410 91-009、pp.81-82、動燃事業団(1991. 3)(1991).  
Power Reactor and Nuclear Fuel Development Corporation: Current Status for Research and Development of Geological Disposal -FY1989-, PNC Technical Report, PNC TN1410 91-009, pp.81-82, Power Reactor and Nuclear Fuel Development Corporation (1991. 3)(1991).
- 2 佐藤治夫：緩衝材及び岩体中での核種の拡散・収着データ及びその機構、動燃技術資料「性能評価研究情報交換会報告書」、PNC TN1410 91-079、pp.275-286、動燃事業団(1991.10)(1991).  
H. Sato: PNC Technical Report "Report of Information Exchange Meeting on the Performance Assessment Research", PNC TN1410 91-079, pp.275-286, Power Reactor and Nuclear Fuel Development Corporation (1991. 10)(1991).

Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The kind of the rock is granite, which is regarded as one of the candidate host rocks for repository and major rock in Japan.
- 2) Since the breakthrough plots to calculate De are not enough, there is a possibility that the breakthrough curve did not reach steady-state. The porosity of the sample is too large for common granite, but the De is quite low. The reliability of this De is therefore judged to be not so high. Therefore, this porosity is not adopted in this report. The Da is not reported in this literature.
- 3) The Do of TcO<sub>4</sub><sup>-</sup> is estimated 2.0e-9 m<sup>2</sup>/s at 25°C [Sato, H., Yui, M. and Yoshikawa, H. (1996): Ionic Diffusion Coefficients of Cs<sup>+</sup>, Pb<sup>2+</sup>, Sm<sup>3+</sup>, Ni<sup>2+</sup>, SeO<sub>4</sub><sup>2-</sup> and TcO<sub>4</sub><sup>-</sup> in Free Water Determined from Conductivity Measurements, Journal of Nuclear Science and Technology, Vol.33, No.12, pp.950-955.].
- 4) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 5) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

ページ #1 - "酸性結晶質岩De/Da(U)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	U:De(m2/s)	U:log De	Do(m2/s) 6)	Formation factor(FF) 7)	Geometric factor(Gf) 8)
0	U-233	18	UO22+	2.0	Granite 1)		0.700	2.20e-14	-13.658	4.30e-10	6.10e-05	0.00730
1	U-233	18	UO22+	2.0	Granite 1)		0.700	4.20e-14	-13.377	4.30e-10	9.80e-05	0.0140
2	U-233	18	UO22+	2.0	Granite 1)		0.700	4.40e-14	-13.357	4.30e-10	1.00e-04	0.0150
3	U-233	18	UO22+	2.0	Biotitic granite 3)		0.700	2.20e-14	-13.658	4.30e-10	5.10e-05	0.00730
4	U-233	18	UO22+	2.0	Biotitic granite 3)		0.700	4.20e-14	-13.377	4.30e-10	9.80e-05	0.0140
5	U-233	18	UO22+	2.0	Biotitic granite 3)		0.700	4.40e-14	-13.357	4.30e-10	1.00e-04	0.0150
6	U-233	18	Carbonate complex 4)	-4.0	Granite 1)			1.40e-13	-12.854	7.20e-10	0.000190	
7	U-233	18	Carbonate complex 4)	-4.0	Granite 1)			1.40e-13	-12.854	7.20e-10	0.000190	
8	U-233	18	Carbonate complex 4)	-4.0	Granite 1)			1.40e-13	-12.854	7.20e-10	0.000190	
9	U-233	18	UO22+ 9)	2.0	Granite 1)		0.800					
10	U-233	18	UO22+ 9)	2.0	Granite 1)		0.800					
11	U-233	18	UO22+	2.0	Granite 1)		0.800					
12	U-233	18	UO22+	2.0	Granite 1)		0.800					

ページ #2 - "酸性結晶質岩De/Da(U)data"

	Rock capacity factor	U:Da(m <sup>2</sup> /s)	U:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	0.0180	1.22e-12	-11.914	1	Through-diffusion	0.1M KCl	4.00	640Bq/ml U-233		room		aerobic 2)
1	0.0180	2.33e-12	-11.633	1	Through-diffusion	0.1M KCl	4.00	640Bq/ml U-233		room		aerobic 2)
2	0.0180	2.44e-12	-11.613	1	Through-diffusion	0.1M KCl	4.00	640Bq/ml U-233		room		aerobic 2)
3	0.0110	2.00e-12	-11.699	2	Through-diffusion	0.1M KCl	4.00	7.7e-6M	about 350days	20-25(room)		aerobic
4	0.0250	1.68e-12	-11.775	2	Through-diffusion	0.1M KCl	4.00	7.7e-6M	about 350days	20-25(room)		aerobic
5	0.0120	3.67e-12	-11.435	2	Through-diffusion	0.1M KCl	4.00	7.7e-6M	about 350days	20-25(room)		aerobic
6				3	Through-diffusion	NaHCO <sub>3</sub> +NaNO <sub>2</sub>	8.30	560Bq/ml		25.0		aerobic 5)
7				3	Through-diffusion	NaHCO <sub>3</sub> +NaNO <sub>2</sub>	8.30	560Bq/ml		25.0		aerobic 5)
8				3	Through-diffusion	NaHCO <sub>3</sub> +NaNO <sub>2</sub>	8.30	560Bq/ml		25.0		aerobic 5)
9		6.00e-13	-12.222	4	In-diffusion	Deionized water	5.00	2e-5 M(Uranyl) 10)	54days	30.0		aerobic 11)
10		2.00e-12	-11.699	4	In-diffusion	Deionized water	5.00	2e-5 M(Uranyl) 10)	70days	30.0		aerobic 11)
11		2.00e-12	-11.699	5	In-diffusion	Deionized water	5.00	2e-5 M(Uranyl) 10)	58days	30.0		aerobic 11)
12		7.00e-12	-11.165	5	In-diffusion	Deionized water	5.00	2e-5 M(Uranyl) 10)	252days	30.0		aerobic 11)

ページ #1 - “酸性結晶質岩De/Da(U)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 山口徹治、坂本義昭、関根敬一：花崗岩中におけるUO<sub>22+</sub>の有効拡散係数、日本原子力学会1993年秋の大会、M28、p.666(1993).  
Yamaguchi, T., Sakamoto, Y., Sekine, K.: Effective Diffusivity of UO<sub>22+</sub> in Granite, 1993 Fall Meeting of the Atomic Energy Society of Japan, M28, p.666 (1993).
- 2 Yamaguchi, T., Sakamoto, Y., Nakayama, S., Vandergraaf, T. T.: Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, Journal of Contaminant Hydrology, 26, pp.109-117, Elsevier Science B.V.(1997).
- 3 山口徹治：稲田花崗岩中におけるアクチニド元素の拡散、動燃技術資料「地層処分研究情報交換会報告書」、PNC TN1100 96-010、pp.156-160、動燃事業団(1996.12)(1996).  
Yamaguchi, T.: Diffusivity of Actinides in a Granite from Inada, Ibaraki, Japan, PNC Technical Report "Report of Information Exchange Meeting on the Research and Development for Geological Disposal of Radioactive Waste", PNC TN1100 96-010, pp.156-160, Power Reactor and Nuclear Fuel Development Corporation (1996. 10)(1996).
- 4 Idemitsu, K., Furuya, H., Murayama, K., Inagaki, Y.: Diffusivity of Uranium(VI) in Water-Saturated Inada Granite, Scientific Basis for Nuclear Waste Management XV (Sombret, C. G. ed.), Materials Research Society Symposium Proceedings, Vol.257, pp.625-632, Materials Research Society, Pittsburgh, Pennsylvania (1992).
- 5 Idemitsu, K., Furuya, H., Murayama, K., Asao, H., Inagaki, Y.: Primary Diffusion Path of Uranium(VI) in Water-Saturated Inada Granite, Proceedings of the International Conference on Nuclear Waste Management and Environmental Remediation (Alexandra, D., Baker, R. and Kohout, R. eds.), the American Society of Mechanical Engineers, pp.207-212 (1993).

Footnote:

- 1) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The kind of the rock is granite, which is regarded as one of the candidate host rocks for repository and major host rock in Japan.
- 2) The rock capacity factor was averaged. The actual rock capacity factor was between 0.011 and 0.025.  
The Da values were calculated from the average rock capacity factor and De values.
- 3) The biotitic granite was sampled from ground surface of the Inada mine, Ibaraki-ken, Japan.
- 4) The dominant chemical species of uranium in the solution was estimated

ページ #2 - “酸性結晶質岩De/Da(U)data”

to be  $\text{UO}_2(\text{CO}_3)_3^{4-}$  from the pH of the solution and the chemical conditions of the porewater in the literature.

- 5) The porewater used in this experiment is a mixture of  $\text{NaHCO}_3$  and  $\text{NaNO}_2$  and the total concentrations of  $\text{CO}_3^{2-}$  and  $\text{NO}_2^-$  in the tracer are respectively 0.1 and 0.01 M.  
The  $D_e$  described is an average value of data obtained from experiments in triplicate. The error of the  $D_e$  was evaluated to be  $1.4 \pm 0.1 \times 10^{-13} \text{ m}^2/\text{s}$  in the literature.
- 6) The  $D_o$  of  $\text{UO}_2^{2+}$  is estimated  $4.3 \times 10^{-10} \text{ m}^2/\text{s}$  at  $25^\circ\text{C}$  [Marcus, Y. (1997): Ion Properties, Marcel Dekker, Inc., New York, U. S. A., p.169.]. The  $D_o$  of  $\text{UO}_2(\text{CO}_3)_3^{4-}$  is estimated  $7.2 \pm 0.5 \times 10^{-10} \text{ m}^2/\text{s}$  at  $25^\circ\text{C}$  [literature 3]. This  $D_o$ , not directly measured value, was estimated based on the Stokes equation by the ionic radius calculated from the molecular structure.
- 7) The formation factor (FF) was calculated based on the equation:  $\text{FF} = D_e/D_o$ . Where  $D_e$  and  $D_o$  is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 8) The geometric factor (Gf) is also called tortuosity factor ( $T_f$ ), and was calculated based on the equation:  $G_f = D_p/D_o = D_e/(\phi \cdot D_o) = \text{FF}/\phi$ . Where  $D_p$  is the porewater diffusion coefficient,  $D_o$  is the free water diffusion coefficient,  $D_e$  is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.
- 9) The species of U in the porewater has not been described, but it is probably  $\text{UO}_2^{2+}$  judging from the pH of the porewater (pH5.0).
- 10)  $\text{UO}_2$  was dissolved in nitric acid at  $20^\circ\text{C}$  and diluted. An uranyl solution of  $2 \times 10^{-5} \text{ M U(VI)}$  which pH is 3.0, was used in the experiment.
- 11) The porosity is reported to be  $0.8 \pm 0.05\%$  in the literature.  $D_e$  can not be obtained from this method. In this literature, two kinds of  $D_a$  values,  $D_f$  and  $D_m$  are reported. The  $D_f$  is the diffusion in fissures which are perpendicular to the specimen surface, and the other is the diffusion into matrix which is parallel to the specimen surface. Since diffusion in rock matrix is controlled by  $D_f$ , the  $D_f$  was adopted in this database.

ページ #1 - "酸性結晶質岩De/Da(分-)data"

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	Uranine:De(m2/s)	Uranine:log De	Do(m2/s) 4)	Formation factor(FF) 5)	Geometric factor(Gf) 6)	Rock capacity factor
0	Uranine 1)	19			Granite 2)	2.60	0.740	4.70e-14	-13.328	4.50e-10	1.00e-04	0.0140	0.0150
1	Uranine 1)	19			Granite 2)	2.60	0.740	3.50e-14	-13.458	4.50e-10	7.80e-05	0.0110	0.0170
2	Uranine 1)	19			Granite 2)	2.60	0.740	5.00e-14	-13.301	4.50e-10	0.000110	0.0150	0.0710
3	Uranine 1)	19			Granite 2)	2.60	0.700	4.70e-14	-13.328	4.50e-10	1.00e-04	0.0150	0.0150
4	Uranine 1)	19			Granite 2)	2.60	0.700	3.50e-14	-13.458	4.50e-10	7.80e-05	0.0110	0.0170
5	Uranine 1)	19			Granite 2)	2.60	0.700	5.00e-14	-13.301	4.50e-10	0.000110	0.0150	0.0710

ページ #2 - "酸性結晶質岩De/Da(ケレン)data"

	Uranine:Da(m2/s)	Uranine:log Da	Literature No.	Method	Porewater	pH	Tracer	Contact time	Temperature(℃)	Eh(mV)	Others
0	3.10e-12	-11.509	1	Through-diffusion	Distilled water		10000ppm uranine	150days	probably room		aerobic 3)
1	2.10e-12	-11.878	1	Through-diffusion	Distilled water		10000ppm uranine	150days	probably room		aerobic 3)
2	7.00e-13	-12.155	1	Through-diffusion	Distilled water		10000ppm uranine	150days	probably room		aerobic 3)
3	3.10e-12	-11.509	2	Through-diffusion			1%uranine solution		room		aerobic 3)
4	2.10e-12	-11.678	2	Through-diffusion			1%uranine solution		room		aerobic 3)
6	7.00e-13	-12.155	2	Through-diffusion			1%uranine solution		room		aerobic 3)



ページ #1 - “酸性結晶質岩De/Da(ウラニ)data”

REFERENCE AND FOOTNOTE

Reference:

- 1 熊田政弘、岩井孝幸、佐川民雄、鈴木隆次、西山勝栄：岩石コアを用いた放射性核種の拡散実験、JAERI-M 90-179、日本原子力研究所(1990. 10)(1990).  
Kumata, M., Iwai, T., Sagawa, T., Suzuki, T., Nishiyama, K.: Diffusion Experiment of a Radionuclide in Granitic Rock Cores, JAERI Report, JAERI-M 90-179, Japan Atomic Energy Research Institute (1990. 10) (1990).
- 2 Yamaguchi, T., Sakamoto, Y., Nakayama, S., Vandergraaf, T. T.: Effective Diffusivity of the Uranyl Ion in a Granite from Inada, Ibaraki, Japan, Journal of Contaminant Hydrology, 26, pp.109-117, Elsevier Science B.V.(1997).

Footnote:

- 1) Uranine is an aromatic organic compound,  $C_{20}H_{10}O_5Na_2$ , with a molecular weight of 376.28. This was used as a non-sorbing tracer.
- 2) The rock sample was sampled from Inada, Ibaraki-ken, Japan. The kind of the rock is granite, which is regarded as one of the candidate host rocks for repository and major rock in Japan.
- 3) The Da values were calculated from rock capacity factors and De values. The porosity of the granite sample described was averaged based on data obtained from porosity measurements by water saturation method (water intrusion method).
- 4) The Do of uranine, not directly measured value, has been estimated  $4.5e-10$  m<sup>2</sup>/s at 25°C based on the Hyduk-Laudie equation [Skagius, K. and Neretnieks, I. (1985): Properties and Diffusivities of Some Non-sorbing Species in Crystalline Rocks, SKB 85-03.]. The same Do has been estimated also based on the Stokes equation by the ionic radius calculated from the molecular structure by Yamaguchi et al. [literature 2].
- 5) The formation factor (FF) was calculated based on the equation:  $FF=De/Do$ . Where De and Do is the effective diffusion coefficient and free water diffusion coefficient, respectively.
- 6) The geometric factor (Gf) is also called tortuosity factor (Tf), and was calculated based on the equation:  $Gf=Dp/Do=De/(\phi \cdot Do)=FF/\phi$ . Where Dp is the porewater diffusion coefficient, Do is the free water diffusion coefficient, De is the effective diffusion coefficient,  $\phi$  is the porosity and FF is the formation factor.

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
0	Ba	1	Ba2+	2.0	Granite		0.680	2.10e-13	-12.678	8.50e-10
1	Ba	1	Ba2+	2.0	Granite		0.680	3.50e-13	-12.456	8.50e-10
2	Ba	1	Ba2+	2.0	Granite		0.680	3.00e-13	-12.523	8.50e-10
3	Ba	1	Ba2+	2.0	Granite		0.680	2.50e-13	-12.602	8.50e-10
4	Ba	1	Ba2+	2.0	Granite		0.680	2.90e-13	-12.538	8.50e-10
5	Ba	1	Ba2+	2.0	Granite		0.680	1.90e-13	-12.721	8.50e-10
6	Be	2	Be2+	2.0	Granite		0.680	3.00e-13	-12.523	6.00e-10
7	Be	2	Be2+	2.0	Granite		0.680	3.30e-13	-12.481	6.00e-10
8	Cl	3	Cl-	-1.0	Granodiorite		0.880	4.50e-13	-12.347	2.00e-09
9	Cl	3	Cl-	-1.0	Granodiorite		0.880	7.10e-13	-12.149	2.00e-09
10	Cl	3	Cl-	-1.0	Granodiorite		0.880	5.10e-13	-12.292	2.00e-09
11	Cl	3	Cl-	-1.0	Granodiorite		0.990	4.50e-13	-12.347	2.00e-09
12	Cl	3	Cl-	-1.0	Granodiorite		0.990	7.10e-13	-12.149	2.00e-09
13	Cl	3	Cl-	-1.0	Granodiorite		0.990	5.10e-13	-12.292	2.00e-09
14	Cl	3	Cl-	-1.0	Granodiorite	2.64	1.20	4.50e-13	-12.347	2.00e-09
15	Cl	3	Cl-	-1.0	Granodiorite	2.64	1.20	7.10e-13	-12.149	2.00e-09
16	Cl	3	Cl-	-1.0	Granodiorite	2.64	1.20	5.10e-13	-12.292	2.00e-09
17	Cl	3	Cl-	-1.0	Granodiorite	2.64	1.20	7.40e-13	-12.131	2.00e-09
18	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	1.70e-11	-10.770	2.00e-09
19	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	8.10e-12	-11.092	2.00e-09
20	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	4.80e-12	-11.319	2.00e-09
21	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	1.40e-11	-10.854	2.00e-09
22	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	6.90e-12	-11.161	2.00e-09
23	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	3.90e-12	-11.409	2.00e-09
24	Cl	3	Cl-	-1.0	Fracture fillings	2.43	5.60	2.10e-11	-10.678	2.00e-09
25	Cl	3	Cl-	-1.0	Fracture fillings	2.43	5.60	1.40e-11	-10.854	2.00e-09
26	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	1.00e-11	-11.000	2.00e-09
27	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	8.30e-12	-11.081	2.00e-09
28	Cl	3	Cl-	-1.0	Fracture fillings	2.43	5.60	1.80e-11	-10.745	2.00e-09
29	Cl	3	Cl-	-1.0	Granodiorite		2.30	1.00e-11	-11.000	2.00e-09
30	Cl	3	Cl-	-1.0	Altered granodiorite		3.20	8.30e-12	-11.081	2.00e-09
31	Cl	3	Cl-	-1.0	Fracture fillings		5.60	1.80e-11	-10.745	2.00e-09
32	Cl	3	Cl-	-1.0	Granodiorite		1.00	7.41e-13	-12.130	2.00e-09
33	Cl	3	Cl-	-1.0	Granite		0.990	4.50e-13	-12.347	2.00e-09

	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH
0	0.000250	0.0360				Through-diffusion	0.1M KCl	4.00
1	0.000410	0.0610				Through-diffusion	0.1M KCl	4.00
2	0.000350	0.0520				Through-diffusion	0.1M KCl	4.00
3	0.000290	0.0430				Through-diffusion	0.1M KCl	4.00
4	0.000340	0.0500				Through-diffusion	0.1M KCl	4.00
5	0.000220	0.0330				Through-diffusion	0.1M KCl	4.00
6	5.00e-05	0.00740				Through-diffusion	0.1M KCl	4.00
7	5.50e-05	0.00810				Through-diffusion	0.1M KCl	4.00
8	0.000230	0.0260				Through-diffusion	Distilled water	
9	0.000360	0.0410				Through-diffusion	Distilled water	
10	0.000260	0.0300				Through-diffusion	Distilled water	
11	0.000230	0.0230				Through-diffusion	Distilled water	
12	0.000360	0.0360				Through-diffusion	Distilled water	
13	0.000260	0.0260				Through-diffusion	Distilled water	
14	0.000230	0.0190				Through-diffusion	Distilled water	5.50
15	0.000360	0.0300				Through-diffusion	Distilled water	5.50
16	0.000260	0.0220				Through-diffusion	Distilled water	5.50
17	0.000370	0.0310	0.0370	2.00e-11	-10.700	Through-diffusion	Distilled water	5.80
18	0.00850	0.370	3.00	5.60e-12	-11.252	Through-diffusion	groundwater	8.70
19	0.00410	0.180	1.20	6.70e-12	-11.174	Through-diffusion	groundwater	8.70
20	0.00240	0.100	0.280	1.70e-11	-10.770	Through-diffusion	groundwater	8.70
21	0.00700	0.220	2.50	5.70e-12	-11.244	Through-diffusion	groundwater	8.77
22	0.00350	0.110	1.30	5.50e-12	-11.260	Through-diffusion	groundwater	8.77
23	0.00200	0.0630	0.200	2.00e-11	-10.699	Through-diffusion	groundwater	8.77
24	0.0110	0.200	1.20	1.80e-11	-10.745	Through-diffusion	groundwater	8.92
25	0.00700	0.130	0.410	3.40e-11	-10.469	Through-diffusion	groundwater	8.92
26	0.00500	0.220		9.80e-12	-11.009	Through-diffusion	groundwater	8.70
27	0.00420	0.130		1.00e-11	-11.000	Through-diffusion	groundwater	8.77
28	0.00900	0.160		2.60e-11	-10.585	Through-diffusion	groundwater	8.92
29	0.00500	0.220		9.80e-12	-11.009	Through-diffusion	groundwater	8.70
30	0.00420	0.130		1.00e-11	-11.000	Through-diffusion	groundwater	8.77
31	0.00900	0.160		2.60e-11	-10.585	Through-diffusion	groundwater	8.92
32	0.000370	0.0370		2.00e-11	-10.699	Through-diffusion	Distilled water	6.00
33	0.000230	0.0230				Through-diffusion	Distilled water	5.50

	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
0	Ba(NO3)2		25.0		aerobic
1	Ba(NO3)2		25.0		aerobic
2	Ba(NO3)2		25.0		aerobic
3	Ba(NO3)2		25.0		aerobic
4	Ba(NO3)2		25.0		aerobic
5	Ba(NO3)2		25.0		aerobic
6	BeSO4		25.0		aerobic
7	BeSO4		25.0		aerobic
8	60000ppmCl(NaCl)	90days	room		aerobic
9	60000ppmCl(NaCl)	90days	room		aerobic
10	60000ppmCl(NaCl)	90days	room		aerobic
11	60000ppmCl(NaCl)	90days	room		aerobic
12	60000ppmCl(NaCl)	90days	room		aerobic
13	60000ppmCl(NaCl)	90days	room		aerobic
14	10%NaCl	90days	25.0		aerobic
15	10%NaCl	90days	25.0		aerobic
16	10%NaCl	90days	25.0		aerobic
17	0.6M Cl 3)		25.0		aerobic
18	0.06M(2127ppm)	30days	25.0		aerobic
19	0.06M(2127ppm)	30days	25.0		aerobic
20	0.06M(2127ppm)	30days	25.0		aerobic
21	0.06M(2127ppm)	30days	25.0		aerobic
22	0.06M(2127ppm)	30days	25.0		aerobic
23	0.06M(2127ppm)	30days	25.0		aerobic
24	0.06M(2127ppm)	30days	25.0		aerobic
25	0.06M(2127ppm)	30days	25.0		aerobic
26	0.06M(2127ppm)	30days	25.0		aerobic
27	0.06M(2127ppm)	30days	25.0		aerobic
28	0.06M(2127ppm)	30days	25.0		aerobic
29			25.0		aerobic
30			25.0		aerobic
31			25.0		aerobic
32	0.6M Cl		20.0		aerobic
33	60000ppmCl(NaCl)		probably room		aerobic

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
34	Cl	3	Cl-	-1.0	Granite		0.990	7.10e-13	-12.149	2.00e-09
35	Cl	3	Cl-	-1.0	Granite		0.990	5.10e-13	-12.292	2.00e-09
36	Cl	3	Cl-	-1.0	Granodiorite			7.40e-13	-12.131	2.00e-09
37	Cl	3	Cl-	-1.0	Granodiorite	2.73	2.30	1.00e-11	-11.000	2.00e-09
38	Cl	3	Cl-	-1.0	Altered granodiorite	2.61	3.20	8.30e-12	-11.081	2.00e-09
39	Cl	3	Cl-	-1.0	Fracture fillings	2.43	5.60	1.80e-11	-10.745	2.00e-09
40	Co	4	Co2+	2.0	Granite		0.680	2.10e-13	-12.678	7.30e-10
41	Co	4	Co2+	2.0	Granite		0.680	3.40e-13	-12.469	7.30e-10
42	Co	4	Co2+	2.0	Granite		0.680	3.10e-13	-12.509	7.30e-10
43	Co	4	Co2+	2.0	Granite		0.680	2.60e-13	-12.585	7.30e-10
44	Co	4	Co2+	2.0	Granite		0.680	2.20e-13	-12.658	7.30e-10
45	Co	4	Co2+	2.0	Granite		0.680	3.50e-13	-12.456	7.30e-10
46	Co	4	Co2+	2.0	Granite		0.680	2.50e-13	-12.602	7.30e-10
47	Co	4	Co2+	2.0	Granite		0.680	1.60e-13	-12.796	7.30e-10
48	Co-60	4	Probably Co2+	2.0	Granite		0.800			7.30e-10
49	Co-60	4	Probably Co2+	2.0	Granite		0.800			7.30e-10
50	Co-60	4	Probably Co2+	2.0	Granite		0.800			7.30e-10
51	Co-60	4	Probably Co2+	2.0	Granite		0.800			7.30e-10
52	Cs	5	Cs+	1.0	Granodiorite	2.64	1.20	1.40e-12	-11.854	2.10e-09
53	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	7.80e-12	-11.108	2.10e-09
54	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	2.50e-12	-11.602	2.10e-09
55	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	5.00e-12	-11.301	2.10e-09
56	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	2.50e-12	-11.602	2.10e-09
57	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	4.80e-12	-11.319	2.10e-09
58	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	6.00e-12	-11.222	2.10e-09
59	Cs	5	Cs+	1.0	Fracture fillings	2.43	5.60	1.40e-11	-10.854	2.10e-09
60	Cs	5	Cs+	1.0	Fracture fillings	2.43	5.60	2.20e-11	-10.658	2.10e-09
61	Cs	5	Cs+	1.0	Granodiorite		0.880	1.40e-12	-11.854	2.10e-09
62	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	5.10e-12	-11.292	2.10e-09
63	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	1.20e-11	-10.921	2.10e-09
64	Cs	5	Cs+	1.0	Fracture fillings	2.43	5.60	1.80e-11	-10.745	2.10e-09
65	Cs	5	Cs+	1.0	Granodiorite		2.30	5.10e-12	-11.292	2.10e-09
66	Cs	5	Cs+	1.0	Altered granodiorite		3.20	1.20e-11	-10.921	2.10e-09
67	Cs	5	Cs+	1.0	Fracture fillings		5.60	1.80e-11	-10.745	2.10e-09

	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH
34	0.000360	0.0360				Through-diffusion	Distilled water	5.50
35	0.000260	0.0260				Through-diffusion	Distilled water	5.50
36	0.000370		0.140	2.00e-11	-10.699	Through-diffusion	Distilled water	5.80
37	0.00500	0.220		9.80e-12	-11.009	Through-diffusion	groundwater	8.70
38	0.00420	0.130		1.00e-11	-11.000	Through-diffusion	groundwater	8.77
39	0.00900	0.160		2.60e-11	-10.585	Through-diffusion	groundwater	8.92
40	0.000290	0.0420				Through-diffusion	0.1M KCl	4.00
41	0.000470	0.0680				Through-diffusion	0.1M KCl	4.00
42	0.000420	0.0620				Through-diffusion	0.1M KCl	4.00
43	0.000360	0.0520				Through-diffusion	0.1M KCl	4.00
44	0.000300	0.0440				Through-diffusion	0.1M KCl	4.00
45	0.000480	0.0710				Through-diffusion	0.1M KCl	4.00
46	0.000340	0.0500				Through-diffusion	0.1M KCl	4.00
47	0.000220	0.0320				Through-diffusion	0.1M KCl	4.00
48				2.00e-11	-10.699	In-diffusion	Deionized water	4.30
49				8.00e-12	-11.697	In-diffusion	Deionized water	6.00
50				2.00e-11	-10.699	In-diffusion	Deionized water	7.70
51				2.00e-11	-10.699	In-diffusion	Deionized water	9.40
52	0.000670	0.0560	0.140	1.00e-11	-11.000	Through-diffusion	Distilled water	5.80
53	0.00370	0.160	0.650	1.20e-11	-10.921	Through-diffusion	groundwater	8.70
54	0.00120	0.0520	0.190	1.30e-11	-10.886	Through-diffusion	groundwater	8.70
55	0.00240	0.100	0.700	7.10e-12	-11.149	Through-diffusion	groundwater	8.70
56	0.00120	0.0380	0.700	3.20e-12	-11.495	Through-diffusion	groundwater	8.77
57	0.00230	0.0720	0.910	5.30e-12	-11.276	Through-diffusion	groundwater	8.77
58	0.00290	0.0910	1.20	5.10e-12	-11.292	Through-diffusion	groundwater	8.77
59	0.00670	0.120	1.30	1.10e-11	-10.959	Through-diffusion	groundwater	8.92
60	0.01000	0.180	1.70	1.30e-11	-10.886	Through-diffusion	groundwater	8.92
61	0.000670	0.0760		1.00e-11	-11.000	Through-diffusion	Distilled water	6.00
62	0.00240	0.100		1.10e-11	-10.959	Through-diffusion	groundwater	8.70
63	0.000570	0.0180		4.50e-12	-11.347	Through-diffusion	groundwater	8.77
64	0.000860	0.0150		1.20e-11	-10.921	Through-diffusion	groundwater	8.92
65	0.00240	0.100		1.10e-11	-10.959	Through-diffusion	groundwater	8.70
66	0.000570	0.0180		4.50e-12	-11.347	Through-diffusion	groundwater	8.77
67	0.000860	0.0150		1.20e-11	-10.921	Through-diffusion	groundwater	8.92

	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
34	60000ppmCl(NaCl)		probably room		aerobic
35	60000ppmCl(NaCl)		probably room		aerobic
36	0.6M.Cl		25.0		aerobic
37	0.06M(2127ppm)	30days	25.0		aerobic
38	0.06M(2127ppm)	30days	25.0		aerobic
39	0.06M(2127ppm)	30days	25.0		aerobic
40	CoCl2		25.0		aerobic
41	CoCl2		25.0		aerobic
42	CoCl2		25.0		aerobic
43	CoCl2		25.0		aerobic
44	CoCl2		25.0		aerobic
45	CoCl2		25.0		aerobic
46	CoCl2		25.0		aerobic
47	CoCl2		25.0		aerobic
48	2KBq/ml	7days	30.0		aerobic
49	2KBq/ml	7days	30.0		aerobic
50	2KBq/ml	7days	30.0		aerobic
51	2KBq/ml	7days	30.0		aerobic
52	0.1M CsCl		25.0		aerobic
53	0.01M(1330ppm)CsCl	30days	25.0		aerobic
54	0.01M(1330ppm)CsCl	30days	25.0		aerobic
55	0.01M(1330ppm)CsCl	30days	25.0		aerobic
56	0.01M(1330ppm)CsCl	30days	25.0		aerobic
57	0.01M(1330ppm)CsCl	30days	25.0		aerobic
58	0.01M(1330ppm)CsCl	30days	25.0		aerobic
59	0.01M(1330ppm)CsCl	30days	25.0		aerobic
60	0.01M(1330ppm)CsCl	30days	25.0		aerobic
61	0.1M CsCl		25.0		aerobic
62	0.01M(1330ppm)CsCl	30days	25.0		aerobic
63	0.01M(1330ppm)CsCl	30days	25.0		aerobic
64	0.01M(1330ppm)CsCl	30days	25.0		aerobic
65			25.0		aerobic
66			25.0		aerobic
67			25.0		aerobic

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
68	Cs	5	Cs+	1.0	Granodiorite		1.00	1.36e-12	-11.866	2.10e-09
69	Cs	5	Cs+	1.0	Granodiorite			1.40e-12	-11.854	2.10e-09
70	Cs	5	Cs+	1.0	Granodiorite	2.73	2.30	5.10e-12	-11.292	2.10e-09
71	Cs	5	Cs+	1.0	Altered granodiorite	2.61	3.20	1.20e-11	-10.921	2.10e-09
72	Cs	5	Cs+	1.0	Fracture fillings	2.43	5.60	1.80e-11	-10.745	2.10e-09
73	Cs	5	Cs+	1.0	Granodiorite		0.880	1.40e-12	-11.854	2.10e-09
74	Cs-134	5	Cs+	1.0	Granite					2.10e-09
75	Cs-134	5	Cs+	1.0	Biotitic granite	2.70	0.374			2.10e-09
76	Cs-134	5	Cs+	1.0	Granite		0.800			2.10e-09
77	Cs-134	5	Cs+	1.0	Granite		0.800			2.10e-09
78	Cs-134	5	Cs+	1.0	Granite		0.800			2.10e-09
79	Cs-134	5	Cs+	1.0	Granite		0.800			2.10e-09
80	Cs-134	5	Cs+	1.0	Granite		0.800			2.10e-09
81	Cs-134	5	Cs+	1.0	Granite		0.800			2.10e-09
82	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	5.00e-12	-11.301	2.10e-09
83	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	4.80e-12	-11.319	2.10e-09
84	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	6.20e-12	-11.208	2.10e-09
85	H-3	6	H2O	0.0	Altered granodiorite	2.61	3.20	1.70e-11	-10.770	2.10e-09
86	H-3	6	H2O	0.0	Altered granodiorite	2.61	3.20	2.00e-11	-10.699	2.10e-09
87	H-3	6	H2O	0.0	Altered granodiorite	2.61	3.20	1.70e-11	-10.770	2.10e-09
88	H-3	6	H2O	0.0	Fracture fillings	2.43	5.60	2.70e-11	-10.569	2.10e-09
89	H-3	6	H2O	0.0	Fracture fillings	2.43	5.60	2.80e-11	-10.553	2.10e-09
90	H-3	6	H2O	0.0	Fracture fillings	2.43	5.60	1.60e-11	-10.796	2.10e-09
91	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	5.30e-12	-11.276	2.10e-09
92	H-3	6	H2O	0.0	Altered granodiorite	2.61	3.20	1.80e-11	-10.745	2.10e-09
93	H-3	6	H2O	0.0	Granodiorite		2.30	5.30e-12	-11.276	2.10e-09
94	H-3	6	H2O	0.0	Altered granodiorite		3.20	1.80e-11	-10.745	2.10e-09
95	H-3	6	H2O	0.0	Fracture fillings		5.60	2.40e-11	-10.620	2.10e-09
96	H-3	6	H2O	0.0	Granodiorite	2.73	2.30	5.30e-12	-11.276	2.10e-09
97	H-3	6	H2O	0.0	Altered granodiorite	2.61	3.20	1.80e-11	-10.745	2.10e-09
98	I	7	I-	-1.0	Granite		0.800	2.60e-12	-11.585	2.00e-09
99	I	7	I-	-1.0	Granite		0.800	1.90e-12	-11.721	2.00e-09
100	I	7	I-	-1.0	Granite		0.770	2.60e-12	-11.585	2.00e-09
101	I	7	I-	-1.0	Granite		0.770	1.90e-12	-11.721	2.00e-09



	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH
68	0.000650	0.0650		1.00e-11	-11.000	Through-diffusion	Distilled water	6.00
69	0.000670		0.140	1.00e-11	-11.000	Through-diffusion	Distilled water	5.80
70	0.00240	0.100		1.10e-11	-10.959	Through-diffusion	groundwater	8.70
71	0.000570	0.0180		4.50e-12	-11.347	Through-diffusion	groundwater	8.77
72	0.000860	0.0150		1.20e-11	-10.921	Through-diffusion	groundwater	8.92
73	0.000670	0.0760		1.00e-11	-11.000	Through-diffusion	Distilled water	6.00
74				3.10e-14	-13.509	Advection diffusion	Distilled water	
75				8.00e-12	-11.097	In-diffusion	Distilled water	
76				1.00e-11	-11.000	In-diffusion	Deionized water	4.30
77				1.00e-11	-11.000	In-diffusion	Deionized water	6.00
78				1.00e-11	-11.000	In-diffusion	Deionized water	7.70
79				1.00e-11	-11.000	In-diffusion	Deionized water	9.40
80				1.50e-11	-10.824	In-diffusion	Deionized water	4.20
81				3.00e-11	-10.523	In-diffusion	Deionized water	4.20
82	0.00240	0.100	0.00	2.20e-10	-9.6580	Through-diffusion	groundwater	8.70
83	0.00230	0.100	0.00	2.10e-10	-9.6780	Through-diffusion	groundwater	8.70
84	0.00300	0.130	0.0330	1.90e-10	-9.7210	Through-diffusion	groundwater	8.70
85	0.00810	0.250	0.00	5.30e-10	-9.2760	Through-diffusion	groundwater	8.77
86	0.00950	0.300	0.00	6.30e-10	-9.2010	Through-diffusion	groundwater	8.77
87	0.00810	0.250	0.00	5.30e-10	-9.2760	Through-diffusion	groundwater	8.77
88	0.0130	0.230	0.0560	4.80e-10	-9.3190	Through-diffusion	groundwater	8.92
89	0.0130	0.240	0.0560	5.00e-10	-9.3010	Through-diffusion	groundwater	8.92
90	0.00760	0.140	0.0560	2.90e-10	-9.5380	Through-diffusion	groundwater	8.92
91	0.00250	0.110		2.10e-10	-9.6780	Through-diffusion	groundwater	8.70
92	0.00860	0.180		5.60e-10	-9.2520	Through-diffusion	groundwater	8.77
93	0.00250	0.110		2.10e-10	-9.6780	Through-diffusion	groundwater	8.70
94	0.00860	0.180		5.60e-10	-9.2520	Through-diffusion	groundwater	8.77
95	0.0110	0.200		4.30e-10	-9.3670	Through-diffusion	groundwater	8.92
96	0.00250	0.110		2.10e-10	-9.6780	Through-diffusion	groundwater	8.70
97	0.00860	0.180		5.60e-10	-9.2520	Through-diffusion	groundwater	8.77
98	0.00130	0.160	0.0950	2.74e-11	-10.562	Through-diffusion	Deionized water	
99	0.000950	0.120	0.140	1.36e-11	-10.866	Through-diffusion	Deionized water	
100	0.00130	0.170	0.0900	2.89e-11	-10.539	Through-diffusion	Distilled water	
101	0.000950	0.120	0.130	1.46e-11	-10.836	Through-diffusion	Distilled water	

	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
68	0.1M CsCl		20.0		aerobic
69	0.1M CsCl		25.0		aerobic
70	0.01M(1330ppm)CsCl	30days	25.0		aerobic
71	0.01M(1330ppm)CsCl	30days	25.0		aerobic
72	0.01M(1330ppm)CsCl	30days	25.0		aerobic
73	0.1M CsCl		20.0		aerobic
74	CsCl(+Cs-134)		probably room		aerobic
75	Cs-134(+Cs carrier)	51 days	70.0		aerobic
76	2kBq/ml	7days	30.0		aerobic
77	2kBq/ml	7days	30.0		aerobic
78	2kBq/ml	7days	30.0		aerobic
79	2kBq/ml	7days	30.0		aerobic
80	2kBq/ml	7days	40.0		aerobic
81	2kBq/ml	7days	56.0		aerobic
82	6000Bq/ml	23days	23.0		aerobic
83	6000Bq/ml	23days	23.0		aerobic
84	6000Bq/ml	23days	23.0		aerobic
85	6000Bq/ml	23days	23.0		aerobic
86	6000Bq/ml	23days	23.0		aerobic
87	6000Bq/ml	23days	23.0		aerobic
88	6000Bq/ml	23days	23.0		aerobic
89	6000Bq/ml	23days	23.0		aerobic
90	6000Bq/ml	23days	23.0		aerobic
91	6000Bq/ml	23days	23.0		aerobic
92	6000Bq/ml	23days	23.0		aerobic
93	2MBq/5ml		23.0		aerobic
94	2MBq/5ml		23.0		aerobic
95	2MBq/5ml		23.0		aerobic
96	6000Bq/ml	23days	23.0		aerobic
97	6000Bq/ml	23days	23.0		aerobic
98	1M(12700ppm)KI		probably room		aerobic
99	1M(12700ppm)KI		probably room		aerobic
100	1M(12700ppm)KI		probably room		aerobic
101	1M(12700ppm)KI		probably room		aerobic

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
102	I-125	7	I-	-1.0	Granite		0.400	4.40e-13	-12.357	2.00e-09
103	K	8	K+	1.0	Granite	2.60	0.880	7.70e-13	-12.114	2.00e-09
104	K	8	K+	1.0	Granite	2.60	0.880	1.10e-12	-11.959	2.00e-09
105	K	8	K+	1.0	Granite	2.60	0.880	8.30e-13	-12.081	2.00e-09
106	K	8	K+	1.0	Granite	2.60	0.880	1.80e-12	-11.745	2.00e-09
107	K	8	K+	1.0	Granite	2.60	0.880	8.30e-13	-12.081	2.00e-09
108	K	8	K+	1.0	Granite	2.60	0.880	1.80e-12	-11.745	2.00e-09
109	Mg	9	Mg2+	2.0	Granite		0.680	1.90e-13	-12.721	7.10e-10
110	Mg	9	Mg2+	2.0	Granite		0.680	3.30e-13	-12.481	7.10e-10
111	Mg	9	Mg2+	2.0	Granite		0.680	3.00e-13	-12.523	7.10e-10
112	Mg	9	Mg2+	2.0	Granite		0.680	2.90e-13	-12.538	7.10e-10
113	Mg	9	Mg2+	2.0	Granite		0.680	2.30e-13	-12.638	7.10e-10
114	Mg	9	Mg2+	2.0	Granite		0.680	3.60e-13	-12.444	7.10e-10
115	Mg	9	Mg2+	2.0	Granite		0.680	2.40e-13	-12.620	7.10e-10
116	Mg	9	Mg2+	2.0	Granite		0.680	1.90e-13	-12.721	7.10e-10
117	Na	10	Na+	1.0	Granodiorite	2.64	1.20	4.40e-13	-12.357	1.30e-09
118	Na	10	Na+	1.0	Granodiorite	2.64	1.20	5.20e-13	-12.284	1.30e-09
119	Na	10	Na+	1.0	Granodiorite	2.64	1.20	4.70e-13	-12.328	1.30e-09
120	Na	10	Na+	1.0	Granodiorite	2.73	2.30	6.50e-12	-11.187	1.30e-09
121	Na	10	Na+	1.0	Granodiorite	2.73	2.30	2.30e-12	-11.638	1.30e-09
122	Na	10	Na+	1.0	Granodiorite	2.73	2.30	5.20e-12	-11.284	1.30e-09
123	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	1.20e-11	-10.921	1.30e-09
124	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	5.10e-12	-11.292	1.30e-09
125	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	4.30e-12	-11.367	1.30e-09
126	Na	10	Na+	1.0	Fracture fillings	2.43	5.60	2.10e-11	-10.678	1.30e-09
127	Na	10	Na+	1.0	Fracture fillings	2.43	5.60	1.70e-11	-10.770	1.30e-09
128	Na	10	Na+	1.0	Fracture fillings	2.43	5.60			1.30e-09
129	Na	10	Na+	1.0	Granodiorite		2.30	4.70e-12	-11.328	1.30e-09
130	Na	10	Na+	1.0	Altered granodiorite		3.20	7.20e-12	-11.143	1.30e-09
131	Na	10	Na+	1.0	Fracture fillings		5.60	1.90e-11	-10.721	1.30e-09
132	Na	10	Na+	1.0	Granodiorite	2.73	2.30	4.70e-12	-11.328	1.30e-09
133	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	7.20e-12	-11.143	1.30e-09
134	Na	10	Na+	1.0	Fracture fillings	2.43	5.60	1.90e-11	-10.721	1.30e-09
135	Na	10	Na+	1.0	Granodiorite	2.73	2.30	4.70e-12	-11.328	1.30e-09

	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH
102	0.000220	0.0550	0.340	1.30e-12	-11.886	Through-diffusion	groundwater	
103	0.000390	0.0440				conductivity	0.1M KCl	
104	0.000550	0.0630				conductivity	0.1M KCl	
105	0.000410	0.0470				conductivity	0.1M KCl	
106	0.000900	0.100				conductivity	0.1M KCl	
107	0.000420	0.0470				conductivity	0.1M KCl	
108	0.000900	0.100				conductivity	0.1M KCl	
109	0.000270	0.0390				Through-diffusion	0.1M KCl	4.00
110	0.000460	0.0680				Through-diffusion	0.1M KCl	4.00
111	0.000420	0.0620				Through-diffusion	0.1M KCl	4.00
112	0.000410	0.0600				Through-diffusion	0.1M KCl	4.00
113	0.000320	0.0480				Through-diffusion	0.1M KCl	4.00
114	0.000510	0.0750				Through-diffusion	0.1M KCl	4.00
115	0.000340	0.0500				Through-diffusion	0.1M KCl	4.00
116	0.000270	0.0390				Through-diffusion	0.1M KCl	4.00
117	0.000340	0.0280				Through-diffusion	Distilled water	5.50
118	0.000400	0.0330				Through-diffusion	Distilled water	5.50
119	0.000360	0.0300				Through-diffusion	Distilled water	5.50
120	0.00500	0.220	0.0850	8.00e-11	-10.097	Through-diffusion	groundwater	8.70
121	0.00180	0.0770				Through-diffusion	groundwater	8.70
122	0.00400	0.170	0.960	5.40e-12	-11.268	Through-diffusion	groundwater	8.70
123	0.00920	0.290	1.43	8.40e-12	-11.076	Through-diffusion	groundwater	8.77
124	0.00390	0.120	0.660	7.70e-12	-11.114	Through-diffusion	groundwater	8.77
125	0.00330	0.100	0.330	1.30e-11	-10.886	Through-diffusion	groundwater	8.77
126	0.0160	0.290	1.11	1.90e-11	-10.721	Through-diffusion	groundwater	8.92
127	0.0130	0.230	0.850	2.00e-11	-10.699	Through-diffusion	groundwater	8.92
128						Through-diffusion	groundwater	8.92
129	0.00360	0.160		4.30e-11	-10.367	Through-diffusion	groundwater	8.70
130	0.00550	0.170		9.70e-12	-11.013	Through-diffusion	groundwater	8.77
131	0.0150	0.260		2.00e-11	-10.699	Through-diffusion	groundwater	8.92
132	0.00360	0.160		4.30e-11	-10.367	Through-diffusion	groundwater	8.70
133	0.00550	0.170		9.70e-12	-11.013	Through-diffusion	groundwater	8.77
134	0.0150	0.260		2.00e-11	-10.699	Through-diffusion	groundwater	8.92
135	0.00360	0.160		4.30e-11	-10.367	Through-diffusion	groundwater	8.70

	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
102	Sr12(3.4e-4M)		probably room		aerobic
103	0.1M KCl		probably room		aerobic
104	0.1M KCl		probably room		aerobic
105	0.1M KCl		probably room		aerobic
106	0.1M KCl		probably room		aerobic
107	0.1M KCl		probably room		aerobic
108	0.1M KCl		probably room		aerobic
109	MgCl2		25.0		aerobic
110	MgCl2		25.0		aerobic
111	MgCl2		25.0		aerobic
112	MgCl2		25.0		aerobic
113	MgCl2		25.0		aerobic
114	MgCl2		25.0		aerobic
115	MgCl2		25.0		aerobic
116	MgCl2		25.0		aerobic
117	10%NaCl		25.0		aerobic
118	10%NaCl		25.0		aerobic
119	10%NaCl		25.0		aerobic
120	0.05M(1150ppmNa)	30days	25.0		aerobic
121	0.05M(1150ppmNa)	30days	25.0		aerobic
122	0.05M(1150ppmNa)	30days	25.0		aerobic
123	0.05M(1150ppmNa)	30days	25.0		aerobic
124	0.05M(1150ppmNa)	30days	25.0		aerobic
125	0.05M(1150ppmNa)	30days	25.0		aerobic
126	0.05M(1150ppmNa)	30days	25.0		aerobic
127	0.05M(1150ppmNa)	30days	25.0		aerobic
128	0.05M(1150ppmNa)	30days	25.0		aerobic
129			25.0		aerobic
130			25.0		aerobic
131			25.0		aerobic
132	0.05M(1150ppmNa)	30days	25.0		aerobic
133	0.05M(1150ppmNa)	30days	25.0		aerobic
134	0.05M(1150ppmNa)	30days	25.0		aerobic
135	0.05M(1150ppmNa)	30days	25.0		aerobic

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
136	Na	10	Na+	1.0	Altered granodiorite	2.61	3.20	7.20e-12	-11.143	1.30e-09
137	Na	10	Na+	1.0	Fracture fillings	2.43	5.60	1.90e-11	-10.721	1.30e-09
138	Ni	11	Ni2+	2.0	Granodiorite	2.64	1.20	5.70e-13	-12.244	6.60e-10
139	Ni	11	Ni2+	2.0	Granite		0.680	2.00e-13	-12.699	6.60e-10
140	Ni	11	Ni2+	2.0	Granite		0.680	3.40e-13	-12.469	6.60e-10
141	Ni	11	Ni2+	2.0	Granite		0.680	3.00e-13	-12.523	6.60e-10
142	Ni	11	Ni2+	2.0	Granite		0.680	2.90e-13	-12.538	6.60e-10
143	Ni	11	Ni2+	2.0	Granite		0.680	2.40e-13	-12.620	6.60e-10
144	Ni	11	Ni2+	2.0	Granite		0.680	3.60e-13	-12.444	6.60e-10
145	Ni	11	Ni2+	2.0	Granite		0.680	2.50e-13	-12.602	6.60e-10
146	Ni	11	Ni2+	2.0	Granite		0.680	1.90e-13	-12.721	6.60e-10
147	Ni	11	Ni2+	2.0	Granodiorite		0.880	5.70e-13	-12.244	6.60e-10
148	Ni	11	Ni2+	2.0	Granodiorite		1.00	5.72e-13	-12.243	6.60e-10
149	Ni	11	Ni2+	2.0	Granodiorite			5.70e-13	-12.244	6.60e-10
150	Ni	11	Ni2+	2.0	Granodiorite		0.880	5.70e-13	-12.244	6.60e-10
151	Np-237	12	NpO2+	1.0	Granite		0.740	2.10e-13	-12.678	1.20e-09
152	Np-237	12	NpO2+	1.0	Granite		0.740	2.90e-13	-12.538	1.20e-09
153	Np-237	12			Granite	2.64	0.490	4.60e-13	-12.337	
154	Np-237	12			Granite	2.64	0.490	4.60e-13	-12.337	
155	Np-237	12			Granite	2.64	0.490	4.60e-13	-12.337	
156	Np-237	12	NpO2+	1.0	Granite			2.10e-13	-12.678	1.20e-09
157	Np-237	12	NpO2+	1.0	Granite			2.90e-13	-12.538	1.20e-09
158	Pu-239	13	Carbonate complex	-2.0	Granite			5.10e-14	-13.292	
159	Pu-239	13	Carbonate complex	-2.0	Granite			5.10e-14	-13.292	
160	Pu-239	13	Carbonate complex	-2.0	Granite			5.10e-14	-13.292	
161	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	3.20e-12	-11.495	8.10e-10
162	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	7.20e-13	-12.143	8.10e-10
163	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	1.90e-12	-11.721	8.10e-10
164	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20			8.10e-10
165	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20	2.40e-12	-11.620	8.10e-10
166	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20	3.30e-12	-11.481	8.10e-10
167	Se	14	SeO32-	-2.0	Fracture fillings	2.43	5.60	5.70e-12	-11.244	8.10e-10
168	Se	14	SeO32-	-2.0	Fracture fillings	2.43	5.60	4.90e-12	-11.310	8.10e-10
169	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	1.90e-12	-11.721	8.10e-10

	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m2/s)	log Da	Method	Porewater	pH
136	0.00550	0.170		9.70e-12	-11.013	Through-diffusion	groundwater	8.77
137	0.0150	0.260		2.00e-11	-10.699	Through-diffusion	groundwater	8.92
138	0.000860	0.0720	0.0570	1.00e-11	-11.000	Through-diffusion	Distilled water	5.80
139	0.000300	0.0450				Through-diffusion	0.1M KCl	4.00
140	0.000520	0.0760				Through-diffusion	0.1M KCl	4.00
141	0.000450	0.0670				Through-diffusion	0.1M KCl	4.00
142	0.000440	0.0650				Through-diffusion	0.1M KCl	4.00
143	0.000360	0.0530				Through-diffusion	0.1M KCl	4.00
144	0.000550	0.0800				Through-diffusion	0.1M KCl	4.00
145	0.000380	0.0560				Through-diffusion	0.1M KCl	4.00
146	0.000290	0.0420				Through-diffusion	0.1M KCl	4.00
147	0.000860	0.0980		1.00e-11	-11.000	Through-diffusion	Disrilled water	6.00
148	0.000870	0.0870		1.00e-11	-11.000	Through-diffusion	Disrilled water	6.00
149			0.0570	1.00e-11	-11.000	Through-diffusion	Disrilled water	5.80
150	0.000860	0.0980		1.00e-11	-11.000	Through-diffusion	Disrilled water	5.80
151	0.00180	0.240	0.0160	1.30e-11	-10.886	Through-diffusion	groundwater	7.40
152	0.00240	0.330	0.00400	7.20e-11	-10.143	Through-diffusion	groundwater	7.40
153			0.240	1.92e-12	-11.717	Through-diffusion	1M NaHCO3	8.60
154			0.240	1.92e-12	-11.717	Through-diffusion	1M NaHCO3	8.60
155			0.240	1.92e-12	-11.717	Through-diffusion	1M NaHCO3	8.60
156	0.00180		0.0160	1.30e-11	-10.886	Through-diffusion	groundwater	7.40
157	0.00240		0.00400	7.20e-11	-10.143	Through-diffusion	groundwater	7.40
158						Through-diffusion	NaHCO3+NaNO2	9.30
159						Through-diffusion	NaHCO3+NaNO2	9.30
160						Through-diffusion	NaHCO3+NaNO2	9.30
161	0.00400	0.170	0.320	1.00e-10	-10.000	Through-diffusion	groundwater	8.70
162	0.000880	0.0390	0.170	4.30e-12	-11.367	Through-diffusion	groundwater	8.70
163	0.00230	0.100	0.360	5.30e-12	-11.276	Through-diffusion	groundwater	8.70
164				1.40e-12	-11.854	Through-diffusion	groundwater	8.77
165	0.00300	0.0930	0.180	1.30e-11	-10.886	Through-diffusion	groundwater	8.77
166	0.00410	0.130	0.350	9.30e-12	-11.032	Through-diffusion	groundwater	8.77
167	0.00700	0.130	0.700	8.10e-12	-11.092	Through-diffusion	groundwater	8.92
168	0.00600	0.110	1.31	3.70e-12	-11.432	Through-diffusion	groundwater	8.92
169	0.00230	0.100		6.50e-12	-11.187	Through-diffusion	groundwater	8.70

	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
136	0.05M(1150ppmNa)	30days	25.0		aerobic
137	0.05M(1150ppmNa)	30days	25.0		aerobic
138	0.1MNiCl <sub>2</sub>	137days	25.0		aerobic
139	NiCl <sub>2</sub>		25.0		aerobic
140	NiCl <sub>2</sub>		25.0		aerobic
141	NiCl <sub>2</sub>		25.0		aerobic
142	NiCl <sub>2</sub>		25.0		aerobic
143	NiCl <sub>2</sub>		25.0		aerobic
144	NiCl <sub>2</sub>		25.0		aerobic
145	NiCl <sub>2</sub>		25.0		aerobic
146	NiCl <sub>2</sub>		25.0		aerobic
147	0.1MNiCl <sub>2</sub>		20.0		aerobic
148	0.1MNiCl <sub>2</sub>		20.0		aerobic
149	0.1MNiCl <sub>2</sub>		25.0		aerobic
150	0.1MNiCl <sub>2</sub>		20.0		aerobic
151	1117Bq/ml	about 180days	probably room		aerobic
152	1117Bq/ml	about 180days	probably room		aerobic
153	356Bq/ml	about 350days	26±1	-500~-400	reducing
154	356Bq/ml	about 350days	26±1	-500~-400	reducing
155	356Bq/ml	about 350days	26±1	-500~-400	reducing
156			room		aerobic
157			room		aerobic
158	570Bq/ml(e-6M)		25.0		aerobic
159	570Bq/ml(e-6M)		25.0		aerobic
160	570Bq/ml(e-6M)		25.0		aerobic
161	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic
162	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic
163	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic
164	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic
165	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic
166	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic
167	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic
168	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic
169	6e-4M(50ppmSe)SeO <sub>2</sub>	36days	22.0		anaerobic



	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
170	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20	2.90e-12	-11.538	8.10e-10
171	Se	14	SeO32-	-2.0	Granodiorite		2.30	1.90e-12	-11.721	8.10e-10
172	Se	14	SeO32-	-2.0	Altered granodiorite		3.20	2.90e-12	-11.538	8.10e-10
173	Se	14	SeO32-	-2.0	Fracture fillings		5.60	5.30e-12	-11.276	8.10e-10
174	Se	14	SeO42-	-2.0	Granodiorite					1.00e-09
175	Se	14	SeO32-	-2.0	Granodiorite	2.73	2.30	1.90e-12	-11.721	8.10e-10
176	Se	14	SeO32-	-2.0	Altered granodiorite	2.61	3.20	2.90e-12	-11.538	8.10e-10
177	Sm	15	Sm3+	3.0	Granodiorite	2.64	1.20	5.70e-13	-12.244	6.10e-10
178	Sm	15	Sm3+	3.0	Granodiorite		0.880	5.70e-13	-12.244	6.10e-10
179	Sm	15	Sm3+	3.0	Granodiorite		1.00	5.74e-13	-12.241	6.10e-10
180	Sm	15	Sm3+	3.0	Granodiorite			5.70e-13	-12.244	6.10e-10
181	Sm	15	Sm3+	3.0	Granodiorite		0.880	5.70e-13	-12.244	6.10e-10
182	Sr	16	Sr2+	2.0	Granite		0.700	2.00e-13	-12.699	7.90e-10
183	Sr	16	Sr2+	2.0	Granite		0.700	2.10e-13	-12.678	7.90e-10
184	Sr	16	Sr2+	2.0	Granite		0.700	2.60e-13	-12.585	7.90e-10
185	Sr	16	Sr2+	2.0	Granite		0.700	2.60e-13	-12.585	7.90e-10
186	Sr	16	Sr2+	2.0	Granite		0.700	3.10e-13	-12.509	7.90e-10
187	Sr	16	Sr2+	2.0	Granite		0.700	3.60e-13	-12.444	7.90e-10
188	Sr	16	Sr2+	2.0	Granite		0.700	3.20e-12	-11.495	7.90e-10
189	Sr	16	Sr2+	2.0	Granite		0.700	9.50e-12	-11.022	7.90e-10
190	Sr	16	Sr2+	2.0	Granite		0.700	1.70e-11	-10.770	7.90e-10
191	Sr	16	Sr2+	2.0	Granite		0.680	2.10e-13	-12.678	7.90e-10
192	Sr	16	Sr2+	2.0	Granite		0.680	3.60e-13	-12.444	7.90e-10
193	Sr	16	Sr2+	2.0	Granite		0.680	3.10e-13	-12.509	7.90e-10
194	Sr	16	Sr2+	2.0	Granite		0.680	2.60e-13	-12.585	7.90e-10
195	Sr	16	Sr2+	2.0	Granite		0.680	2.60e-13	-12.585	7.90e-10
196	Sr	16	Sr2+	2.0	Granite		0.680	2.00e-13	-12.699	7.90e-10
197	Sr	16	Sr2+	2.0	Granite			2.70e-13	-12.569	7.90e-10
198	Sr	16	Sr2+	2.0	Granite			1.00e-11	-11.000	7.90e-10
199	Sr	16	Sr2+	2.0	Granite		0.700	2.70e-13	-12.569	7.90e-10
200	Sr	16	Sr2+	2.0	Granite		0.400	1.60e-12	-11.796	7.90e-10
201	Sr	16	Sr2+	2.0	Granite			1.00e-11	-11.000	7.90e-10
202	Sr	16	Sr2+	2.0	Granite			2.00e-11	-10.699	7.90e-10
203	Sr	16	Sr2+	2.0	Granite			1.80e-13	-12.745	7.90e-10

	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m2/s)	log Da	Method	Porewater	pH
170	0.00360	0.110		1.10e-11	-10.959	Through-diffusion	groundwater	8.77
171	0.00230	0.100		6.50e-12	-11.187	Through-diffusion	groundwater	8.70
172	0.00360	0.110		1.10e-11	-10.959	Through-diffusion	groundwater	8.77
173	0.00650	0.120		5.90e-12	-11.229	Through-diffusion	groundwater	8.92
174				4.30e-11	-10.367	Advection diffusion	Distilled	
175	0.00230	0.100		6.50e-12	-11.187	Through-diffusion	groundwater	8.70
176	0.00360	0.100		1.10e-11	-10.959	Through-diffusion	groundwater	8.77
177	0.000930	0.0780	0.0570	1.00e-11	-11.000	Through-diffusion	Distilled water	5.80
178	0.000930	0.110		1.00e-11	-11.000	Through-diffusion	Distilled water	6.00
179	0.000940	0.0940		1.00e-11	-11.000	Through-diffusion	Distilled water	6.00
180	0.000930		0.0570	1.00e-11	-11.000	Through-diffusion	Distilled water	5.80
181	0.000930	0.110		1.00e-11	-11.000	Through-diffusion	Distilled water	6.00
182	0.000250	0.0360	0.0190	1.00e-11	-11.000	Through-diffusion	0.1M KCl	4.0000
183	0.000270	0.0380	0.00200	1.00e-10	-10.000	Through-diffusion	0.1M KCl	4.0000
184	0.000330	0.0470	0.0220	1.18e-11	-10.928	Through-diffusion	0.1M KCl	4.0000
185	0.000330	0.0470	0.00300	8.67e-11	-10.062	Through-diffusion	0.1M KCl	4.0000
186	0.000390	0.0580	0.0190	1.63e-11	-10.788	Through-diffusion	0.1M KCl	4.0000
187	0.000460	0.0650	0.0140	2.57e-11	-10.590	Through-diffusion	0.1M KCl	4.0000
188	0.00410	0.580	2.40	1.33e-12	-11.876	Through-diffusion	Deionized water	5.5000
189	0.0120	1.70	1.50	6.33e-12	-11.199	Through-diffusion	Deionized water	5.5000
190	0.0220	3.10	1.70	1.00e-11	-11.000	Through-diffusion	Deionized water	5.5000
191	0.000270	0.0390				Through-diffusion	0.1M KCl	4.0000
192	0.000460	0.0670				Through-diffusion	0.1M KCl	4.0000
193	0.000390	0.0580				Through-diffusion	0.1M KCl	4.0000
194	0.000330	0.0480				Through-diffusion	0.1M KCl	4.0000
195	0.000330	0.0480				Through-diffusion	0.1M KCl	4.0000
196	0.000250	0.0370				Through-diffusion	0.1M KCl	4.0000
197	0.000340					Through-diffusion	0.1M KCl	
198	0.0130					Through-diffusion	Deionized water	
199	0.000340	0.0490	0.0130	2.08e-11	-10.682	Through-diffusion	0.1M KCl	4.0000
200	0.00200	0.510	1.20	1.30e-11	-10.886	Through-diffusion	groundwater	
201	0.0130		2.00	5.00e-12	-11.301	Through-diffusion	0.1M KCl	4.0000
202	0.0260		3.00	6.67e-12	-11.176	Through-diffusion	0.1M KCl	4.0000
203	0.000160		0.0380	4.74e-12	-11.324	Through-diffusion	Deionized water	4.0000

	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
170	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic
171			Temp. in G.B.		anaerobic
172			Temp. in G.B.		anaerobic
173			Temp. in G.B.		anaerobic
174	SeO2(1000ppmSe)		probably room		aerobic
175	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic
176	6e-4M(50ppmSe)SeO2	36days	22.0		anaerobic
177	0.1M SmCl3	137days	25.0		aerobic
178	0.1M SmCl3		20.0		aerobic
179	0.1M SmCl3		20.0		aerobic
180	0.1M SmCl3		25.0		aerobic
181	0.1M SmCl3		20.0		aerobic
182	Sr(NO3)2		25.0		aerobic
183	Sr(NO3)2		25.0		aerobic
184	Sr(NO3)2		25.0		aerobic
185	Sr(NO3)2		25.0		aerobic
186	Sr(NO3)2		25.0		aerobic
187	Sr(NO3)2		25.0		aerobic
188	Sr(NO3)2		25.0		aerobic
189	Sr(NO3)2		25.0		aerobic
190	Sr(NO3)2		25.0		aerobic
191	Sr(NO3)2		25.0		aerobic
192	Sr(NO3)2		25.0		aerobic
193	Sr(NO3)2		25.0		aerobic
194	Sr(NO3)2		25.0		aerobic
195	Sr(NO3)2		25.0		aerobic
196	Sr(NO3)2		25.0		aerobic
197	1M Sr(NO3)2		probably room		aerobic
198	1M Sr(NO3)2		probably room		aerobic
199			25.0		aerobic
200	SrI2(3.4e-3M)	300days	probably room		aerobic
201			probably room		aerobic
202			probably room		aerobic
203					aerobic

	Element(nuclide)	Element No.	Species	Charge	Solid	Dry density(g/cm3)	Porosity(%)	De(m2/s)	log De	Do(m2/s)
204	Sr	16	Sr2+	2.0	Granite			3.20e-13	-12.495	7.90e-10
205	Sr	16	Sr2+	2.0	Granite		0.800			
206	Sr	16	Sr2+	2.0	Granite		0.800			
207	Sr	16	Sr2+	2.0	Granite		0.800			
208	Sr	16	Sr2+	2.0	Granite		0.800			
209	Tc-99	17	TcO4-	-1.0	Granite		7.70	4.20e-14	-13.377	2.00e-09
210	Tc-99	17	TcO4-	-1.0	Granite		1.00	4.20e-14	-13.377	2.00e-09
211	U-233	18	UO22+	2.0	Granite		0.700	2.20e-14	-13.658	4.30e-10
212	U-233	18	UO22+	2.0	Granite		0.700	4.20e-14	-13.377	4.30e-10
213	U-233	18	UO22+	2.0	Granite		0.700	4.40e-14	-13.357	4.30e-10
214	U-233	18	UO22+	2.0	Biotitic granite		0.700	2.20e-14	-13.658	4.30e-10
215	U-233	18	UO22+	2.0	Biotitic granite		0.700	4.20e-14	-13.377	4.30e-10
216	U-233	18	UO22+	2.0	Biotitic granite		0.700	4.40e-14	-13.357	4.30e-10
217	U-233	18	Carbonate complex	-4.0	Granite			1.40e-13	-12.854	7.20e-10
218	U-233	18	Carbonate complex	-4.0	Granite			1.40e-13	-12.854	7.20e-10
219	U-233	18	Carbonate complex	-4.0	Granite			1.40e-13	-12.854	7.20e-10
220	U-233	18	UO22+	2.0	Granite		0.800			
221	U-233	18	UO22+	2.0	Granite		0.800			
222	U-233	18	UO22+	2.0	Granite		0.800			
223	U-233	18	UO22+	2.0	Granite		0.800			
224	Uranine	19			Granite	2.6000	0.740	4.70e-14	-13.328	4.50e-10
225	Uranine	19			Granite	2.6000	0.740	3.50e-14	-13.456	4.50e-10
226	Uranine	19			Granite	2.6000	0.740	5.00e-14	-13.301	4.50e-10
227	Uranine	19			Granite	2.6000	0.700	4.70e-14	-13.328	4.50e-10
228	Uranine	19			Granite	2.6000	0.700	3.50e-14	-13.456	4.50e-10
229	Uranine	19			Granite	2.6000	0.700	5.00e-14	-13.301	4.50e-10

	Formation factor(FF)	Geometric factor(Gf)	Rock capacity factor	Da(m <sup>2</sup> /s)	log Da	Method	Porewater	pH
204	0.000410					Through-diffusion	Deionized water	4.0000
205				1.00e-11	-11.000	In-diffusion	Deionized water	4.3000
206				4.00e-12	-11.398	In-diffusion	Deionized water	6.0000
207				1.00e-11	-11.000	In-diffusion	Deionized water	7.7000
208				1.00e-11	-11.000	In-diffusion	Deionized water	9.4000
209	2.10e-05	0.00210				Through-diffusion	Distilled water	8.55~8.60
210	2.10e-05	0.00210		4.20e-12	-11.377	Through-diffusion	Distilled water	
211	5.10e-05	0.00730	0.0180	1.22e-12	-11.914	Through-diffusion	0.1M KCl	4.00
212	9.80e-05	0.0140	0.0180	2.33e-12	-11.633	Through-diffusion	0.1M KCl	4.00
213	1.00e-04	0.0150	0.0180	2.44e-12	-11.613	Through-diffusion	0.1M KCl	4.00
214	5.10e-05	0.00730	0.0110	2.00e-12	-11.699	Through-diffusion	0.1M KCl	4.00
215	9.80e-05	0.0140	0.0250	1.68e-12	-11.775	Through-diffusion	0.1M KCl	4.00
216	1.00e-04	0.0150	0.0120	3.67e-12	-11.435	Through-diffusion	0.1M KCl	4.00
217	0.000190					Through-diffusion	NaHCO <sub>3</sub> +NaNO <sub>2</sub>	9.30
218	0.000190					Through-diffusion	NaHCO <sub>3</sub> +NaNO <sub>2</sub>	9.30
219	0.000190					Through-diffusion	NaHCO <sub>3</sub> +NaNO <sub>2</sub>	9.30
220				6.00e-13	-12.222	In-diffusion	Deionized water	5.00
221				2.00e-12	-11.699	In-diffusion	Deionized water	5.00
222				2.00e-12	-11.699	In-diffusion	Deionized water	5.00
223				7.00e-12	-11.155	In-diffusion	Deionized water	5.00
224	1.00e-04	0.0140	0.0150	3.10e-12	-11.509	Through-diffusion	Distilled water	
225	7.80e-05	0.0110	0.0170	2.10e-12	-11.678	Through-diffusion	Distilled water	
226	0.000110	0.0150	0.0710	7.00e-13	-12.155	Through-diffusion	Distilled water	
227	1.00e-04	0.0150	0.0150	3.10e-12	-11.509	Through-diffusion		
228	7.80e-05	0.0110	0.0170	2.10e-12	-11.678	Through-diffusion		
229	0.000110	0.0160	0.0710	7.00e-13	-12.155	Through-diffusion		

	Tracer	Contact time	Temperature(°C)	Eh(mV)	Others
204					aerobic
205	2kBq/ml Sr	7days	30.0		aerobic
206	2kBq/ml Sr	7days	30.0		aerobic
207	2kBq/ml Sr	7days	30.0		aerobic
208	2kBq/ml Sr	7days	30.0		aerobic
209	14kBq/ml	20days	23.0		aerobic
210	14kBq/ml	20days	room		aerobic
211	640Bq/ml U-233		room		aerobic
212	640Bq/ml U-233		room		aerobic
213	640Bq/ml U-233		room		aerobic
214	7.7e-6M	about 350days	20-25(room)		aerobic
215	7.7e-6M	about 350days	20-25(room)		aerobic
216	7.7e-6M	about 350days	20-25(room)		aerobic
217	560Bq/ml		25.0		aerobic
218	560Bq/ml		25.0		aerobic
219	560Bq/ml		25.0		aerobic
220	2e-5 M(U:uranyl)	54days	30.0		aerobic
221	2e-5 M(U:uranyl)	70days	30.0		aerobic
222	2e-5 M(U:uranyl)	58days	30.0		aerobic
223	2e-5 M(U:uranyl)	252days	30.0		aerobic
224	10000ppm uranine	150days	probably room		aerobic
225	10000ppm uranine	150days	probably room		aerobic
226	10000ppm uranine	150days	probably room		aerobic
227	1%uranine solution		room		aerobic
228	1%uranine solution		room		aerobic
229	1%uranine solution		room		aerobic