Diffusion Behaviour of Se in Compacted Sodium Bentonite under Reducing Conditions

April 1997

Tokai Works
Power Reactor and Nuclear Fuel Development Corporation

公　開　資　料
PNC TN8410 97-075
1 9 9 7 年 4 月

Diffusion Behaviour of Se in Compacted Sodium Bentonite under Reducing Conditions

要　旨

高レベル放射性廃棄物地層処分の性能評価において，深部地質環境における酸化還元条件は，還元性と考えられており，酸化還元条件に鋭敏な元素の1つであるSeは，条件によって価数が変化することが知られている。しかしながら，ベントナイト中のSeの拡散に関して還元条件下で研究された例は未だ見られず，実験的証拠が不足している。本報告では，ベントナイト中のSeの見掛けの拡散係数をベントナイト密度をパラメーターに還元条件下にて取得すると共に，還元環境下でのベントナイト中のSeの拡散挙動について検討した結果を記述する。Na型ベントナイトのクニゲルV1中のSeの見掛けの拡散係数を濃度プロファイル法によりベントナイト密度800～1800kg·m^{-3}の範囲において還元条件(Eh vs. SHE -373～-363mV)及び室温(23.6～23.7℃)にて取得した。実験は，N_2雰囲気のグローブボックス内(O_2＜1ppm)で行い，間隙水の還元条件は，酸化還元電位をモニタリングしながら還元溶液とベントナイトを焼結フィルターを介して接触させることにより維持した。また，間隙水の酸化還元電位を確認するため，圧密ベントナイトを介しての酸化還元電位の伝播性を還元剤$Na_2S_2O_4$を用いてベントナイト密度1800kg·m^{-3}の試料について透過拡散法により実験的に調べた。得られた見掛けの拡散係数は6.1x10^{-11}～4.3x10^{-10}m^{2}·s^{-1}の範囲であり，ベントナイト密度の増加に伴って緩やかに減少する傾向が見られた。還元条件におけるベントナイト間隙水中でのSeの支配化学種は，HSe^-であると考えられ，ベントナイト中のHSe^-の見掛けの拡散係数は，同じ電荷を取るTcO_4^-のそれとほぼ同じであった。しかしながら，大気条件でのベントナイト中のアクチニド元素の見掛けの拡散係数は極めて小さく，間隙水中で複雑な陰イオンの錯体を形成することが知られている。ベントナイト中でのこれらの元素の拡散挙動は，陰イオンが支配的とは言うもののTcO_4^-やHSe^-とは異なるものと思われる。このことから，間隙水中で単純なイオンを形成し，同様な電荷を持つイオンの拡散挙動は類似しているものと考えられる。

東海事業所　環境技術開発部　地層処分開発室

CONTENTS

FIGURES

TABLES

Diffusion Behaviour of Se in Compacted Sodium Bentonite under Reducing Conditions

Haruo Sato*

ABSTRACT

In the performance assessment of geological disposal of high-level radioactive waste in Japan, redox condition in deep geological environment is considered to be reducing, and Se is one of the important redox sensitive elements. However, no studies on diffusion of Se in bentonite under reducing conditions have been reported yet. This paper describes the results of apparent diffusion coefficients of Se in compacted sodium bentonite obtained as a function of bentonite density under reducing conditions and discusses its diffusion behaviour. Apparent diffusion coefficients of Se in compacted sodium bentonite, Kunigel V1 (constituent montmorillonite 46 ~ 49wt%), were obtained in a range of dry densities of bentonite, 800 ~ 1800 $kg \cdot m^{-3}$ under reducing conditions (Eh vs. SHE -373~-363mV) at room temperature (23.6~23.7°C) by in-diffusion method. All the experiments were carried out in an N_2-atmospheric glove box (O_2 < 1ppm) and the reducing conditions of the porewater were maintained by continuous contact between compacted bentonite and reducing solution including 5.7×10^{-4} M-$Na_2S_2O_4$ through a sintered metal filter. The Eh of reducing solution was continuously monitored. Furthermore, a through-diffusion experiment of $Na_2S_2O_4$ was also carried out at a dry density, 1800 $kg \cdot m^{-3}$ in order to check the reducing condition of the porewater. The Eh in the measurement cell was confirmed to be the same as that in the tracer cell. The apparent diffusion coefficients of Se were in the range, 6.1×10^{-11} ~ 4.3×10^{-10} $m^2 \cdot s^{-1}$ and showed a tendency of slight decrease with increasing dry density of bentonite. The dominant species of Se in the porewater under reducing conditions is predicted to be HSe^-, and the apparent diffusion coefficients of HSe^- in the bentonite were approximately the same as those of TcO_4^- taking the same ionic charge. However, those for actinides in bentonite are known to be quite low under oxidizing conditions, and they form dominant anionic complexes in the porewater. Diffusion behaviour of actinides in bentonite seems to be different from those of TcO_4^- and HSe^-. Therefore, diffusion behaviour of ions, forming a simple anion with the same charge in bentonite is shown to be very similar.

---

* Geological Isolation Technology Section, Waste Technology Development Division, Tokai Works, Power Reactor and Nuclear Fuel Development Corporation, 4-33 Muramatsu, Tokai-mura, Ibaraki-ken, 319-11 Japan

1. INTRODUCTION

Diffusion coefficients of key nuclides in compacted bentonite are listed up as one of the important parameters required in the performance assessmsnt of geological disposal of high-level radioactive waste in Japan. Many studies on diffusion of nuclides in compacted bentonite have been reported, focused on Na-typed bentonite to date (For example, Sato et al., 1993). However, almost all the studies have been carried out under aerobic conditions, and redox condition in deep geological environment has not been taken into account. The redox condition in deep geological environment is considered to be reducing, and elements being sensitive to the redox condition are presumed to be different chemical behaviour from that under atmospheric conditions.

Selenium-79 is produced as a fission product in a power reactor and is one of the important radionuclides for performance assessment because of its long half-life of 6.5×10^4 yr. Selenium is a redox sensitive element, and it is well known that the valence changes depending on redox condition. Moreover, for chemical species of Se, it is known that Se forms anion when exisits as an ion in solution, and that it is weak sorptive on bentonite has been clarified experimentally (Shibutani et al., 1994).

We have obtained apparent diffusion coefficients of Se in comacted bentonite in a range of densities of 400 ~ 1800 $kg \cdot m^{-3}$ using crude sodium bentonite, Kunigel V1®, under anaerobic condition (O_2: 2.5ppm)(Sato et al., 1994a, 1994b, 1995). However, these studies were all conducted under atmosphere controlled system, but not under reducing condition.

Selenium can chemically take Se(-II), (0), (IV) and (VI) as valence state, and SeO_3^{2-} species is predicted to be predominant under anaerobic conditions (Ticknor at al., 1988). However, it is well known that HSe^- species is predominant under reducing conditions (Brookins, 1988; Ticknor, 1988). As described above, some studies on diffusion of Se under anaerobic conditions have been reported, but no studies carried out under reducing conditions which is considered to be deep geological environment have been reported yet.

This paper describes the results of apparent diffusion coefficients of Se in compacted sodium bentonite obtained as a function of bentonite density under reducing conditions and discusses its diffusion behaviour.

2. EXPERIMENTALS

2.1 Diffusion experiment

The experiments were carried out by in-diffusion method (Torstenfelt and Allard, 1986). The experimental condition is shown in **Table 1**. Bentonite, Kunigel V1, was dried at 110°C in an oven for over night and was packed into acrylic diffusion columns to get densities of 800, 1400 and 1800 $kg \cdot m^{-3}$, respectively. **Figure 1** shows a schematic view of the diffusion column. Each column has a hole with 20mm in diameter and 20mm thick, in which hole bentonite sample is emplaced. In the packing of the bentonite, the samples of 1400 and 1800 $kg \cdot m^{-3}$ were compacted using a hydraulic press and a punching tool (Sato et al., 1992).

The columns with bentonite were placed in the evacuation chamber of an atmosphere controlled glove box and were evacuated oxygen gas sorbed on the bentonite and it existed in the bentonite pore by exchanging with N_2 gas. They were then put in the glove box purged with N_2 gas. The evacuation was repeated 3 times. Next, doubly distilled water degassed by bubbling with atmospheric gas of the glove box for over night was prepared. Moreover, porewater lowered redox potential by adding a small amount of $Na_2S_2O_4$ (Sodium Hydrosulfite, Junsei Chemical C., Ltd.) to the degassed water was prepared ($5.7x10^{-4}M$-$Na_2S_2O_4$). The columns with bentonite were then immersed in this porewater to be saturated. **Figure 2** shows a concept of the immersion of the columns. The immersion was conducted for 4 weeks. The redox potential of the porewater was also monitored during the immersion and was constantly maintained by adding reductant ($Na_2S_2O_4$) as appropriate. After the bentonite was saturated with the porewater, a tracer solution was prepared by diluting a 1000ppm-Se Standard Solution (Waco Pure Chemical Industries, Ltd.) with aliquot of the porewater used in the immersion of bentonite. The pH of the tracer solution lowered by dissolving of the Se Standard Solution, but was adjusted by NaOH to become pH 7.0 which corresponds to the porewater pH. This solution was then placed for a week for aging of precipitation and was filtered with a 0.2μm pore size filter in order to separate the precipitation from the solution. The concentration of Se in the tracer solution was determined to be 100ppm (corresponding to 1.3×10^{-3}M) from analysis with an ICP emission spectroscopy (detection limit: 0.03ppm, corresponding to 3.8×10^{-7}M). A small amount of this tracer solution (50μl) was pipetted on the surface of bentonite specimen in each column, and a blind lid (bottom of the column) was then shut with bolts as shown in **Figure 3**. On the other hand, the top lid of the column with hole remained as it was in order to come in contact with porewater adjusted redox potential. Then porewater level was adjusted not to reach the position of tracer pipetted. The experiments were run for 3, 6 and 10 days for densities of 800, 1400 and 1800 $kg \cdot m^{-3}$, respectively.

After certain time period, the cylindrical core of bentonite was pushed out with an extruding tool (digital position indicator) and cut into 2mm thick slices with a cutter knife as shown in **Figure 4**. Each slice was put in sample bottles and put those out from glove box. Selenium was extracted from the bentonite slices in a 1M-HNO_3 solution with a liquid/solid ratio of 0.02 $m^3 \cdot kg^{-1}$ for 3 days. The extracted solutions were then filtered with a 0.2μm pore size filter. The concentrations of Se in the filtered solutions were analyzed with an ICP emmision spectroscopy.

2.2 Through-diffusion test of reductant through compacted bentonite

In the diffusion experiments under reducing conditions, the control of redox potential of the porewater in bentonite during the immersion and diffusion experiment was maintained by continuous contact between reducing solution and compacted bentonite through a sintered metal filter. However, whether the bentonite porewater was under reducing condition or not has not been checked. In this mesurement, the conductivity of redox potential (permeativity of reductant) was experimentally investigated through compacted bentonite in order to know indirectly redox potential of the porewater.

The experiment was carried out by through-diffusion method (Kita et al., 1989; Park et al., 1991) in experimental condition shown in **Table 2**. **Figure 5** shows a schematic view of acrylic diffusion cell. Bentonite, Kunigel V1, was dried at 110°C for over night and was packed into sample holder of the diffusion cell to get a density of 1800 $kg{\cdot}m^{-3}$. The size of sample is 20mm in diameter and 5mm thick. The diffusion cell with bentonite was also evacuated oxygen gas sorbed on the bentonite and it existed in the bentonite pore in the evacuation chamber in the same way as diffusion experiment of Se. They were then put in the glove box. Degassed doubly distilled water was also prepared in the same way as in-diffusion experiments. This degassed water was injected with a volume of 100ml into both a tracer and a measurement cell of diffusion cell shown in **Figure 5** in order to saturate bentonite. The saturation was conducted under vacuum conditions (several tens of torr) to accelerate for 2 weeks. After the saturation of bentonite, a small amount of reductant, an $Na_2S_2O_4$ powder was added into the tracer cell to become a concentration of 5.7×10^{-3}M, and the conductivity test of redox potential was started. During the experiment, Eh vs. SCE values of the solutions in both cells were measured as a function of time using an ORP electrode (Toa Electronics Ltd., saturated calomel electrode (SCE) PTS-5011C) by a pH meter (Toa Electronics Ltd., HM-30S). At the same time, temperature of the solutions was also measured with an accuracy of ±0.5°C in order to calculate Eh vs. SHE (Standard Hydrogen Electrode) values.

3. RESULTS AND DISCUSSION

3.1 Through-diffusion test of reductant through compacted bentonite

Figure 6 shows the changes in Eh vs. SCE of solutions in both cells of diffusion cell as a function of time, and **Figure 7** shows the changes in Eh vs. SHE as a function of time. Since electrode used for the measurement of redox potential in this experiment was a saturated calomel electrode (SCE), the measured values were converted to Eh vs. SHE values based on equation proposed by Ostwald; $Eh=ORP+0.2415-0.00079(T-25)$ (Tajima, 1986). Where Eh is the Eh vs. SHE (V), ORP is the Eh vs. SCE (V) and T is the temperature (°C).

As shown in **Figure 7**, the Eh vs. SHE of solution in the measurement cell began to lower after several tens of minutes and became approximately the same value as that of solution in the tracer cell after 3 days. The experiment was continued for 9 days, and a little rise in the Eh was found with increasing time. This probable reason is the decrease in reducing capacity of reductant, and it is presumed that reducing condition is able to be maintained by adding reductant as appropriate. Since the change in redox potential in measurement cell occurs through bentonite porewater, redox potential of the porewater is also considered to lower. From this experiment, redox potential adjusted out of compacted bentonite is considered to become equal to that of the porewater of compacted bentonite after about 3 days for a sample of 5mm in thickness.

3.2 Recovery of Se from bentonite

The recovery of Se from bentonite specimen was estimated based on the total amount of tracer (5.0×10^{-9} kg) introduced a diffusion experiment and the accumulated quantities of Se extracted from each bentonite slice. The recoveries were 76 ~ 100% and were quite acceptable. Therefore, it is judged that correction of recovery in concentration profile of Se in bentonite is not needed.

3.3 Change in temperature

Since diffusion experiments were carried out in an atmosphere controlled glove box which cannot control temperature, the temperature in the glove box was monitored during the experiments. **Figure 8** shows the change in temperature as a function of time in the glove box. As shown in **Figure 8**, the temperature in the glove box during the experiments was relatively stable at about 24°C throughout the saturation of bentonite and diffusion experiments.

3.4 Change in redox potential of porewater

Figure 9 shows the change in Eh vs. SCE of the porewater of bentonite as a function of time coming in contact during the saturation of bentonite and diffusion experiments. As **Figure 9** shows, the Eh vs. SCE was stable at around -600mV (corresponding to about -350mV vs. SHE) throughout all the experiments. Therefore, it is presumed that reducing condition was being maintained.

3.5 Apparent diffusion coefficient

Figures 10 ~ 12 show concentration profiles of Se in compacted bentonite in the direction of the depth from the surface of bentonite specimen (diffusion source), on which tracer solution was pipetted for each density and those as a function of square of distance from the diffusion source. As shown in **Figures**, remarkably high concentration was not found near the diffusion source. In actual, no precipitation of Se was found on the surface of bentonite specimen when tracer solution was pipetted. From this, apparent diffusion coefficient was calculated by the analytical solution in a thin layer source.

Diffusion equation for one-dimensional non-steady state is given by the following equation based on Fick's second law (Torstenfelt et al., 1985; Muurinen et al., 1985).

$$\frac{\partial C(t, X)}{\partial t} = Da\frac{\partial^2 C(t, X)}{\partial X^2} \qquad (1)$$

Where C(t, X) is the concentration of Se per unit volume of bentonite ($kg \cdot m^{-3}$), t is the time (s), X is the distance from the diffusion source (m) and Da is the apparent diffusion coefficient ($m^2 \cdot s^{-1}$).

For one-dimensional diffusion of a planar source consisting of a limited amount of substance in a cylinder of infinite length, the analytical solution of equation (1) is derived based on initial and boundary conditions as follows (Crank, 1975).

Initial condition

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

Boundary condition

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

$$M = \int_0^\infty C(t, X)dX$$

$$C(t, X) = \frac{M}{\sqrt{\pi Da\, t}} exp\left(-\frac{X^2}{4Da\, t}\right) \qquad (2)$$

Where M is the total amount of tracer (Se) per unit area of bentonite specimen ($kg \cdot m^{-2}$).

From equation (2), taking *log* C(t, X) and X^2 as the vertical and the horizontal axes, respectively, the slope , $-1/(4Da \cdot t)$ gives apparent diffusion coefficient from the relation with time. The apparent diffusion coefficients were obtained from the linear least-squares fit to the plot. **Table 3** shows the obtained apparent diffusion coefficients of Se in compacted bentonite, and **Figure 13** shows a dependence of apparent diffusion coefficient on dry density of bentonite. As shown in **Figure 13**, though it is not remarkable, the apparent diffusion coefficients of Se showed a tendency of decrease with increasing dry density of bentonite. The dominant species of Se in the porewater was predicted to be HSe^- by Eh-pH diagrams (Brookins, 1988; Ticknor et al., 1988) around pH 8 ~ 9 which is considered to correspond to pH (Sasaki et al., 1995) of the porewater. The apparent diffusion coefficients obtained in this study have a little higher than those of SeO_3^{2-} obtained under anaerobic conditions and were approximately the same values as those of TcO_4^- having an ionic charge of −1. Shibutani et al. (Shibutani et al., 1992, 1994) have carried out studies on sorption of SeO_3^{2-} on bentonite under anaerobic conditions and have reported that little or no sorption was found on bentonite in a wide pH range. Since it is shown that Se forms anion when exists as an ion in solution, Se is presumed to be weak sorptive on bentonite. It is well known that Tc also takes dominantly TcO_4^- in a wide pH range under atmospheric conditions (Brookins, 1988). Besides, since it is known that distribution coefficient of TcO_4^- on bentonite is generally low (Brandberg and Skagius, 1991), the fact supports that apparent diffusion coefficients of TcO_4^- were the highest values next to those of HTO which is a non-sorbing nuclide on bentonite. To the contrary, actinides such as Np, Am and Pu are considered to form dominantly anions in the porewater of bentonite under oxidizing conditions, but distribution coefficients of these nuclides on bentonite are generally high, and apparent diffusion coefficients of these nuclides in compacted bentonite are also quite low (Sato et al., 1992, 1993). However, chemical behaviour of actinides is complicated, and these nuclides form some anionic complexes in bentonite porewater as well as form a small rate of cations. Therefore, these nuclides would be retarded by the combination of sorption on bentonite, anion-exclusion and molecular filtration (McKinley and Hadermann, 1984) caused by a formation of large complexes (Sato et al., 1992, 1993). Thus diffusion behaviour of these nuclides in bentonite seems to be different from those of TcO_4^- and HSe^-. From this, ions forming a simple anion with the same charge in bentonite porewater are predicted to be similar apparent diffusion coefficients in the same conditions. The surface of bentonite particle is generally known to be negatively charged (Sato et al., 1992) around pH 8 ~ 9. Therefore, that apparent diffusion coefficients of HSe^- in compacted bentonite showed a little higher than those of SeO_3^{2-} would be due to the difference in the effect of anion-exclusion (McKinley and Hadermann, 1984) in compacted bentonite caused by the difference of ionic charge between both species.

4. CONCLUSIONS

(1) Apparent diffusion coefficients of Se in compacted sodium bentonite, Kunigel V1, were obtained in a range of dry densities of 800 ~ 1800 kg·m^{-3} under reducing conditions (Eh vs. SHE −373 ~ −363mV) at room temperature (23.6~23.7°C) by in-diffusion method. All the experiments were carried out in an N_2-atmospheric glove box (O_2 < 1ppm), and the reducing conditions of the porewater were maintained by continuous contact between compacted bentonite and reducing solution including 5.7x10^{-4}M-$Na_2S_2O_4$ through a sintered metal filter. The redox potential of the reducing solution was continuously monitored. Furthermore, a through-diffusion experiment of $Na_2S_2O_4$ was also carried out at a dry density of 1800 kg·m^{-3} in order to check the redox potential of the porewater. The redox potential in the measurement cell was confirmed to be the same as that

in the tracer cell. The apparent diffusion coefficients of Se were in the range, $6.1\times10^{-11} \sim 4.3\times10^{-10}\ m^2 \cdot s^{-1}$ and showed a tendency of slight decrease with increasing dry density of bentonite.

(2) The dominant species of Se in the porewater of bentonite under reducing conditions is predicted to be HSe^-, and the apparent diffusion coefficients of HSe^- in the bentonite were approximately the same as those of TcO_4^- taking the same ionic charge. However, those for actinides in bentonite are known to be quite low under oxidizing conditions, and they form dominant anionic complexes in the porewater. Diffusion behaviour for actinides in bentonite seems to be different from those of TcO_4^- and HSe^-. Therefore, diffusion behaviour of ions, forming a simple anion with the same charge in bentonite is shown to be very similar.

5. REFERENCES

Brandberg, F. and Skagius, K.(1991): Porosity, Sorption and Diffusivity Data Compiled for the SKB 91 Study, *SKB* 91-16.

Brookins, D. G.(1988): *Eh-pH Diagrams for Geochemistry*, Springer-Verlag Berlin Heidelbag.

Crank, J.(1975): *The Mathematics of Diffusion*, 2nd ed., Pergamon Press, Oxford.

Kita, H., Iwai, T., 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], 26-32, [in Japanese].

McKinley, I. G. and Hadermann, J.(1984): Radionuclide Sorption Database for Swiss Safety Assessment, NAGRA Technical Report, *NTB* 84-40.

Muurinen, A., Rantanen, J., Penttila-Hiltunen, P.(1985): Diffusion of Cesium and Strontium in Compacted and in Mixtures of Sodium Bentonite and Crushed Rock, *Scientific Basis for Nuclear Waste Management VIII*, Vol.44, 883-890.

Park, C. K., Han, K. W., 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, 156-160.

Sasaki, Y., Shibata, M., Yui, M., Ishikawa, H.(1995): Experimental Studies on the Interaction of Groudwater with Bentonite, *Materials Research Symposium Proceedings*, Vol.353, 337-344.

Sato, H., Ashida, T., Kohara, Y., Yui, M., Sasaki, N.(1992): Effect of Dry Density on Diffusion of Some Radionuclides in Compacted Sodium Bentonite, *Journal of Nuclear Science and Technology*, 29[9], 873-882.

Sato, H., Uchidate, N., Inui, S., Yoshikawa, H.(1994a): Measurement of Apparent Diffusion Coefficient for Se in Bentonite and Modelling by Electrical Double Layer Theory, *1994 Annual Meeting of the Atomic Energy Society of Japan*, B30, p82, [in Japanese].

Sato, H. and Shibutani, T.(1994b): Study on Adsorption and Diffusion Mechanism of Nuclides in Buffer Material and Geosphere, PNC Technical Review No.91, *PNC TN* 8410 94-284, [in Japanese].

Sato, H., Yui, M., Yoshikawa, H.(1995): Diffusion Behavior for Se and Zr in Sodium Bentonite, *Materials Research Society Symposium Proceedings*, Vol.353, 269-276.

Sato, H. and Yui, M.(1996): Diffusion Behavior of Nuclides in Compacted Bentonite -Dependence of Apparent Diffusivities for Ni^{2+} on Dry Density and Temperature-, *1996 Fall Meeting of the Atomic Energy Society of Japan*, L16, p679, [in Japanese].

Sato, H.(to be published, 1997): Effect of Dry Density and pH on Diffusion for Sm in Compacted Sodium Bentonite, PNC Technical Report.

Shibutani, T., Yoshikawa, H., Sato, H., Yui, M., Umeki, H., Ishiguro, K.(1992): Distribution Coefficients on Bentonite and Rocks, PNC Technical Report, *PNC TN* 8410 92-163, [in Japanese].

Shibutani, T., Yui, M., Yoshikawa, H.(1994): Sorption Mechanism of Pu, Am and Se on Sodium-Bentonite, *Materials Research Society Symposium Proceedings*, Vol.333, 725-730.

Tajima, S.(1986): *"Denki-kagaku-tsuron"(An Introduction to Electrochmistry, 3rd ed.)*, Kyoritsu Press, p102, [in Japanese].

Ticknor, K. V., Harris, D. R., Vandergraaf, T. T.(1988): Sorption/Desorption Studies of Selenium on Fracture-filling Minerals under Aerobic and Anaerobic Coditions, Atomic Energy of Canada Limited Research Company Technical Report, *TR*-453.

Torstenfelt, B., Kipatsi, H., Allard, B.(1985): Mesurements of Ion Mobilities in Clay, *Soil Sci.*, 139[6], 512-516.

Torstenfelt, B. and Allard, B.(1986): Migration of Fission Products and Actinides in Compacted Bentonite, *SKBF/KBS TR* 86-14.

Table 1 Experimental condition in diffusion

Bentonite	Kunigel V1(Kunimine Industries Co. Ltd.)
Dry density	800, 1400, 1800(kg m^{-3})
Method	In-diffusion method
Initial porewater	degassed doubly distilled water + reducing agent($Na_2S_2O_4$): 5.7×10^{-4} M
Atmosphere	under atmosphere controlled condition (N_2-atmosphere, $O_2 < 1$ppm)
Temperature	temperature in glove box (20～25℃)
Tracer	SeO_2 solution(1.3×10^{-3} M SeO_2)
Producibility	n = 2

Table 2 Experimental condition in conductivity test of redox potential in compacted bentonite

Bentonite	Kunigel V1(Kunimine Industries Co. Ltd.)
Dry density	1800(kg m^{-3})
Method	Through-diffusion method
Initial porewater	degassed doubly distilled water
Reductant	$Na_2S_2O_4$(5.7x10^{-3} M)
Atmosphere	under atmosphere controlled condition (N_2-atmophere, O_2< 1ppm)
Temperature	temperature in glove box(20～25℃)
Producibility	n = 1

Table 3 Obtained apparent diffusion coefficients

Dry density ($kg\ m^{-3}$)	Temperature (℃)	Eh vs. SHE(mV)	Da(m^2s^{-1})
800	23.6±0.1	-373	$4.3x10^{-10}$ $1.7x10^{-10}$
1400	23.6±0.1	-368±5.5	$2.2x10^{-10}$ $1.6x10^{-10}$
1800	23.7±0.3	-363±1.1	$2.3x10^{-10}$ $6.1x10^{-11}$

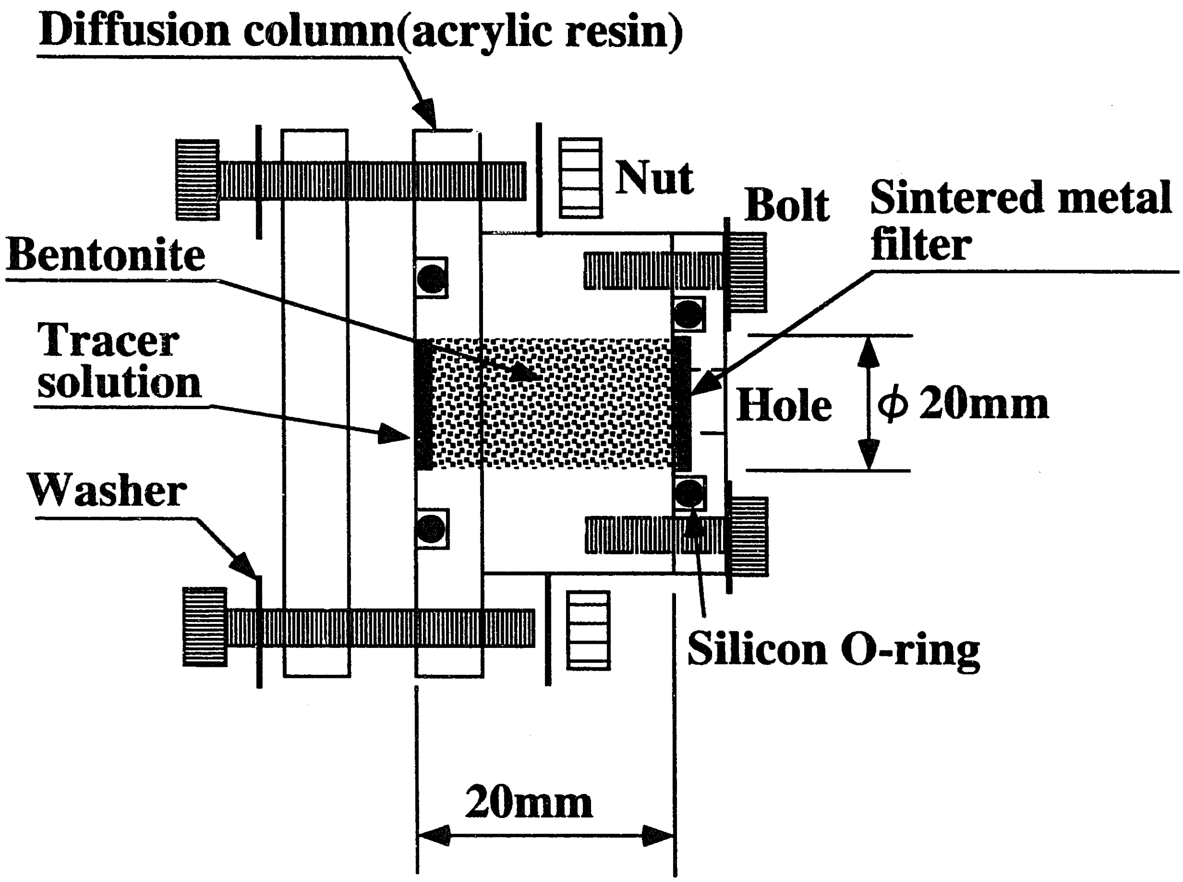

Figure 1 Schematic view of diffusion column

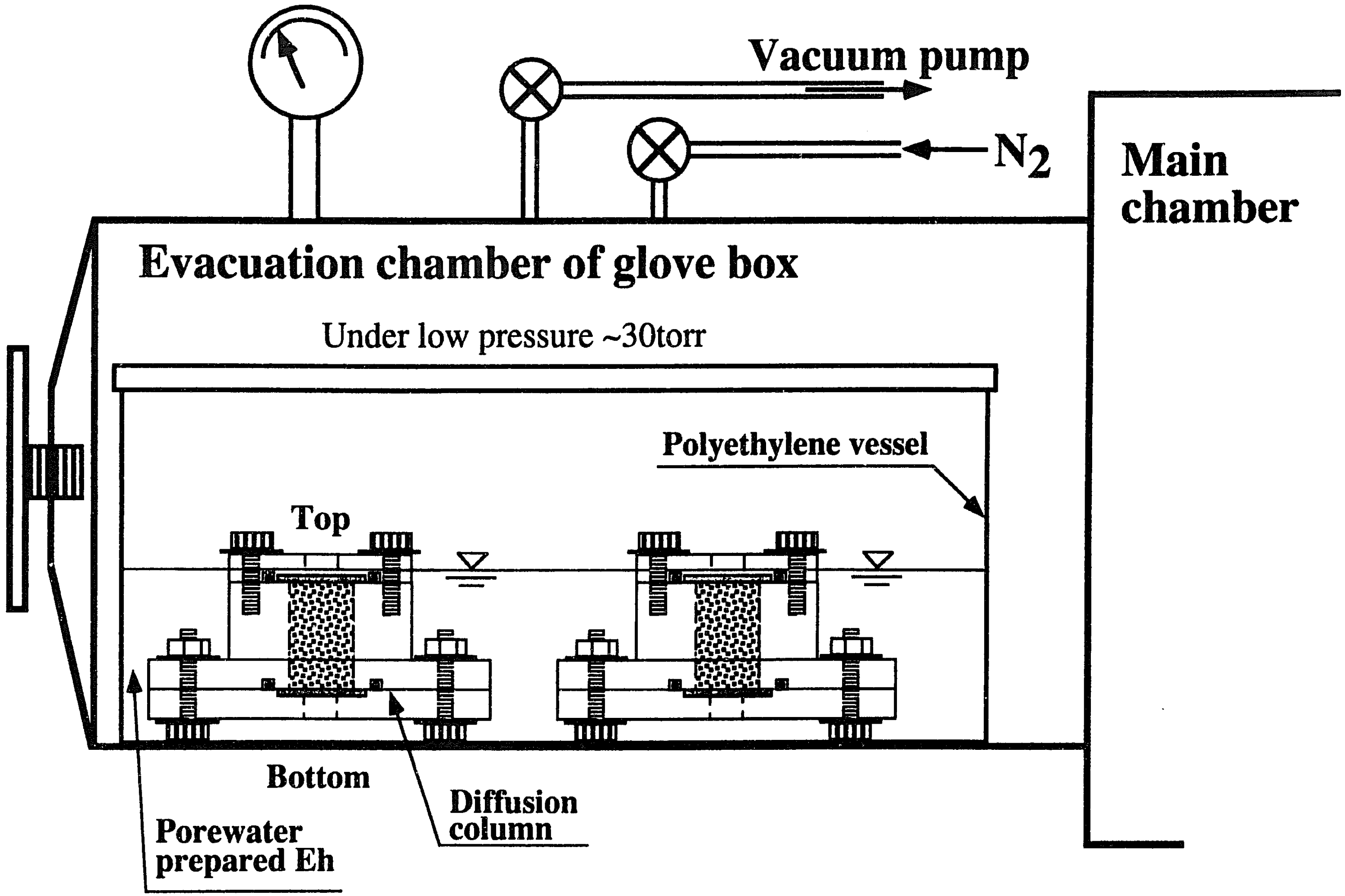

Figure 2 Concept of immersion of diffusion columns with bentonite

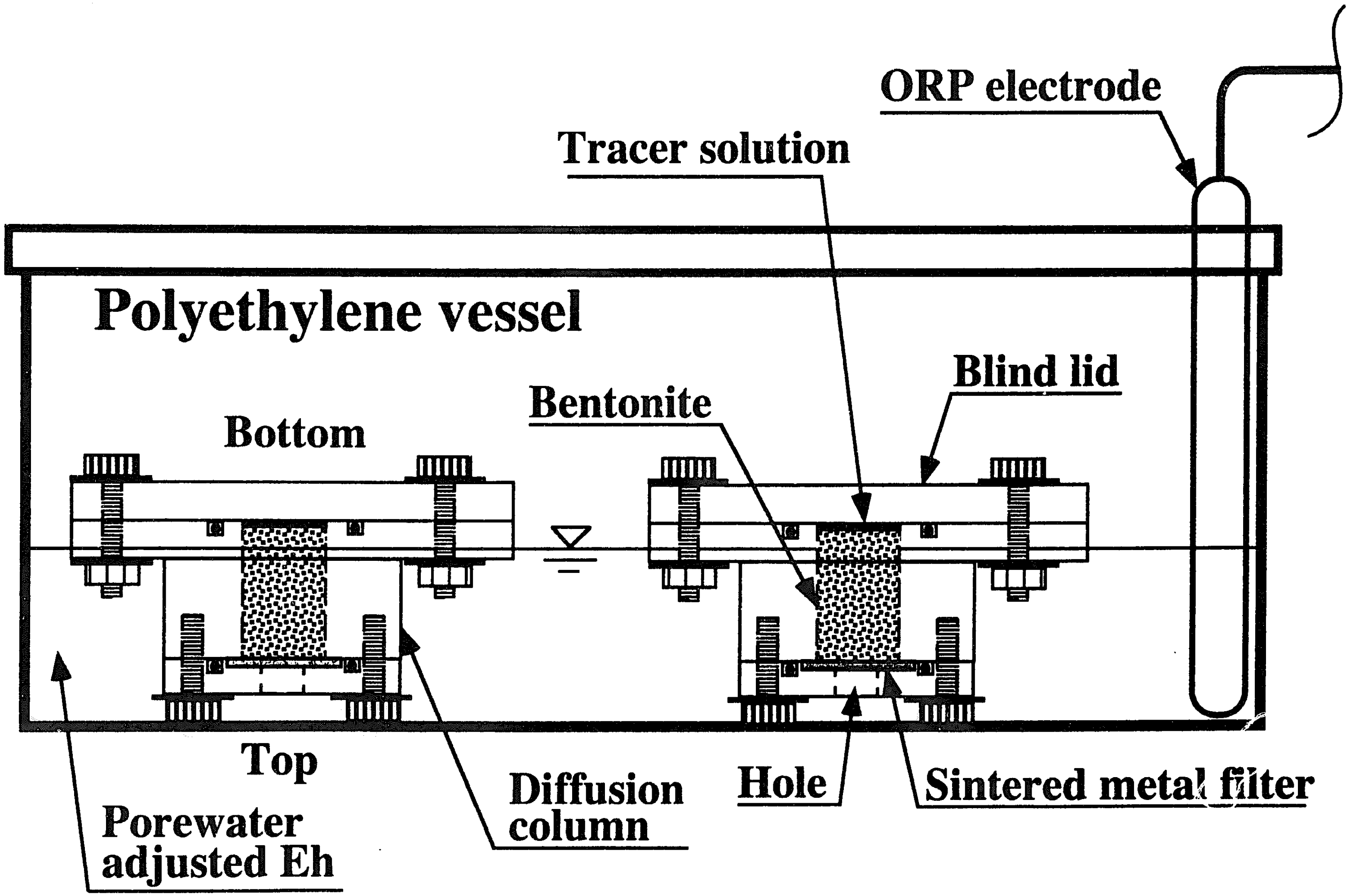

Figure 3 Concept of diffusion experiment

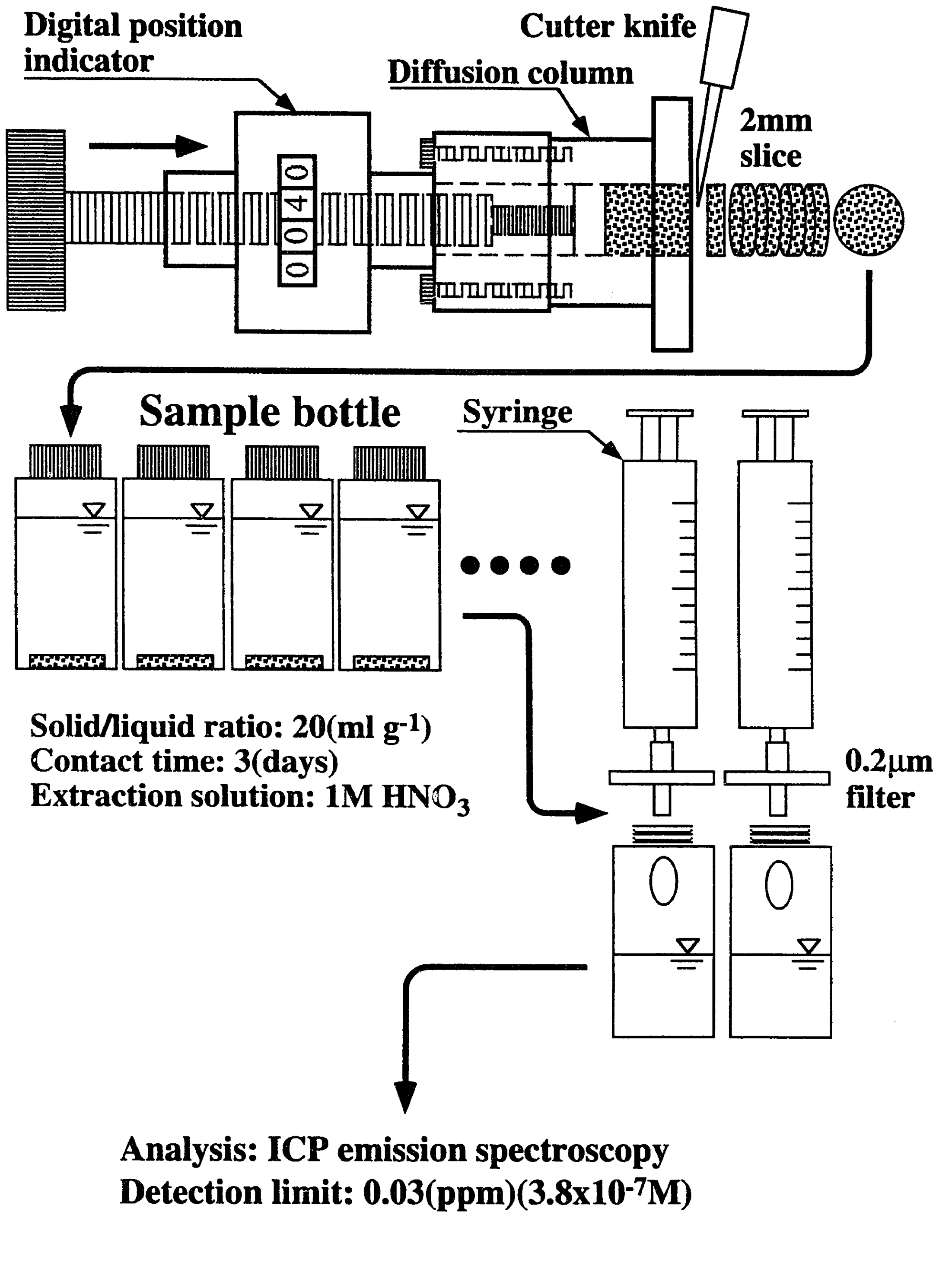

Figure 4 Slice of bentonite and extraction of tracer from the bentonite slice

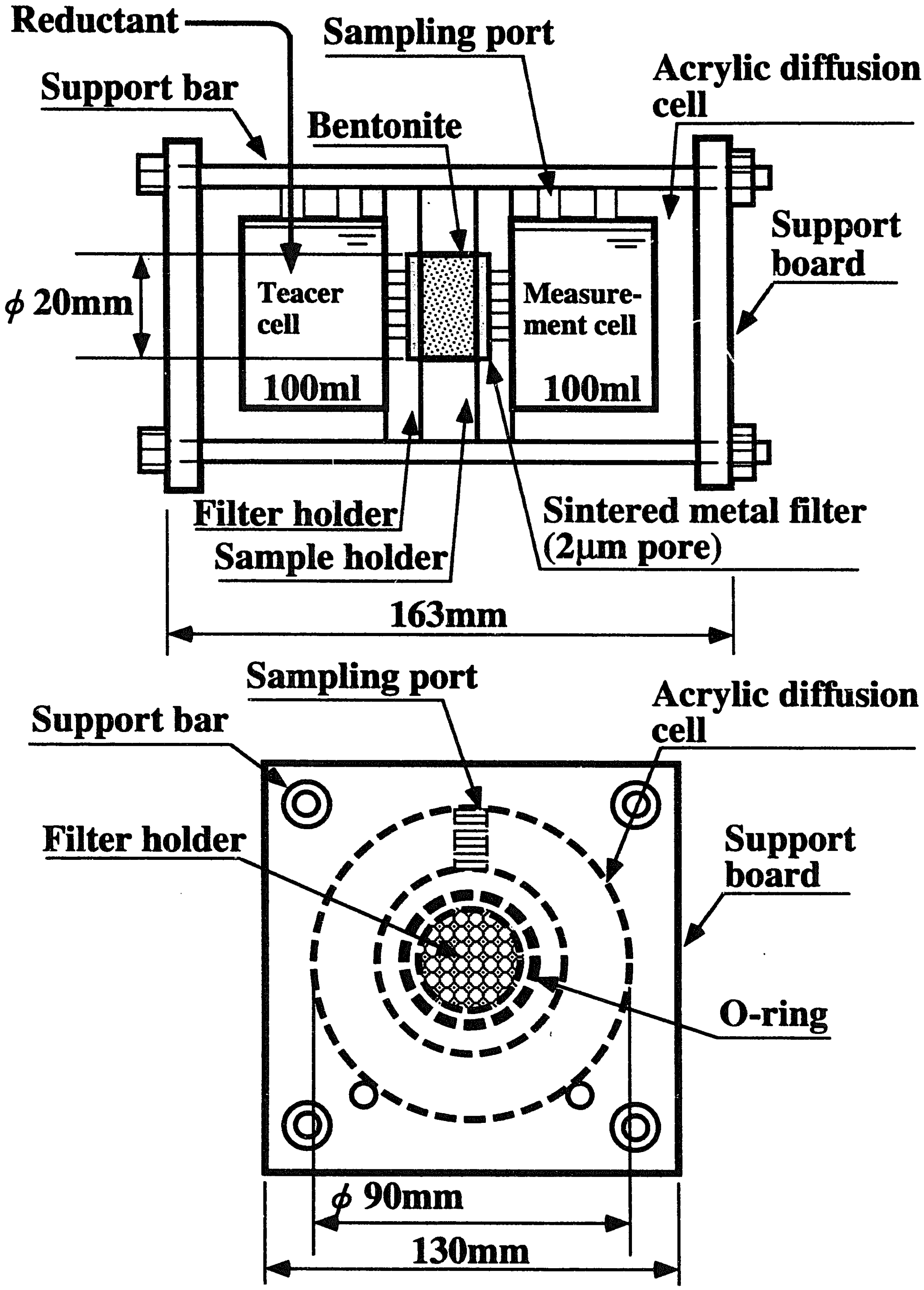

Figure 5 Schematic view of diffusion cell for bentonite experiment

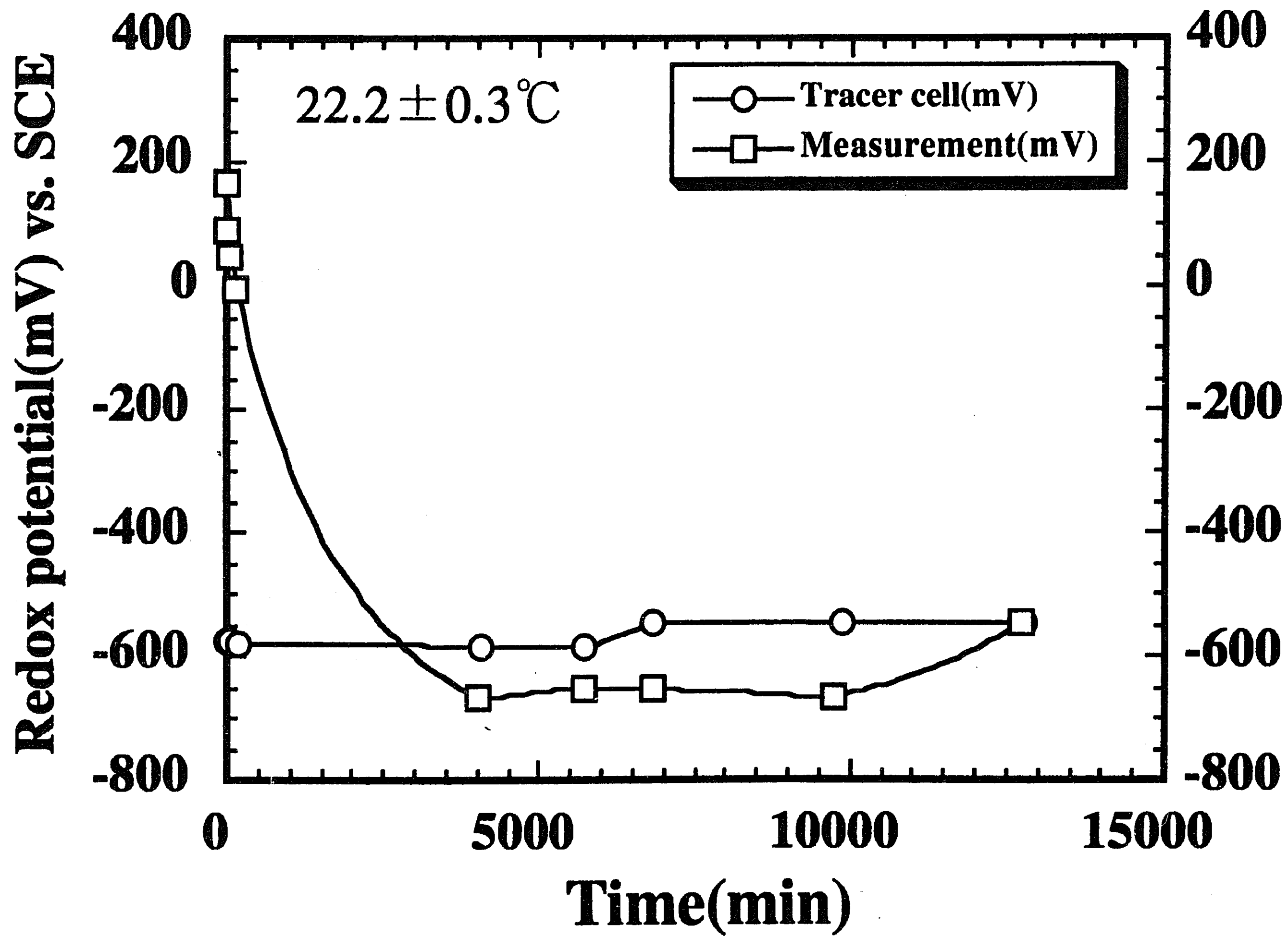

Figure 6 Changes in Eh vs. SCE of solutions in both cells of diffusion cell as a function of time

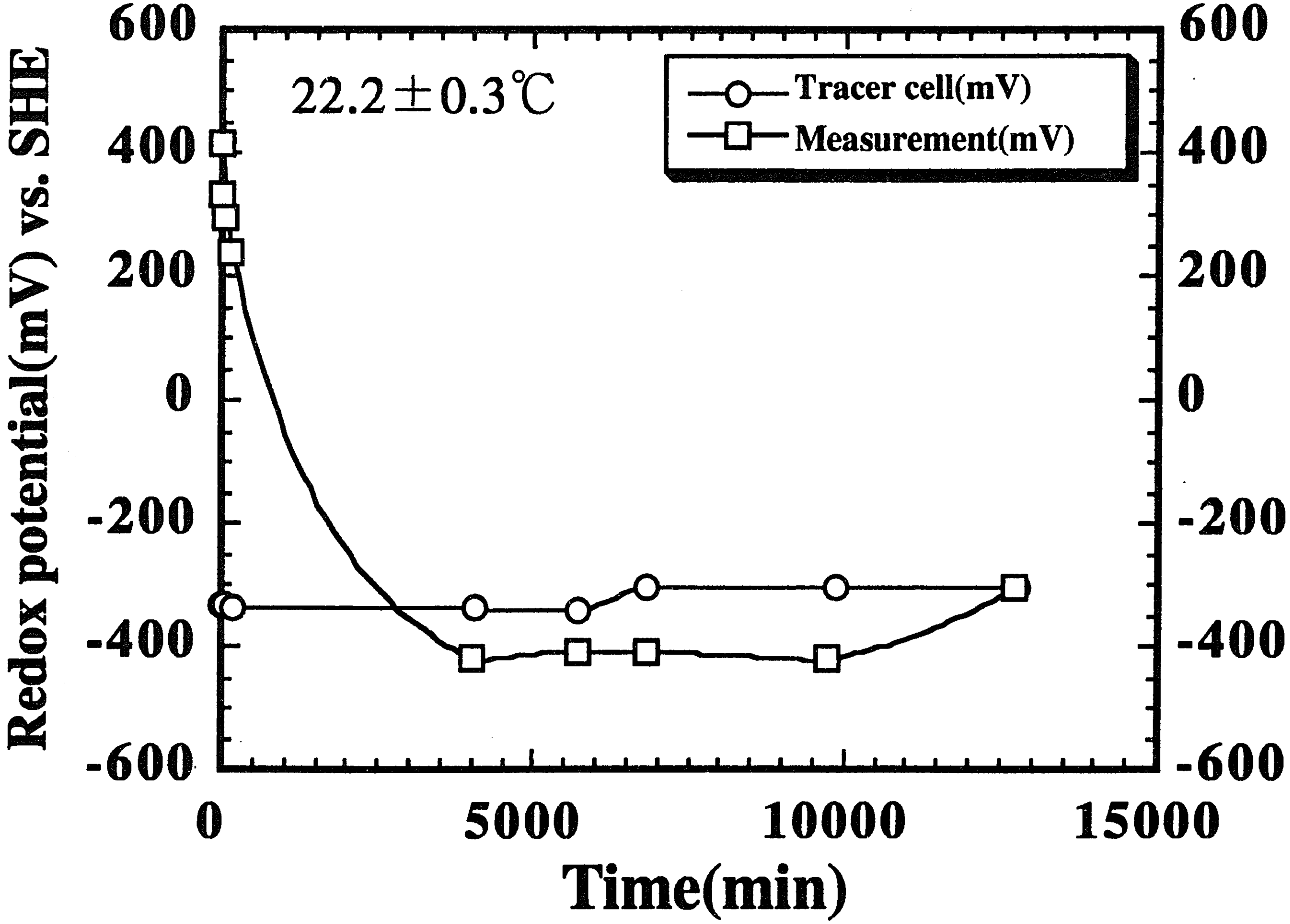

Figure 7 Changes in Eh vs. SHE of solutions in both cells of diffusion cell as a function of time

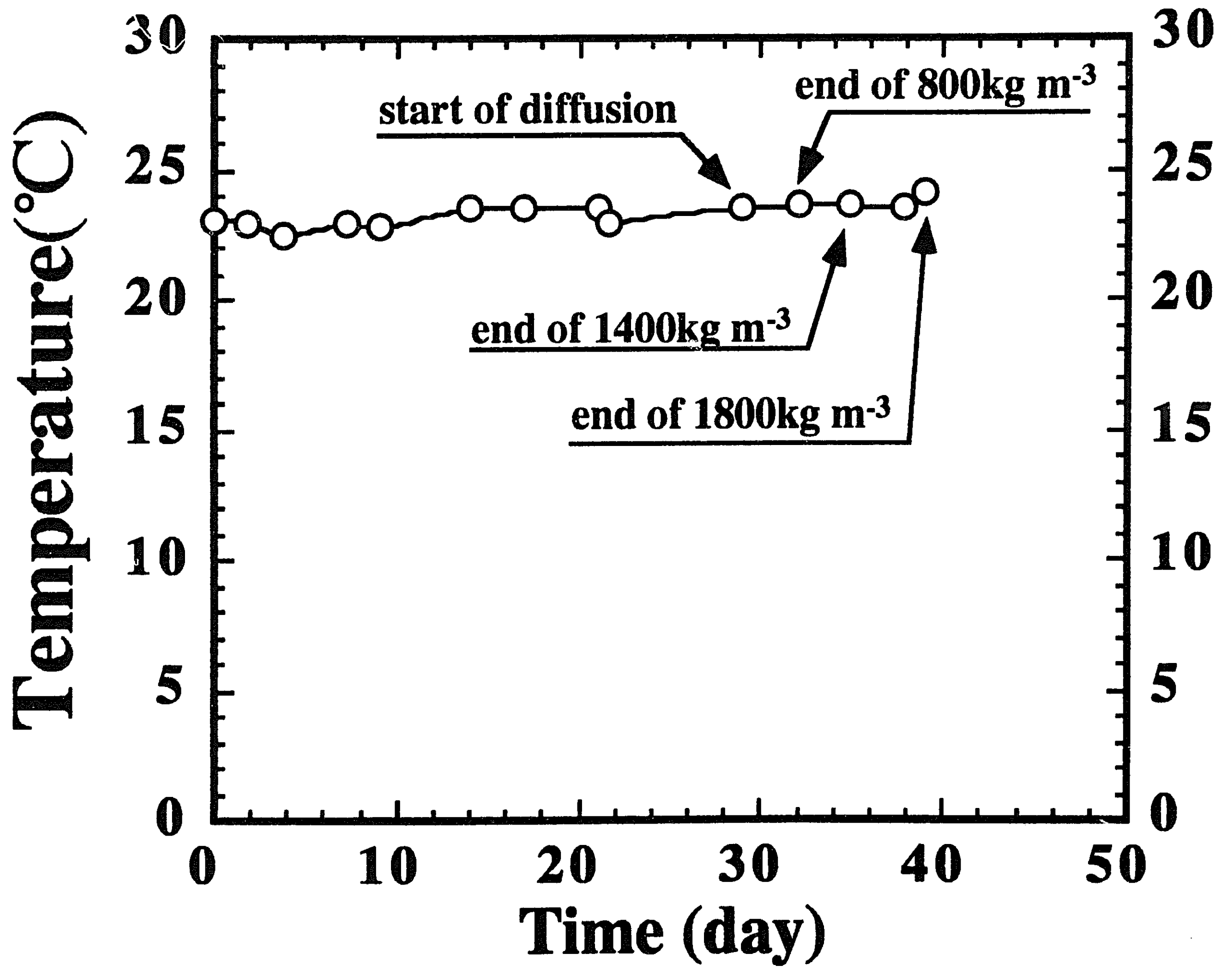

Figure 8 Change in temperature as a function of time in atmosphere controlled glove box

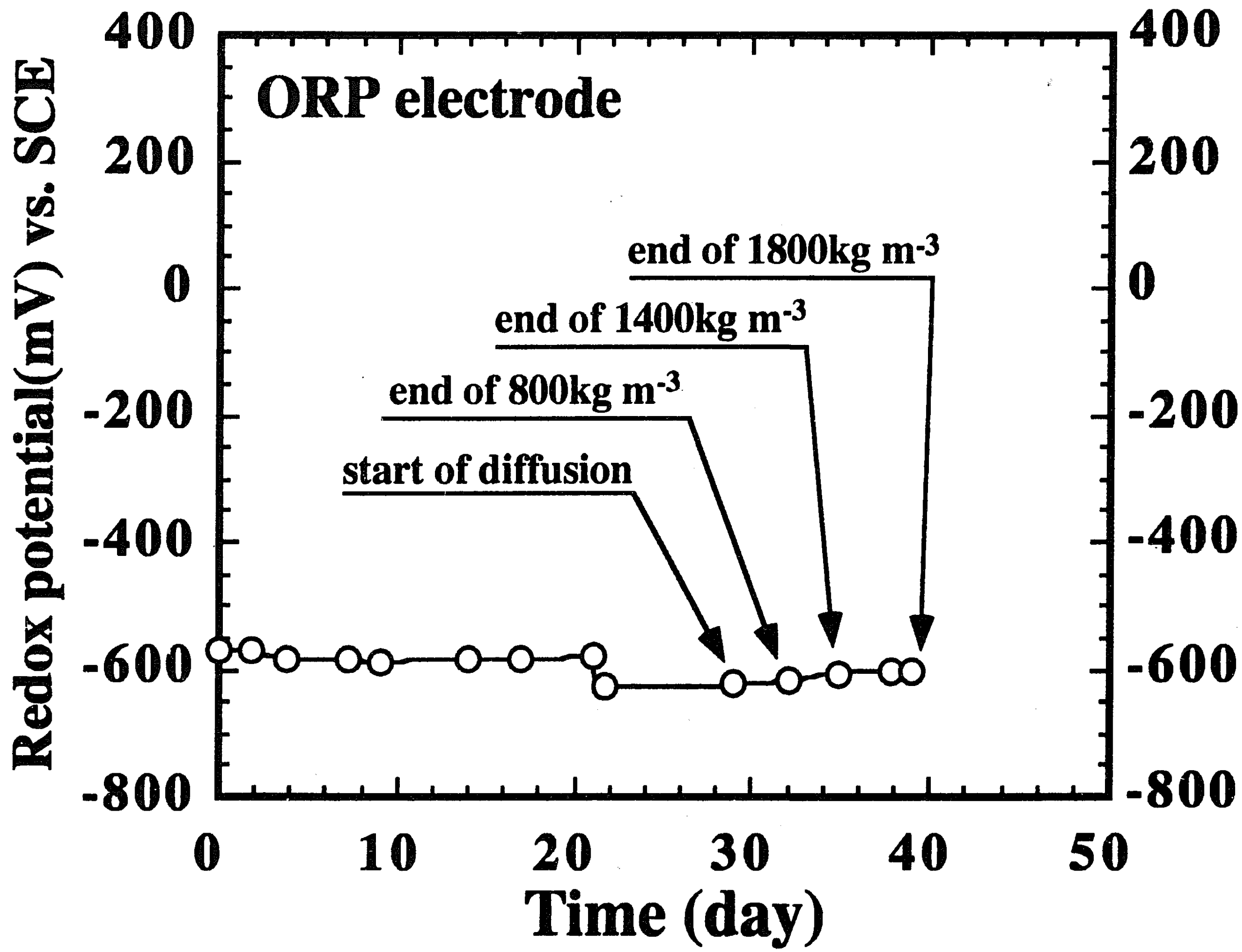

Figure 9 Change in Eh vs. SCE of porewater as a function of time coming in contact during the saturation of bentonite and diffusion experiment

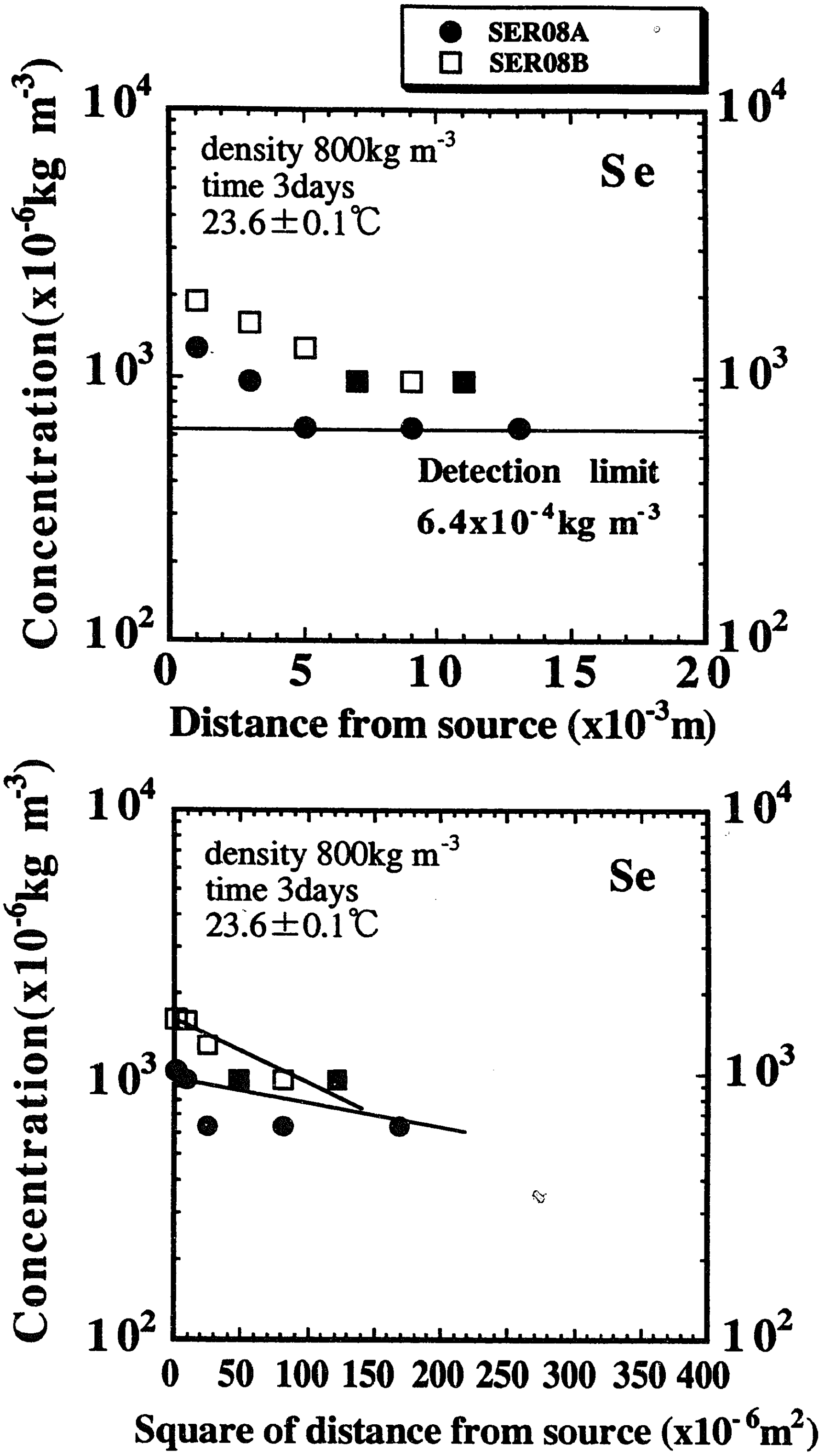

Figure 10 Concentration profiles of Se in compacted bentonite as functions of distance (upper Figure) and square of distance from diffusion source (lower Figure) for a density of 800 kg m^{-3}

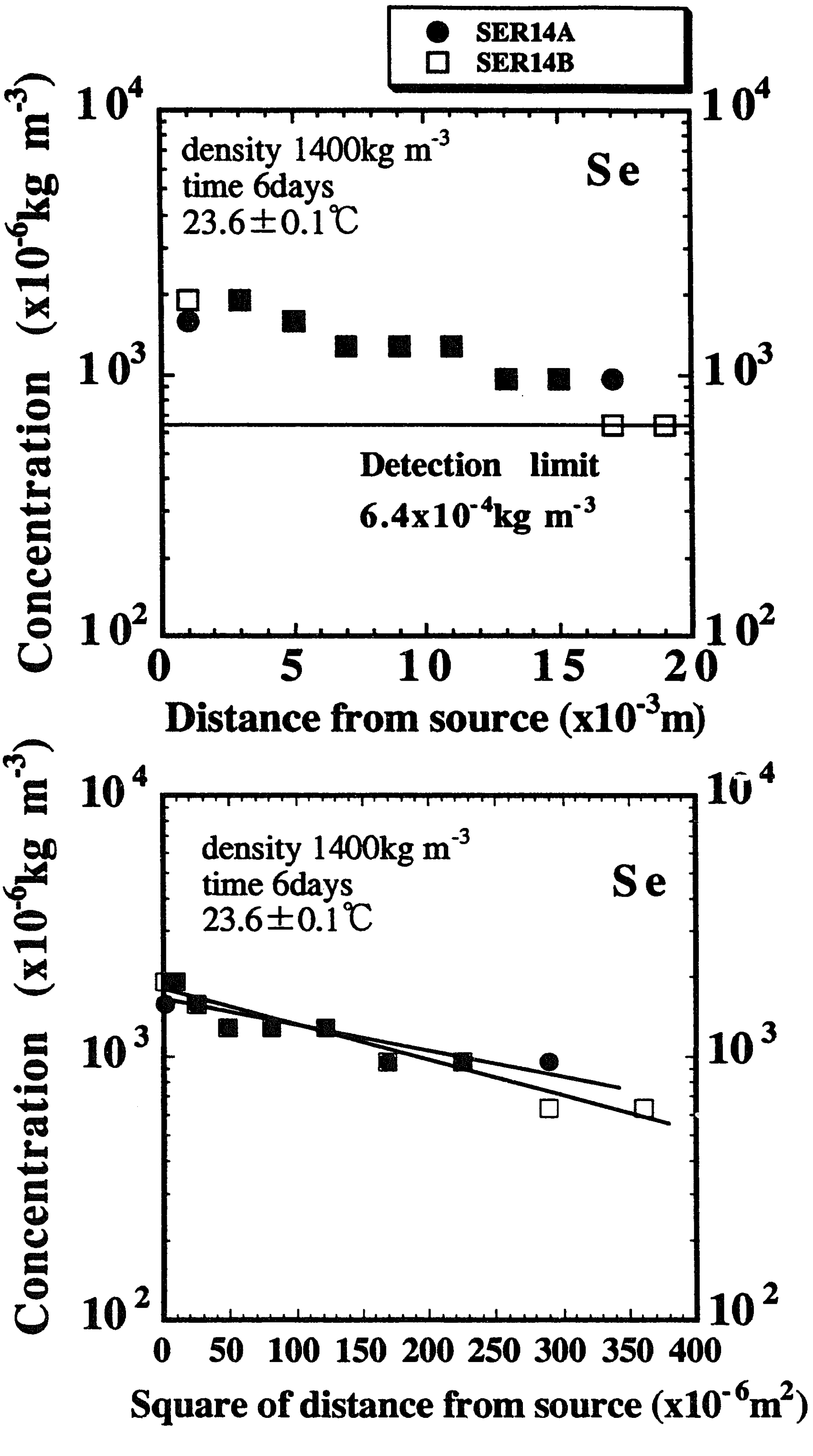

Figure 11 Concentration profiles of Se in compacted bentonite as functions of distance (upper Figure) and square of distance from diffusion source (lower Figure) for a density of 1400 kg m^{-3}

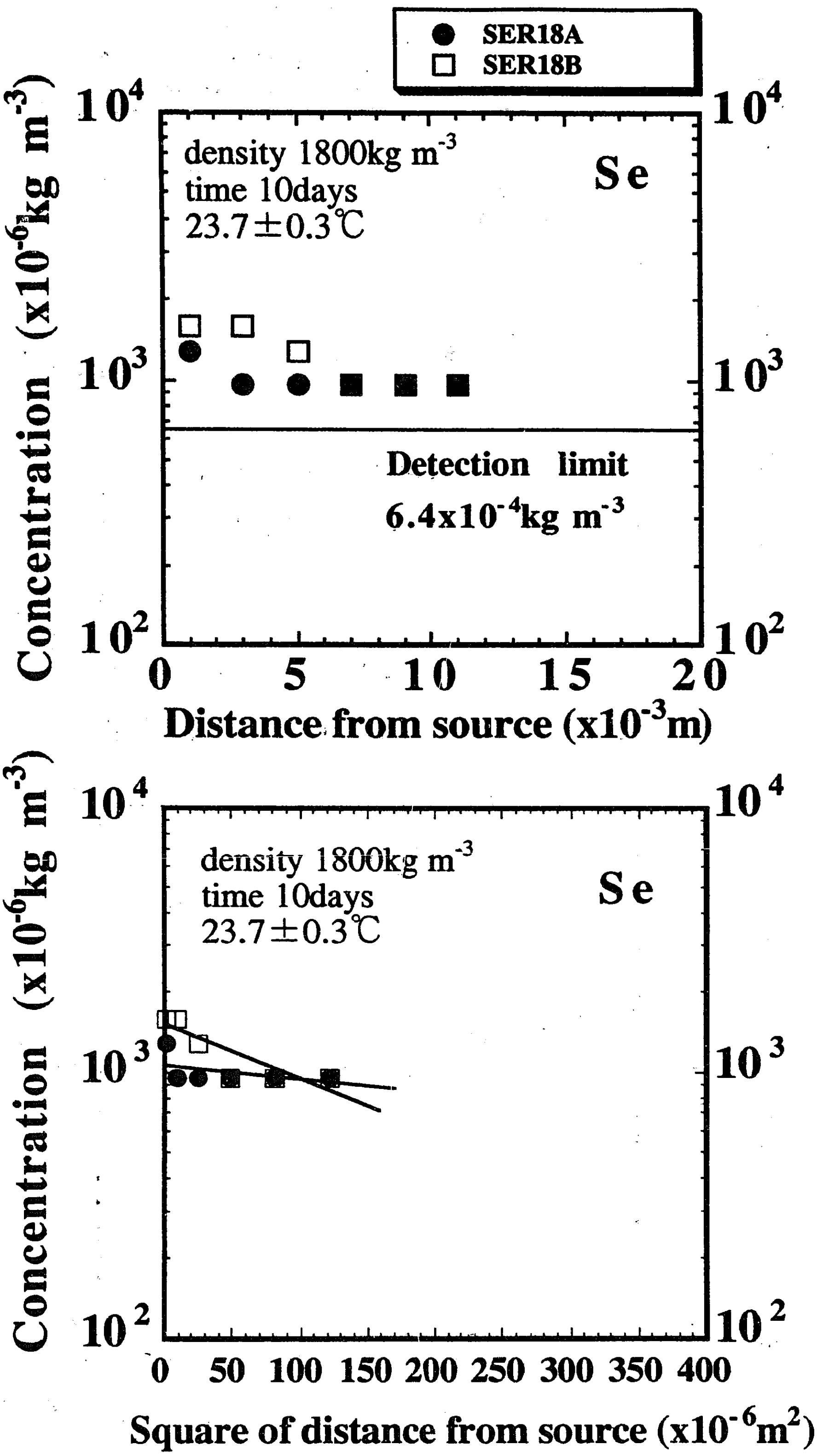

Figure 12 Concentration profiles of Se in compacted bentonite as functions of distance (upper Figure) and square of distance from diffusion source (lower Figure) for a density of 1800 kg m^{-3}

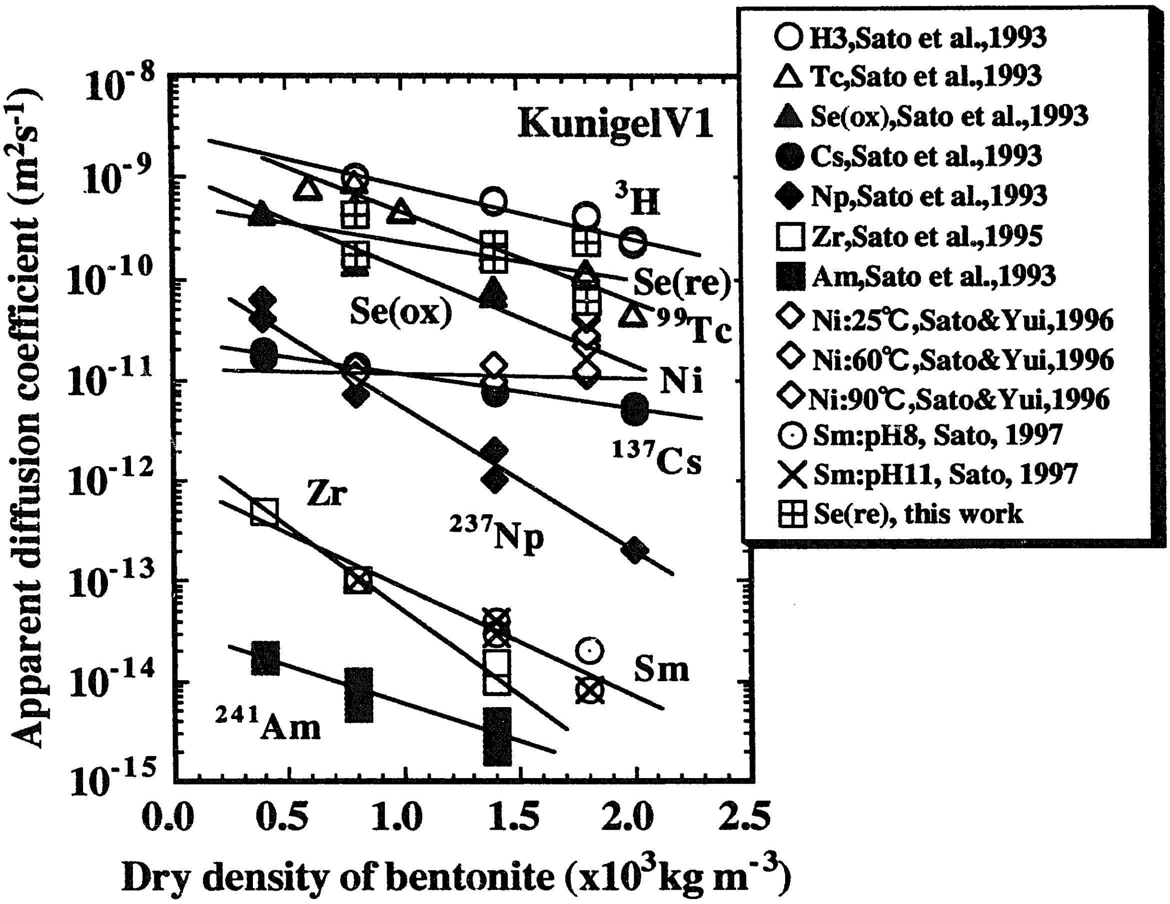

Figure 13 Apparent diffusion coefficients for nuclides and elements as a function of dry density of bentonite obtained to date