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Development of Performance Assessment Methods for TRU Wastes Isolation System

This report was prepared under contract with Power Reactor and Nuclear Fuel Development Corporation

SUMMARY

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本資料についての問合せは下記に願います。注）
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Performance Assessment Method for Transuranic Waste Isolation System And It´s Research and Development Plan

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Abstract

It is expected to accumulate large quantity of transuranic wastes (and long lived β/γ low level radioactive wastes). In the circumstances, Atomic Energy Committee recommended that it is necessary to present applicable technology for isolating transuranic waste till 2000.

The long term safety of isolation technology must be convincingly assessed prior to its application. Therefore models which can predict release and migration of radionuclides with adequate accuracy need to be developed and validated. In this investigation, the purpose of which is to identify principal phenomena for performance assessment and to integrate simplified system models for overall safety assessment.

In 1993 four subjects listed below were investigated.

(1) Scenario development for Performance assessment of TRU waste isolation.

(2) Investigation of assessment methods for important phenomena of TRU waste isolation.

(3) Preliminary assessment for gas generation from TRU wastes.

(4) Development of total performance assessment methdoology and of R&D plan.

Based on these investigations, Basic scenario which involve important phenomena for migration of radionuclides from transuranic waste to biosphere were identified and the influence of gas generation was estimated preliminarily. Also the important item that need to research and develop were selected and the framework of R&D plan was reviewed.

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CONTENTS

1.Scenario Development of Performance Assessment for TRU Waste Isolation System

1.1 Review of Characteristics and Isolation Concepts for TRU Wastes

1.1.1 Characteristics of TRU Wastes

Characteristics of TRU wastes which will be disposed in Japan was preliminarily defined, considering wastes generating from reprocessing plant in Japan, France and England and MOX fuel fabrication plant in Japan. defined material composition was listed in Table 1.

1.1.2 Concepts of Geological Isolation System

Concepts of isolation systems which would be assessed in this study was defined. We considered two types of isolation system, one was vault type and the other was silo type. Defined concepts and major features were described in Table 2.

1.2 Preliminary Screening of FEP's

Features, events and processes which will significantly influence the safety of TRU wastes isolation system were reviewed and combined to construct a reference scenario.

Table-1 Substances which compose TRU waste

Classification	Type	Substance/Structure	Source	Feature/Remarks
Organic materials	Natural (analogous) substance	Cellulose, elastomer	Returnable miscellaneous solid waste	• Possibility of providing as substrates for microorganisms thereby producing gases and degradation product.
		Activated charcoal	PNC chemical sludge	• Ditto
	Synthetic macromolecular compound	Asphalt	Bituminized waste	• Ditto • Influence on sorption characteristics
		Vinyls such as vinyl cloride	Returnable miscellaneous solid waste	• Possibility of providing as substrates for microorganisms thereby producing gases and degradation products.
		Synthetic resins such as polyethylene, epoxy, etc.	Ditto PNC solidified degraded solvent	• Ditto
	Low molecular weight compound	TBP, dodecane	PNC solidified degraded solvent	• Ditto
Inorganics	Silicate	Cement	Various cemented waste, solid waste, structures, filling material	• Naturally exists widely in a large quantity in the crust and principal mineral component of rocks • Generally insoluble to water except for alkali salts
		(Unknown composition and structure)	Hydrothermal solidified waste	
		(Ditto)	Melted incineration ash	
		Borosilicate glass	Miscellaneous solids, vitrified waste, burnable poison	
		Calcium silicate	Micellaneous solid waste (thermal insulators)	
		Silver exchanged zeolite	Spent iodine adsorbent	

inorganics	Oxide	Metal oxides such as AL_2O_3, SiO_2, Fe_2O_3, etc. FP·CP oxides such as Mo, Ru, Zr, Co, etc.	Incineration ash Returnable centrifuge cake MEB clad	
	Process additive (and neutralized salt)	Neutralized salts such as $NaNO_3$, $NaNO_2$, Na_3PO_4, Na_2HPO_4, etc.	Returnable chemical treatment sludge Solidified concentrated liquid waste Chemical treatment sludge (degraded solvent)	· Soluble to water · Possibility of providing as substrates for microorganisms thereby exerting influences on geochemical conditions · Possibility of exerting influences on solubility and sorption characteristics of nuclides
		Co precipitant such as $CaCO_3$, $BaCO_3$, $BaSO_4$, $Fe(OH)_3$, $La(NO_3)_3$, etc.	Returnable chemical treatment sludge, PNC chemical sludge Chemical treatment sludge (degraded solvent, concentrated liquid waste)	· Insoluble to water
	Metal	Standardized alloys such as magnox, zircaloy, stainless steel, inconel, etc.	Magnox swarf, hull, end piece, channel box, burnable poison	· Corrosiveness varies according to composition of each alloy · Change of corrosion rate due to contact of dissimilar metals
		Alloys such as Fe, Cr, Ni, Cu, Zr, etc.	Solidified molten metal, miscellaneous solids, hull, end piece, channel box, burnable poison	· Corrosiveness varies according to composition of alloy

Table - 2 Concepts and Major Features of Isolation System

Concept		
Large Cross Sectional Facility	Vault Type	
	Silo Type	
Engineered Barrier System		
Waste Package	Waste Form	Metals, Metal Oxides, Salts, Silicates, Artificial Polymeric Compounds, Other Organics
	Solidifying Material	Cement, Asphalt, Plastic, etc.
	Waste Container	Metals, Cement
Buffer (Filling) Material	Bentonite, Silica Sand, Cement Concrete, Crushed Rock, etc.	
Artificial Structure	Reinforced Concrete	
Emplacement Depth		
Minimum Depth	Geological structure deeper than 100 [m] : Considering a future human intrusion	

2.State of the Art of Research and Development on Performance Assessment Methods

2.1 Gas Generation and Migration in Barrier System

To assess the effects of gas generation, migration and accumulation, We have to simulate a system with two mobile phases as gas and liquid. Also we have to consider some components in the unsaturated isolation system such as air, other gases and water.

Researches and developments on gas and groundwater migration in unsaturated geological media were implemented in the fields investigating oil and gas reservoir system.

The typical mathematical model used for two-phase flow simulation was given by following equations. The governing equations of two - phase flow are those of mass and momentum balance. These equations are supplemented by other equations. Those are for state of gas and vapor, saturation dependent relative permeability and saturation dependent capillary pressure.

2.2 Influence of Colloids and Chemical Buffer Effect of Cement Material

2.2.1 Influence of Colloids

It has been confirmed that the colloids in the groundwater has markedly enhanced migration of several radionuclides in the geosphere. For this reason, much attention has been attracted recently to the groundwater chemistry of colloids in the field of the study of actinide migration in the geological media. The subjects of research and development on influence of colloids were summarized in table 3.

2.2.2 Chemical Buffer Effect of Cement Material

Investigations on chemical buffering effect of cement materials has implemented especially by NAGRA in Switzerland and by AEA in England.

In Switzerland, in order to describe the long term chemical behavior of cement materials, a series of submodels including the hydration of cement, the thermodynamic properties of calcium - silicate - hydrates and the degradation of cement in natural groundwater have been developed. NAGRA have assessed the degradation of cement materials on the repository condition and following conclusion are drawn.

1)The lifetime of cement materials strongly depends on the chemical composition of the groundwater.

2)The influence of cement composition on the lifetime is not significant rather than influence of groundwater composition.

3)The pH value of pore water is determined by calcium / silicate ratio of cement material.

4)On the selection of specific cement, One should not decide it based on expected lifetime, should based on duration of maintaining high pH value.

AEA also have assessed the long term change of pH value of pore water in the repository using a developed geochemical model which estimate the equilibrium composition between solid phase and liquid phase. Some examples of conclusion derived from recent investigations are listed below.

1)Composition of groundwater is generally important parameter for behavior of chemical buffering.

The important chemical species are Mg^{2+}, dissolved CO_2, SO_4^{2-} and HCO_3^-.

2)Reaction with organic composition of wastes and additives for cement must be considered to evaluate the pH buffering of pore water.

One of the typical cement additive is pulverized fuel ash.

Table - 3 Subjects of Study on Colloids Generation and Migration

Item	Subject	Parameter
Characterization of Colloids	•Sampling Method •Measurement Method for Concentration of Colloid •Measurement Method for Size and Size Distribution of Colloid •Measurement / Evaluation Method for Other Parameters •Stability of Colloid	Concentration of Colloid Size, Size Distribution Surface Potential Chemical Composition Molecular Structure
Mechanisms of Colloid Generation	•Generation from Barrier Materials and Wastes •Generation from Organic / Degradation Products •Generation from Microorganisms •Generation from Rock, Clay and Groundwater Components	pH, Eh, Ionic Strength, Cation Concentration, Colloid Density, Particle Density
Reaction with Radionuclides	•Radionuclide Sorption by Complexation •Influence of Change of Legands •Influence of Interactions between Colloids (Coagulation) •Kinetic Model	Capacity of Electric Double Layer Surface Potential pH, Eh, Ionic Strength, Cation Concentration, Colloid Density
Interaction with Barrier Materials	•Sorption Model •Influence of Interactions between Colloids (Coagulation) •Influence of Sorbed Ion	Capacity of Electric Double Layer Surface Potential pH, Eh, Ionic Strength, Cation Concentration, Colloid Density
Modeling of Migration	•Application of Filtration Model •Assessment Methods for Influence of ElectroStatic Attraction / Repulsion •Definition of Major Phenomena •Evaluation of Parameters for Migration Model •Coupling with Generation, Sorption and Interaction with Barrier Materials •Coupling with Conventional Advection / Diffusion Models	Surface Potential, pH, Eh, Ionic Strength, Cation Concentration, etc.

3.Preliminary Performance Assessment

In this study, as one of the most important phenomena for performance assessment of isolation system for TRU waste, the influence of gas generation in the TRU waste repository was investigated. The purposes of this estimation are preliminary analysis for influence of gas generation on groundwater flow, on pressurization in a repository structure, and on sensitivity of some major parameters. A two-phase flow simulation code "TOUGH" was selected and used for these purposes.

3.1 Definition of Simulation Case

Types of repository, geometry of repository, characteristic values of barriers and other initial conditions are listed in table - 4. And idealized geometry for two types of repository , vault type and silo type, are shown in figure 1 and 2 respectively. For both types of repository, the shape of repository was approximated to rectangular parallelepiped from cylindrical shape.

In this simulation, we assumed that after the repository was closed, pores in the repository was filled instantaneously by groundwater. The depth of repository was defined at 200 [m]. The calculated value of pressure, water and gas saturation and pore water velocity is represented by the value for one discretized element defined for each zones of repository.

<u>Vault type</u>

- Waste zone ; Nearest element from center of vault : 1.1[m] from center of vault.
- Back fill zone ; Upper right corner of waste zone : 5.3[m] from center of vault.
- Buffer zone ; Upper right corner of bentonite zone : 7.4[m] from center of vault.
- Disturbed zone ; Upper right corner of disturbed zone : 19.1[m] from center of vault.
- Host rock zone ; Upper right area of host rock zone : 61.5[m] from center of vault.

<u>Silo type</u>

- Waste zone ; Nearest element from center of silo : 2.5[m] from center axis of vault.

•Structure zone ; Center of side wall in structure zone : 13.0[m] from center axis of vault.

•Buffer zone ; Center of side wall in buffer zone : 14.3[m] from center axis of vault.

•Disturbed zone ; Center of side wall in disturbed zone : 30.0[m] from center axis of vault.

•Host rock zone ; Center of side wall in host rock zone : 75.0[m] from center axis of vault.

3.2 Result and Conclusion

3.2.1 Result

• Vault : reference case (case 1)

Figure-3 shows a pore pressure change vs. time. The maximum pressure is 2.9 [MPa] and appeared in a waste and backfill zone about 10 years after closure. A change of gas saturation vs. time is shown in Figure-4. In the waste zone, about 200 years after closure, gas saturation of pore is nearly 40% and continue to increase. In other words, 40% of pore water, may be contaminated, will be pushed out from waste zone in 200 years.

A pressure contour when the maximum pressure is appeared was shown in Figure-5. Pressure buildup is limited in a repository area. Figure-6 shows the gas saturation contour the time at 200 years after closure. A gas migration front reaches about 50 m or less from the center of repository at 200 years after closure. Figure-7 shows the change of pore water velocity vs. time. The maximum velocity is appeared in backfill zone and in buffer zone, value of the maximum velocity is 0.02 [m/y] or more.

• Vault : sensitivity of capillary pressure (case 2)

In this case we have taken into account the influence of capillary pressure, the maximum value of capillary pressure was set at 0.5 [MPa]. Figure-8 indicate that the maximum pressure increase to 3.3[MPa] and appeared in a waste and backfill zone about 10 years after closure. A change of gas saturation vs. time is shown in Figure-9. In the waste zone, about 200 years after closure, a gas saturation of pore decrease to nearly 30%.

• Vault : sensitivity of absolute permeability (case 3)

In next case, we have reduced absolute permeability to 10^{-19} [m2]. Fig.10 indicate that maximum pressure in the repository increase to 5.6[MPa] and appeared in a waste and backfill zone about 18 years after closure. A change of gas saturation vs. time is shown in Figure-11. In the waste zone, about 200 years after closure, a gas saturation of pore decrease to nearly 30%. A comparison of the effects of buffer permeability and capillary pressure on pore pressure in the repository are compared in Figure-12.

Table-4 Parameters for Simulation Cases

	Vault type			Silo Type		
Model Geometry Outer Size Waste (Filling Material) Buffer Material Structure	Rectangular Parallelepiped(2-Dimention) 1.1mH ×13mW (×200mL) 9.0mH × 9mW (×200mL) 1m／2m(Bentonite／Silica Sand) —			Rectangular Parallelepiped(2-Dimention) 55.5mH ×30mW (×30mL) 50.0mH ×25mW (×25mL) 1.5m(Bentonite／Silica Sand) 1m／1.5m(Reinforced Concrete)		
Characteristics (1)	Density [kg／m^3]	Porosity [−]		Density [kg／m^3]	Porosity [−]	
Waste (Filling Material) Buffer Material Structure Host Rock(Disturbed Zone)	2.4×10^3 2.7×10^3 — 2.6×10^3	0.35 0.30 — 0.03(0.10)		2.4×10^3 2.7×10^3 2.6×10^3 2.6×10^3	0.35 0.30 0.35 0.03(0.10)	
Characteristics (2)	Absolute Permeability [m^2]			Absolute Permeability [m^2]		
Waste (Filling Material) Buffer Material Structure Host Rock(Disturbed Zone)	10^{-15} 10^{-18} (Case 1,2), 10^{-19} (Case 3) — 10^{-17} (10^{-15})			10^{-15} 10^{-18} (Case 1,2), 10^{-19} (Case 3) 10^{-17} 10^{-17} (10^{-15})		
Relative Permeability Function	Corey's Model	Slr=0.25 Sgr=0		Corey's Model	Slr=0.25 Sgr=0	
Capillary Pressure Function	Narasimhan's Model	Pe=0(Case 1,3), 0.5[MPa](Case 2) Po=0(Case 1,3), 0.5[MPa](Case 2) η=1		Narasimhan's Model	Pe=0(Case 1,3), 0.5[MPa](Case 2) Po=0(Case 1,3), 0.5[MPa](Case 2) η=1	
Gas Generation Rate	H_2 : 0.3m^3(STP) ／m^3・y			H_2 : 0.3m^3(STP) ／m^3・y		
Initial Condition	Pressure [MPa]	Pore Water Saturation [−]	Temperature [℃]	Pressure [MPa]	Pore Water Saturation [−]	Temperature [℃]
	2.0	1.0	30	2.0	1.0	30

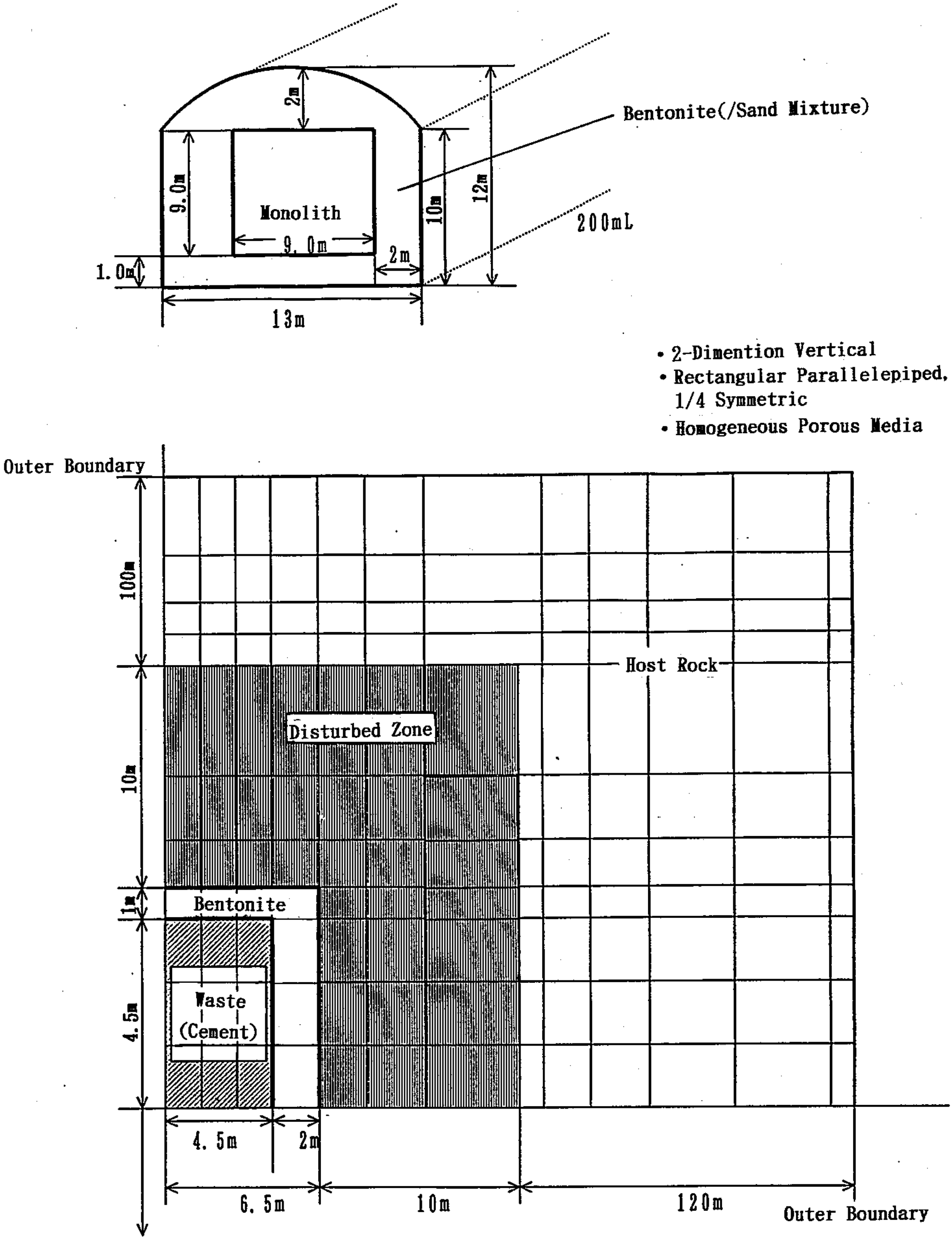


Fig.-1 Idealized Geometry for Vault Type Repository

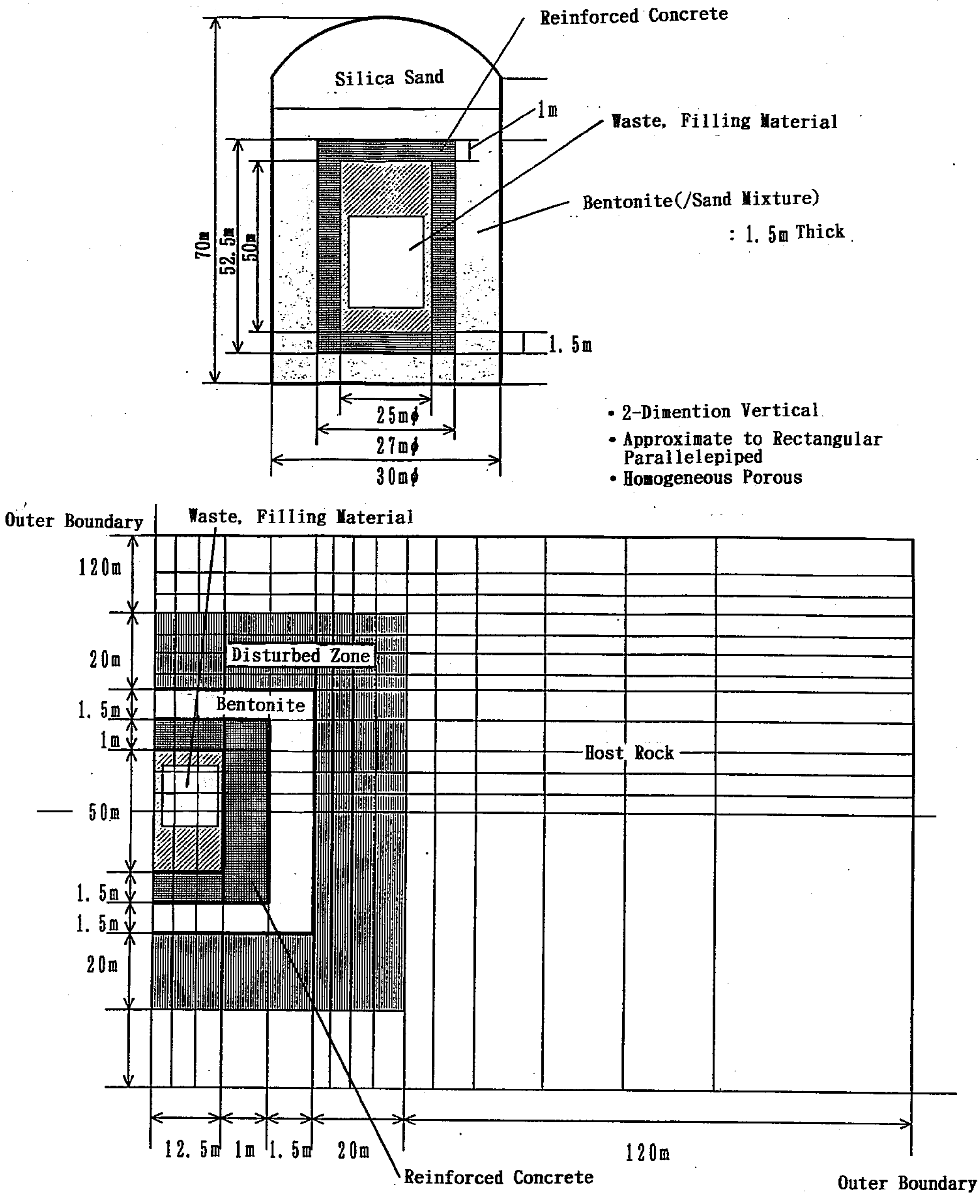


Fig.-2 Idealized Geometry for Silo Type Repository

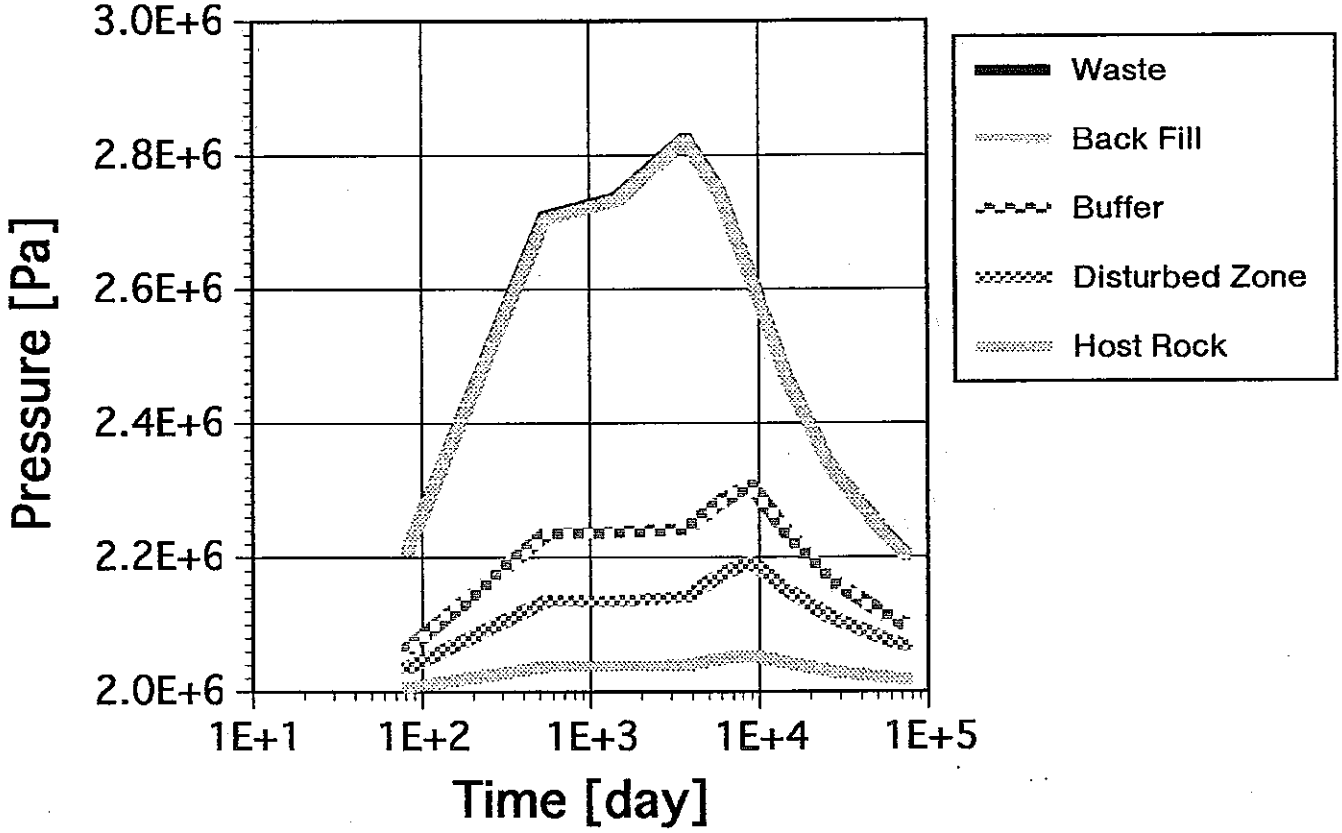


Fig.-3 Pressure for Components (Vault Case1)

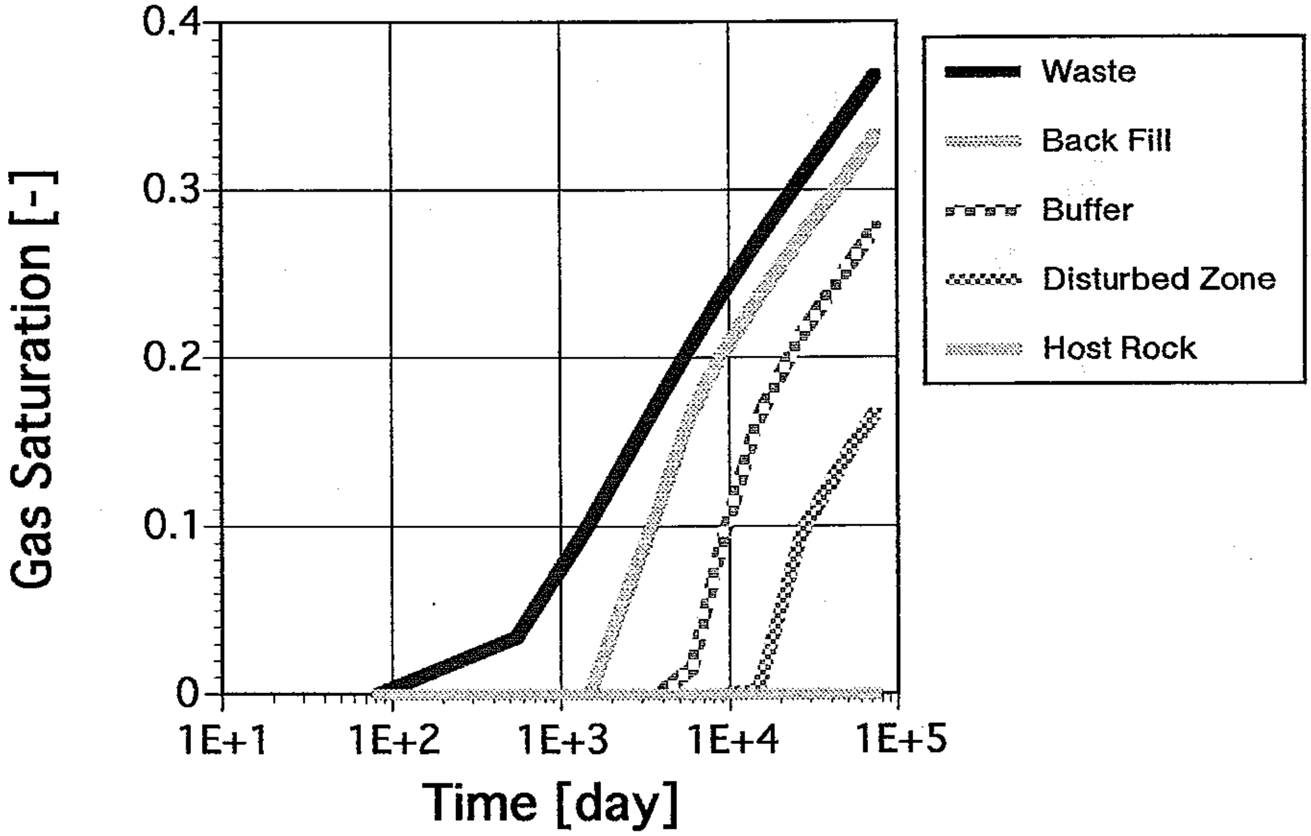


Fig.-4 Gas Saturation for Components (Vault Case1)

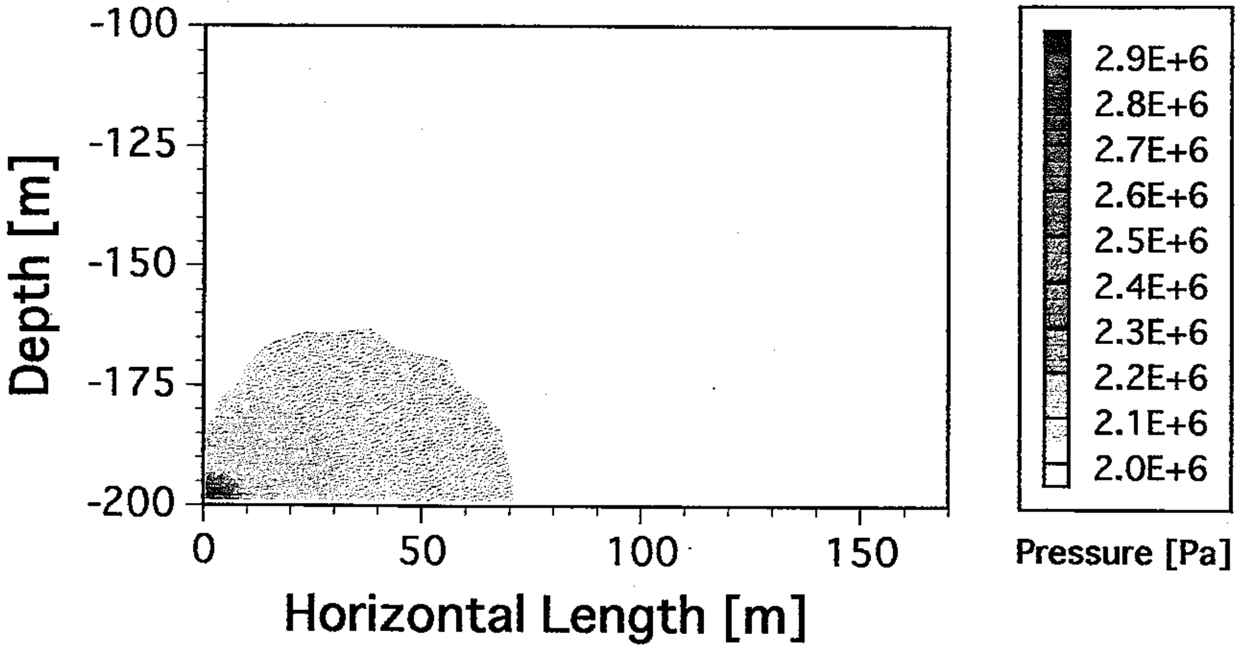


Fig.-5 Pressure Contour (Vault Case1)

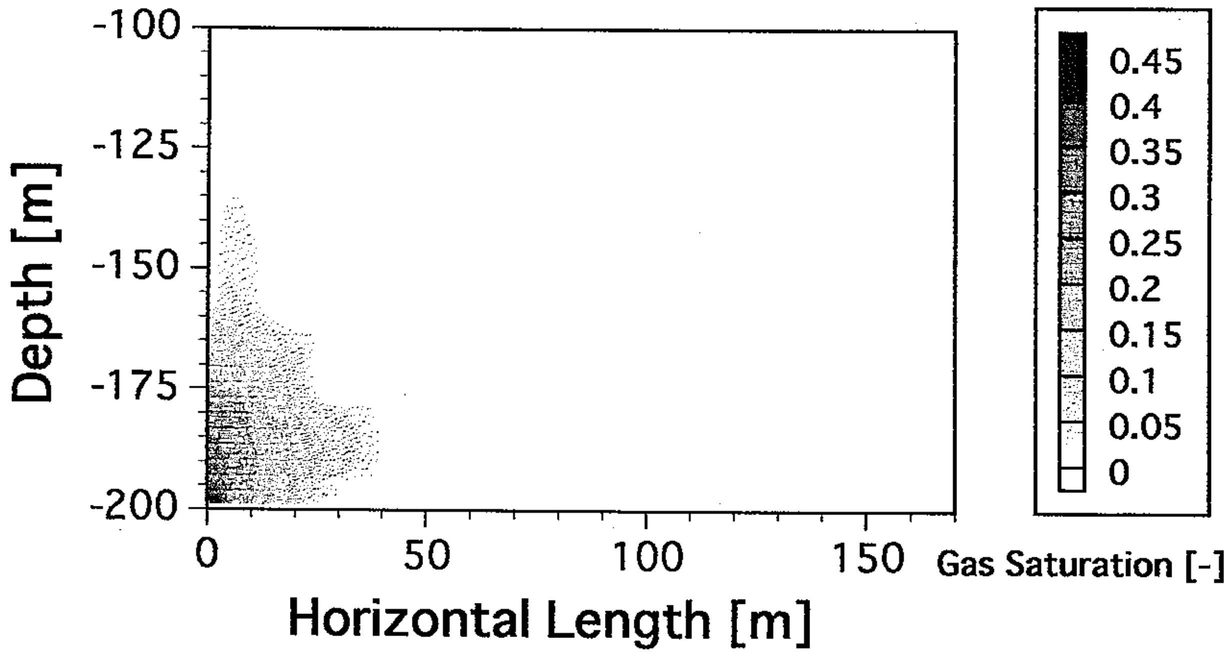
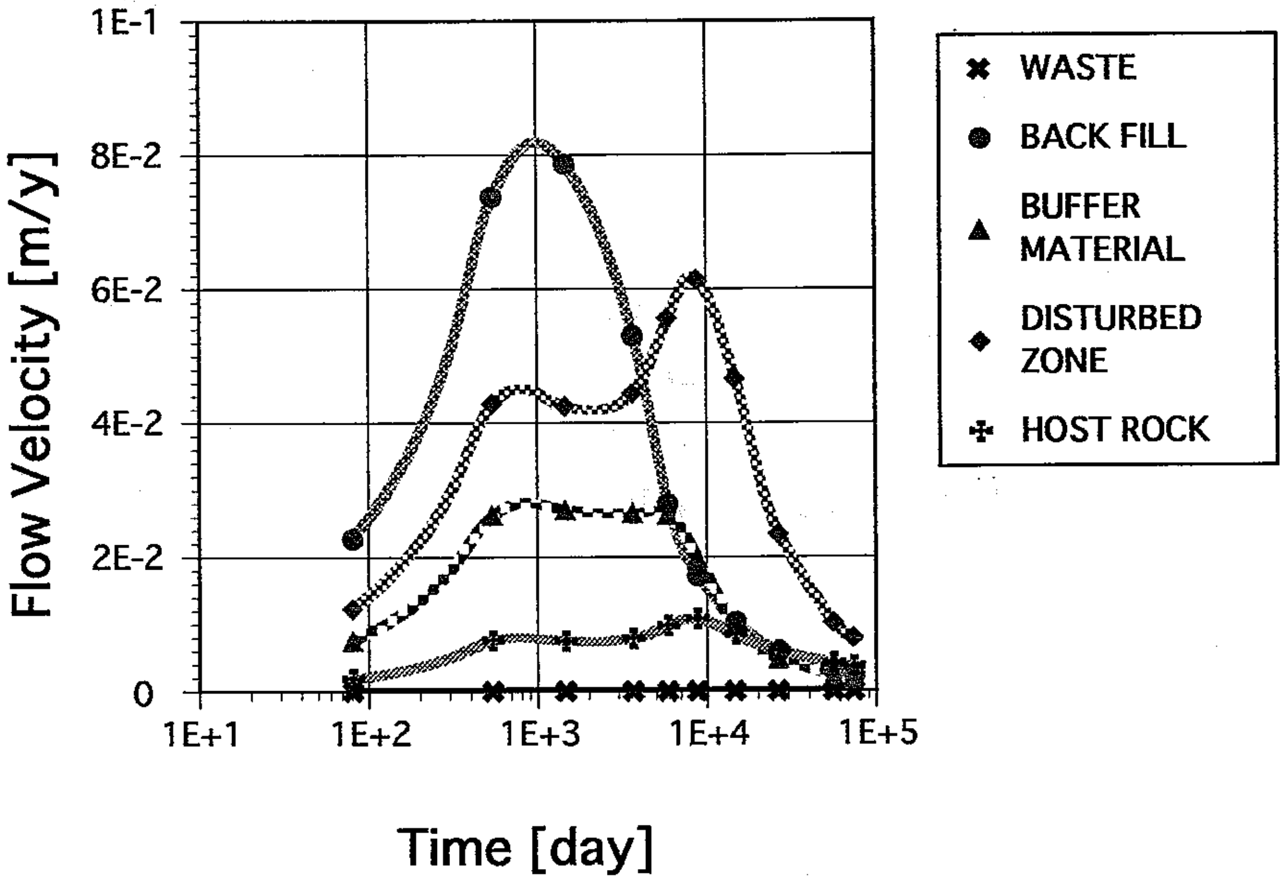


Fig.-6 Gas Saturation Contour (Vault Case1)



Fif.-7 Pore Water Velocity (Vault Case1)

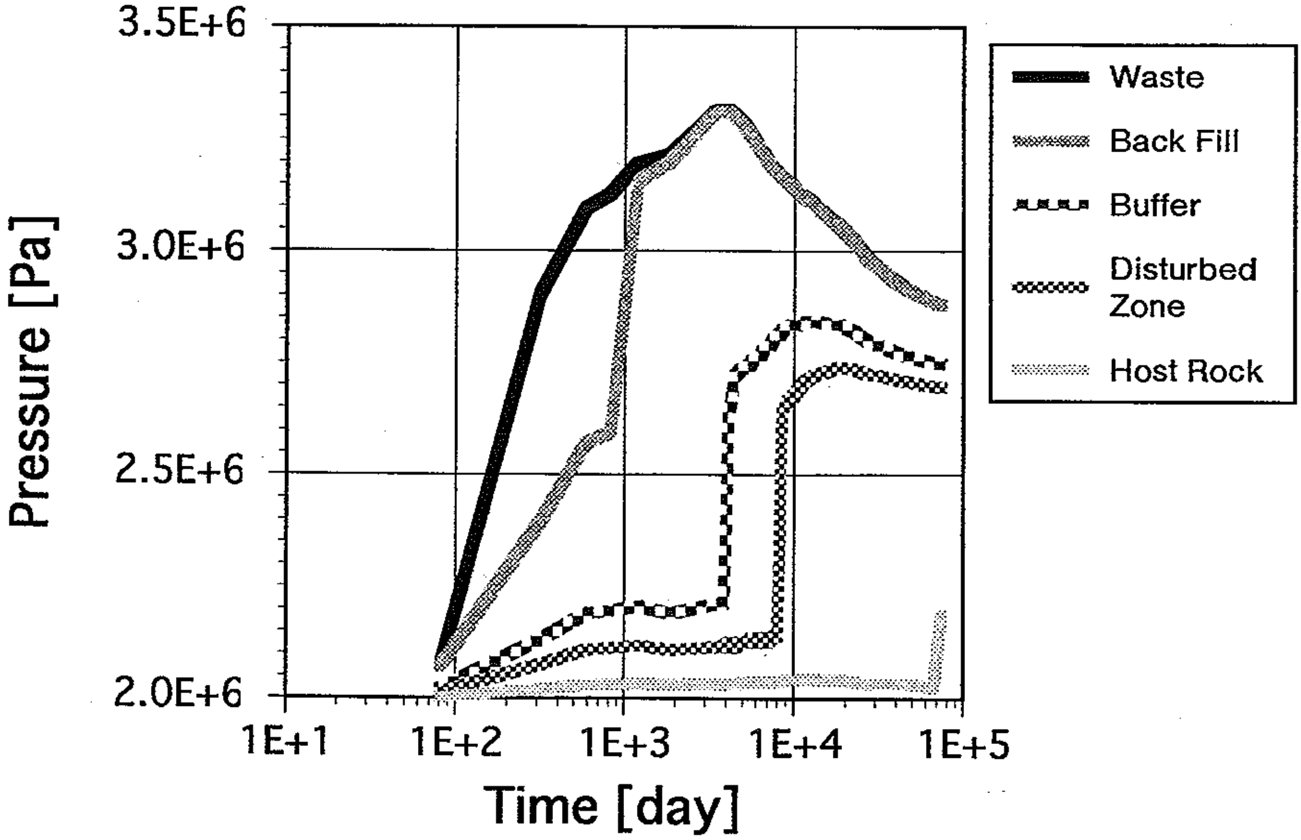


Fig.-8 Pressure for Components (Vault Case2)

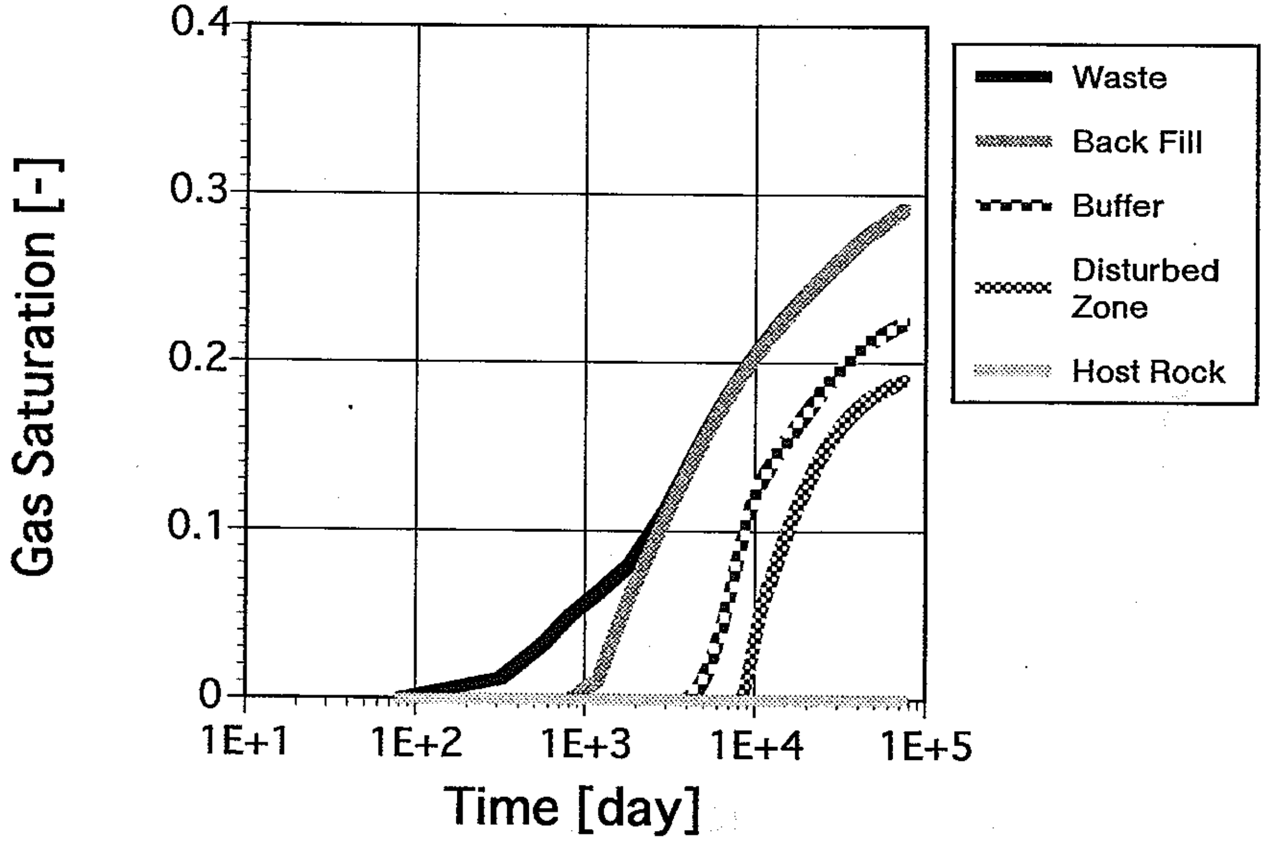


Fig.-9 Gas Saturation for Components (Vault Case2)

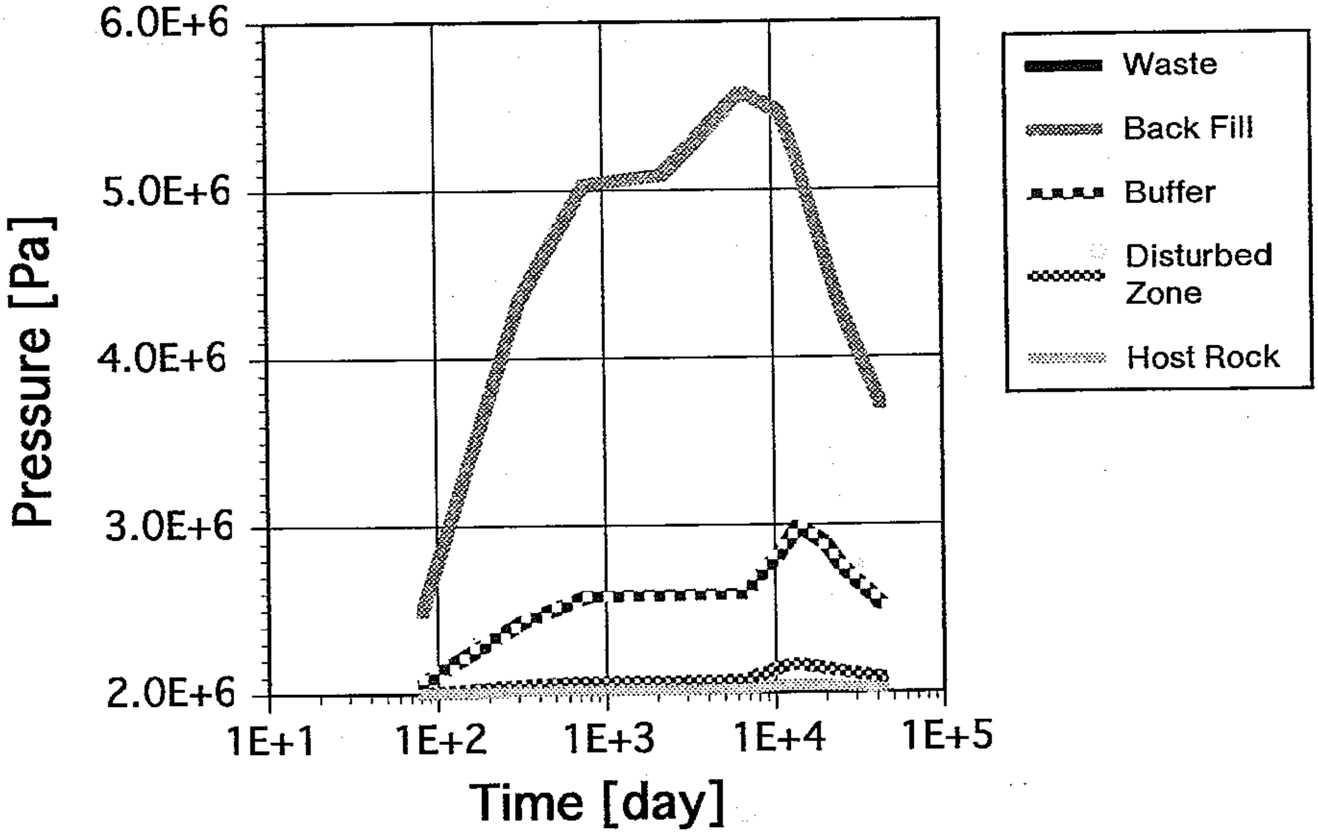


Fig.-10 Pressre for Components (Vault Case3)

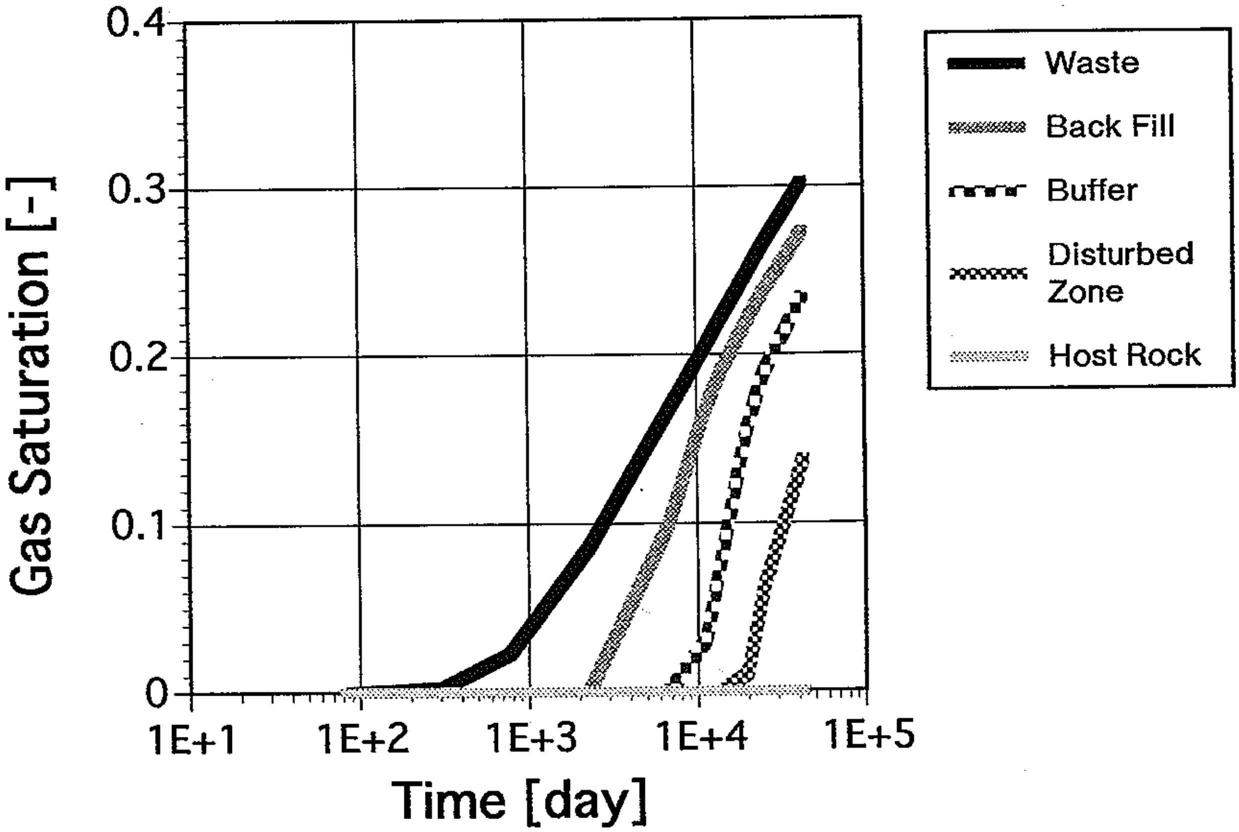
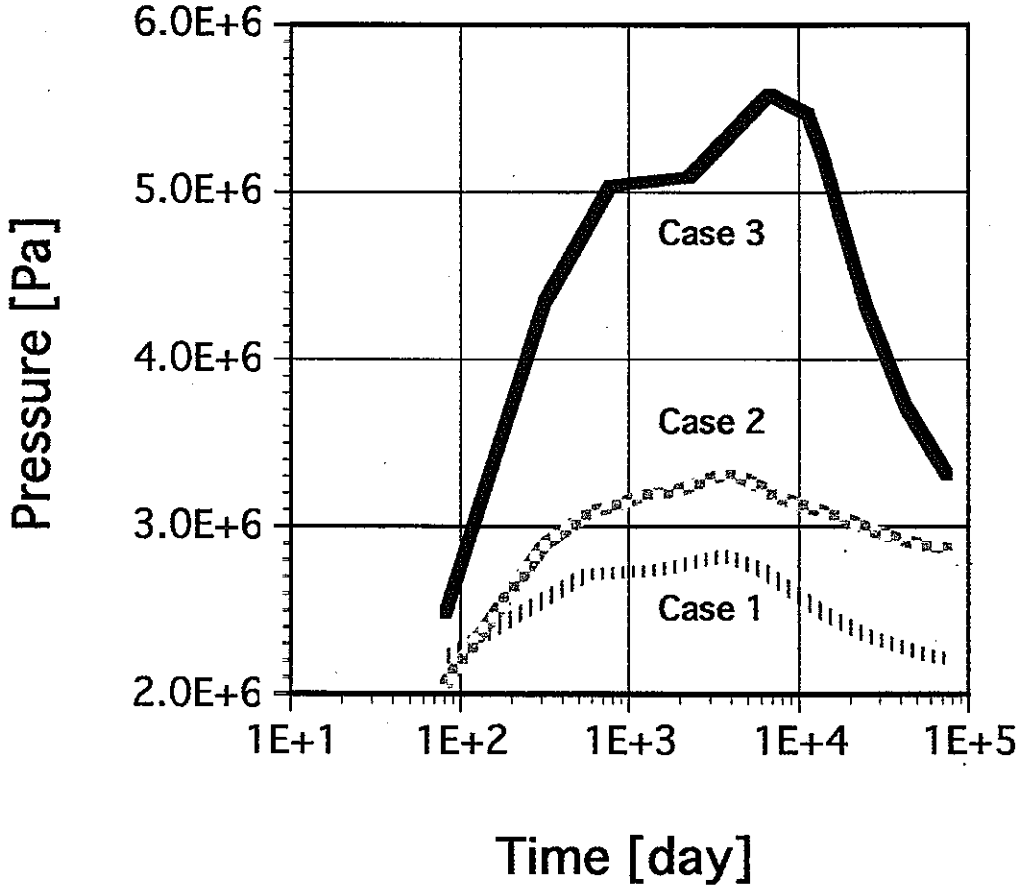


Fig.-11 Gas Saturation for Components (Vault Case3)



Case 1 : Buffer Permeability =1E-19 [m²]
Case 2 : Capillary Pressure < 5E5 [Pa]
Case 3 : Buffer Permeability =1E-18 [m²]

Fig.-12 Effects of Buffer Permeability and Capillary Pressure Variation

3.2.2 Conclusion

The conclusion of this preliminary assessment was arranged from three view points.

1)Pressure buildup

- The pressure buildup in a repository is significantly influenced by permeability of buffer material.
- The simulation cases considering a capillary pressure also indicate increasing maximum pressure, but not significantly than variance of permeability value.
- The pressure buildup in a repository is significantly influenced by permeability of buffer material

2)Gas saturation

- Transport length of gas bubble is also influenced by permeability of barrier materials and capillary pressure.

3)Groundwater flow

- Pore velocity of groundwater might be increased by buildup of gas pressure to the order of 0.01[m/y] or more.
- This indicate a possibility that for some period of time the buffer material zone fill the role of diffusion barrier no longer.

The Subjects for analysis of gas migration was also arranged from same view points.

1)Parameter

- Data collection for permeability of engineering barrier materials and geological materials.
- Data collection for capillary pressure of barrier components.
- Identification of the relationship between two-phase flow parameters, such as relative permeability and capillary pressure, and gas saturation or water saturation.

2)Initial and boundary condition

- Influence of residual air and groundwater intrusion after the closure of repository.
- Influence of gas generation and migration on groundwater flow in wide surrounding

area.

- Simulation for longer period (at least order of ten thousands of years).

3)Simulation model

- Coupling with solute transportation and interaction between solute (radionuclide) and matrix (barriers).
- Simulation models and tools for unisotropic fractured media.
- Coupling with chemical and geochemical interaction between gas and other dissolved components.

4.Model Chain of Performance Assessment and Subjects of Research and Development

4.1 Model Chain and Procedure for Development of Assessment System

The procedure of development and components of comprehensive performance assessment method for TRU waste isolation system was investigated. In this study, FEP's related to the groundwater scenario was considered. Other scenario related to phenomena such as human activity, geological / climatological were excluded. The concept of constructing a comprehensive performance assessment system for TRU waste isolation system was shown in Figure-3.

4.2 Subjects and R&D Program

4.2.1 Subject for R&D on TRU waste disposal

The subjects which should be investigated was selected taking into account the subjects and R&D schedule of high-level radioactive waste disposal. The applicable products from R&D program for geological disposal of high-level radioactive waste was extracted and listed below.

• Scenario development

Radionuclide migration scenario in the near field related to hydrogeology, groundwater chemistry, geochemical reaction, corrosion of metals. Radionuclide migration scenario in the far field related to reaction with natural organics. Waste exposure scenario and human intrusion scenario for geological disposal.

• Performance assessment method of near field barriers

Analysis of hydrology, Corrosion of carbon steel, Radiolysis of water, Analysis of thermal condition, Swelling of bentonite, Creep behavior of rock, Analysis of stress phenomena, Leaching of radionuclide from waste form, Analysis of geochemical condition, Distribution coefficient in the buffer material, Evaluation of radionuclide solubility, Characteristic values of engineered barriers

• Performance assessment method of far field barriers

Analysis of regional hydrology, Evaluation of geological environmental condition, Radionuclide Transport by advection and dispersion, Analysis of geochemical condition, Influence of natural colloid on radionuclide migration, Distribution coefficient in geological media, Evaluation of radionuclide solubility, Characteristic values of geological media

4.2.2 R&D Program for TRU waste disposal

The research and development program for TRU waste disposal should be formulated taking account of R&D program for geological disposal of high-level radioactive waste in order to apply research products mutually. From this point of view R&D program for TRU waste disposal was made based on policy defined below.

1)"Characterization of TRU wastes" should be implemented principally till 1997 so that the basic data of performance assessment can clarify earlier time.

2)In parallel with waste characterization, the concepts of isolation system and phenomena that have to be analyzed should be investigated. And the comprehensive performance assessment system will be developed after scenario development, modeling, validating and simplifying models and integrating models.

3)Basic researches for the phenomena with regard to activity of microorganisms, colloid generation and other recently recommended subjects will be started earlier time.

4)Ultimately comprehensive performance assessment system will be constructed till 2003.

R&D schedule for TRU waste disposal is shown in Figure-14.

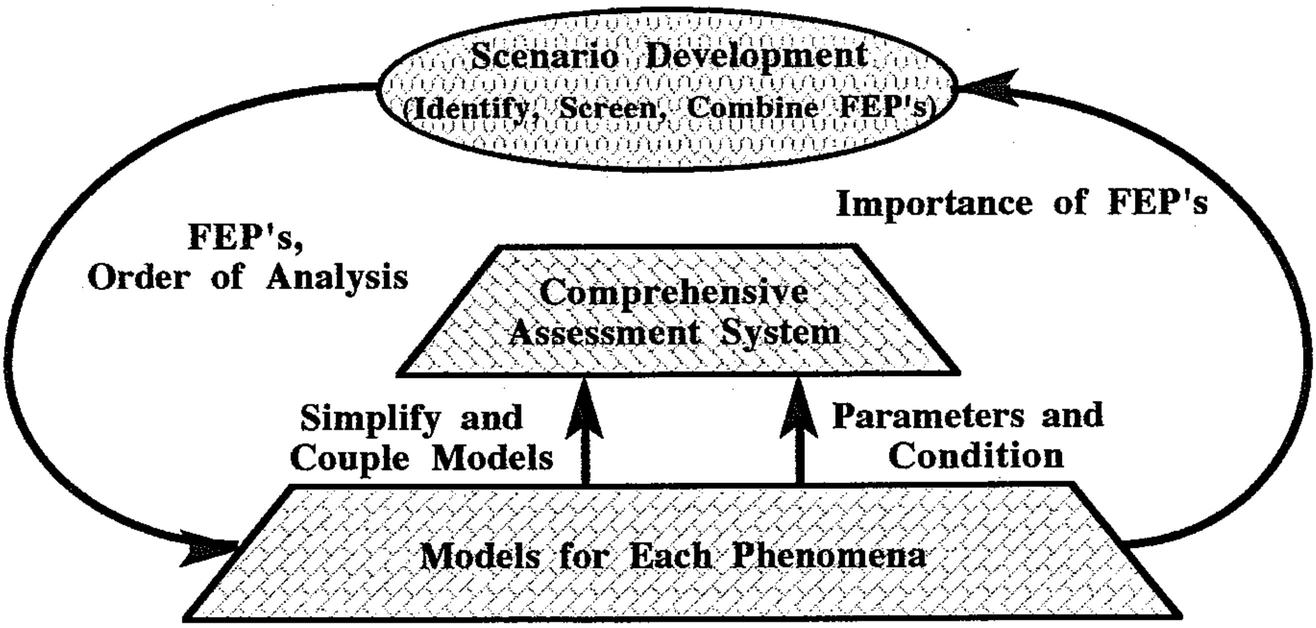


Fig.-13 Concept of comprehensive performance Assessment system

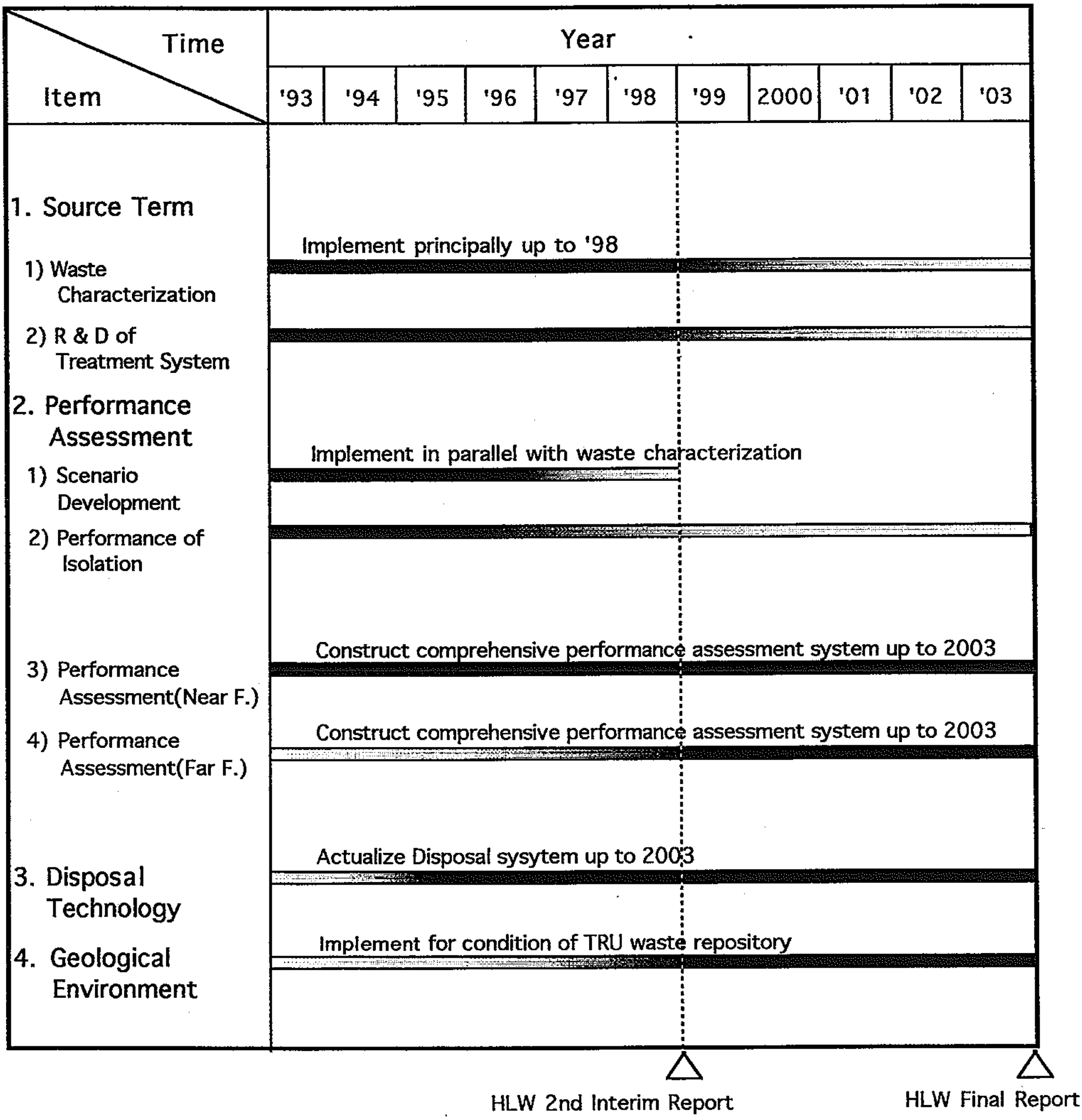


Fig.-14 R&D Schedule for TRU Waste Disposal