Studies on Engineered Barrier System for TRU Waste Isolation with Consideration to Characters of TRU Wastes

ABSTRACT

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Studies on Engineered Barrier System for TRU Waste Isolation with Consideration to Characters of TRU wastes.

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Abstract

This study aimed to discuss with the safe and resonable engineered barieer system of TRU waste isolation and to extract research theme typical to the TRU waste. The followings are items and results at this year.

(1) Arrangement of information about TRU waste
Existed informations about TRU waste were arranged.

(2) Discussion with basic concept of engineered barieer system.
TRU waste isolation systems in overseas countries were surveyed and the components and functions of engineered barrier system were arranged. The method of safety assessment and the chemical behaviors of nuclides were surveyed.

(3) Classification of TRU waste and provisional engineered barrier system
TRU wastes were re-categirized into nine items. Time dependence of hazard index of each waste was calculated. The provisional suitable barrier system were discussed and the hybrid usage of cementaneous materials and bentonite was considered as essential barrier components.

(4) Comparison with safety performance of engineered barieer systems
Calculations of migration through engineered barieer system were carried out with solubility limit and radioactive decay chain. I-129 in iodine adsorbent has the highest release rate. In case of the other wastes Pu-242 and Ra-226 have the peak of flux.
Alternation of Na-bentonite to Ca-bentonite was also calculated.

(5) Extraction of research theme typical to TRU waste.
Based on the above discussions, research theme typical to TRU waste were extracted and hierarchical arrangement was carried out.

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Contents

Chapter 1 Arrangement of informations about TRU waste.

Existed informations about TRU waste were arranged to discuss isolation systems of TRU waste.

The difinition of TRU waste and the basic line of isolation should be considered along the guide proposed by Atomic Energy Commission (AEC) at July in 1991. The basic line proposed that the criteria concentration of TRU waste will be approximately 1 GBq/t of α nuclide. In this report, however TRU waste is assumed as the waste generated in the facility in which a large quantity of TRU nuclides are used, for instance, reprocessing plant.

Characters of TRU waste arranged were condition of generation, treatment technology, nuclide component and concentration, and etc. In Japan, the typical facilities in which a large quantity of TRU nuclides are used are the spent fuel reprocessing plant and the MOX fuel manufacturing plant in Tokai facility of Power Reactor and Nuclear Fuel Development Corporation (PNC). Therefore the actual results in these plants are very inportant and plactical. In the reprocessing plant, wastes are itemized into 20 kinds mainly by the place of generation. In the MOX plant, liquid waste are treated in the same process of the reprocessing plant, and solid wastes are itemized into 3 kinds. These are shown in Table 1.1.

PNC has the actual analysed data of TRU waste which is shown in Table 1.2. The concentration in the Table are for the waste before treatment. For the values of treated wastes concentration change through treatment should be considered. PNC also has the calculation data of Hull/end piece burn up and asphalt solidified waste. These data are shown in Table 1.3 and table 1.4. They are useful to estimate the inventory in each waste.

Table 1.1 Classification of radioactive waste generated in reprocessing plant of PNC

Category			Source	contents
solid waste	Uranuim waste	comb.	U enrichment/denitrization UO_3 storage	paper, wood, polyethlene
		imcomb.		grove, vinyl chloride, gamtape
		noncomb.		metal, glass, concrete, filter
	TRU (solid) waste	comb.	Pu enrichment Pu storage Pu conversion	paper, wood, polyethlene
		imcomb.		grove, vinyl chloride, gamtape
		noncomb.		metal, glass, concrete, filter
	$\beta\gamma$ (solid) waste	comb.	the other processes of U and Pu waste generation	paper, wood, polyethlene
		imcomb.		grove, vinyl chloride, gamtape
		noncomb.		metal, glass, concrete, filter
highly active solid waste	Hull/end piece		pretreatment, dissolution	Hull, assembly attachment
	active solid waste		extraction	filter, stirrer, pump
	wasted jag		analysis	vial, cartridge
liquid waste	high level		1st of separation	extracted liquid
	low level		extraction, gas line	wash water of solbent and gas
	very low level		acid collection/evaporation	drain water
	chemical sludge		chemical treatment of liquid	sludge
	solvent		extraction	TBP(30%)-n-dodecane(70%)
	dilution agent		extraction	TBP(10%)-n-dodecane(90%)
others	iodine adsorbent		gas line	AgX filter (Ag-zeplite, Ag-almina)
	resin		pool water treatment	ion exchange resin
	silicagel		solbent treatment facility	silicagel
	sand		separation	sand
	activated carbon		oil separation	activated carbon
MOX waste	solid waste	comb.	all of MOX plant	paper, wood, polyethlene
		imcomb.		grove, vinyl chloride, gamtape
		noncomb.		metal, glass, concrete, filter

comb. ;combustible
imcomb. ;imcombustible
noncomb. ;noncombustible

Table 1.2 Concentration of total α-nuclides activity in TRU waste of PNC

<table>
<tr><th>source</th><th colspan="2">category</th><th>total α
(MBq/kg)</th><th>total $\beta\gamma$
(MBq/kg)</th></tr>
<tr><td rowspan="8">reprocessing plant</td><td rowspan="3">solid waste</td><td>combustible</td><td rowspan="3">$<10^{0}$</td><td rowspan="3">——</td></tr>
<tr><td>imcombustible</td></tr>
<tr><td>noncombustible</td></tr>
<tr><td colspan="2">Hull/end piece</td><td>$<10^{3}$</td><td>$<10^{3}$</td></tr>
<tr><td colspan="2">low level liquid</td><td>$10^{-1}\sim10^{1}$</td><td>$10^{-1}\sim10^{2}$</td></tr>
<tr><td colspan="2">verylow level liquidquid</td><td>$<10^{-1}$</td><td>$10^{-3}\sim10^{-1}$</td></tr>
<tr><td colspan="2">chemical sludge</td><td>$\fallingdotseq 0$</td><td>$\fallingdotseq 10^{-3}$</td></tr>
<tr><td colspan="2">solvent/dilution agent</td><td>$<10^{1}$</td><td>$<10^{1}$</td></tr>
<tr><td rowspan="3">MOX plant</td><td rowspan="3">solid waste</td><td>combustible</td><td rowspan="3">$10^{0}\sim10^{3}$</td><td rowspan="3">——</td></tr>
<tr><td>imcombustible</td></tr>
<tr><td>noncombustible</td></tr>
</table>

Table 1.3 Fundemental unit of nuclides in Hull/end piece

Inventory = A×B (Ci/kg-HIP of Hull/end piece)

nuclide	A	B (Ci/kg)	nuclide	A	B (Ci/kg)
H - 3	2.59 E - 2	1.67 E -3	U -230	3.59 E -60	1.67 E -3
C - 14	7.54 E - 2	〃	U -232	1.07 E - 6	〃
Cr- 51	6.31 E -35	〃	U -233	1.41 E - 9	〃
Mn- 54	4.11 E - 2	〃	U -234	2.76 E - 5	〃
Fe- 55	5.38 E 1	〃	U -235	3.16 E - 6	〃
Co- 60	1.21 E 2	〃	U -236	2.79 E - 5	〃
Ni- 63	3.23 E 1	〃	U -237	1.30 E - 4	〃
Kr- 85	4.79 E - 1	〃	U -238	3.18 E - 5	〃
Sr- 90	5.55 E 0	〃	U -240	8.65 E -12	〃
Y - 90	5.55 E 0	〃	Np-237	2.42 E - 5	〃
Zr- 93	4.48 E -13	〃	Np-239	5.30 E - 4	〃
Nb- 94	9.94 E -13	〃	Pu-238	1.17 E - 1	〃
Tc- 99	1.16 E - 3	〃	Pu-239	2.89 E - 2	〃
Ru-106	5.55 E - 2	〃	Pu-240	4.04 E - 2	〃
Rh-106	5.55 E - 2	〃	Pu-241	5.15 E - 2	〃
In-113m	1.38 E - 7	〃	Pu-242	2.02 E - 2	〃
Sn-113	1.38 E - 7	〃	Pu-244	5.05 E - 7	〃
Sn-119m	6.23 E - 2	〃	Am-241	1.08 E - 1	〃
Sb-125	2.50 E 1	〃	Am-242m	3.57 E - 5	〃
Te-125m	6.11 E 0	〃	Am-243	2.65 E - 3	〃
I -129	2.49 E - 6	〃	Cm-242	2.87 E - 4	〃
Cs-134	4.17 E - 1	〃	Cm-243	8.05 E - 6	〃
Cs-135	3.53 E - 5	〃	Cm-244	2.74 E - 2	〃
Cs-137	7.10 E 0	〃	Cm-245	1.36 E - 5	〃
Ba-137m	6.75 E 0	〃	Cm-246	1.05 E - 6	〃
Ce-144	2.56 E - 2	〃	Cm-247	6.50 E - 9	〃
Pr-144	2.56 E - 2	〃	Cm-248	2.13 E -10	〃
Pm-147	1.23 E 0	〃	Bk-249	8.40 E -16	〃
Eu-154	1.08 E - 1	〃	Cf-249	1.68 E -12	〃
Ra-226	6.60 E -12	〃	Cf-250	1.67 E -13	〃
Pa-231	9.50 E -10	〃			

Table 1.4 Concentration of nuclides in bituminization waste

nuclide	Bq/m^3	nuclide	Bq/m^3
H - 3	2.20 E 10	U -234	2.80 E 7
C - 14	1.00 E 6	U -235	5.20 E 5
Ni- 63	5.20 E 8	U -236	4.10 E 6
Sr- 90	1.40 E 11	U -238	4.80 E 6
Zr- 93	4.10 E 6	Np-237	5.90 E 5
Nb- 94	1.10 E 6	Pu-238	3.20 E 6
Tc- 99	2.80 E 7	Pu-239	3.20 E 6
I -129	3.30 E 8	Pu-240	4.40 E 9
Cs-135	8.50 E 5	Pu-242	8.10 E 6
Cs-137	1.90 E 11	Am-241	8.10 E 6
		Am-242m	8.10 E 6
		Am-243	8.10 E 6
		Cm-245	8.10 E 6

Chapter 2 Discussion with basic concept of engineered barrier system.

The difinition of TRU waste and the component and the function of engineered barrier system in foreign countries were surveyed to discuss with basic concept of engineered barrier system. Furthermore the method of the safety assessment and the chemical behaviors of nuclides are surveyed.

2.1 Present state of TRU waste isolation in foreign countries

The difinition of TRU waste in each countriy are shown in Table 2.1. In USA, BK, and France, there are the obvious difinition of TRU waste and the criteria value is approximately 3.7 GBq/t (= 0.1 Ci/t) which is higher than the proposed value by AEC Japan.

Components of engineered barrier is the same in each country. It is shown in TAble 2.2. Among these components, only solidified materials and buffer materials will be useful in safety assessment. Therefore these two components must be the essential in the engineered barrier. Cement, asphalt, and plastic are proposed for the solidified materials, and cementaneous materials and clay like as bentonite are proposed for buffer materials.

2.2 Arrangement of safety performance model

The following five items were surveyed and arranged; 1)leachability of nuclide from solidified waste, 2)corrosion of container, 3)nuclide migration through buffer, 4)outer boundary condition of engineered barrier system, and 5) gas generation.

To discribe the leachability many types of model like as solubility limit model or diffusion model are used in the suitable condition. Many of these models are, however, applicable in case that the waste can be assumed homogeneous. The suitable model for miscellaneous waste should be newly considered. In this report, the new hybrid type model was proposed. In this model, the nuclide leaching processes were devided into two regions. The first

Table 2.1 Criteria of nuclides concentration for TRU waste in Various countries

Country	Category	Criteria	Remarks
Japan	TRU waste	not decided (Total-α > 1 GBq/ton)	Atomic Energy Commission
		Total-α > 0.555GBq/ton	License Report for LLW
U.S.A.	TRU waste	α-emitter ($T_{1/2}$>20yr) TRU >100nCi/g (>3.7GBq/ton)	E P A 40CFR 191
	TRU waste	α-emitter ($T_{1/2}$>20yr) TRU, Ra-226>100nCi/g (>3.7GBq/ton)	D O E Order 5820.2
	GTCC	α-emitter ($T_{1/2}$>5yr) TRU >100nCi/g (>3.7GBq/ton)	N R C 10CFR 61
United Kingdom	Intermediate Level waste	α-nuclide >4GBq/ton or β/γ-nuclide>12GBq/ton	D O E
France	Category B (α waste)	α-nuclide >3.7GBq/ton	S C S I N R F S No.Ⅲ.2
Switzer-land	TRU waste	not decided Low/Intermediate level contains α nuclide	N A G R A total disposal amount 4×10^{18}Bq
Sweden	α waste	Low/Intermediate level contains α nuclide	S K B
Germany	contained in pyrogenic waste	Temperature increase in rock < 3 ℃	B f S
Belgium	Category-B MLW-α α-Waste	not decided	NIRAS/ONDRAF
Canada	not decided	not decided	

Table 2.2 Components and functions of waste isolation system.

Barrier	Component	Function of barrier
Engineered Barrier System		
Solidi-fication material	cement/concrete bitumen plastic	• control nuclide release • sorption and control solubility (cement/concrete)
Container	drum can container canister	• eliminate contact of waste and water
Buffer	bentonite mixed bentonite/rock	• control water flow and immersion • sorption
	cement powder grout	• sorption • control solubility
Stracture	concrete pit concrete silo concrete container	• eliminate contact of waste and water • sorption • control solubility
Lining of gallery	concrete lining	• eliminate contact of waste and water • sorption • control solubility
Back fill	bentonite mixed bentonite/rock	• control water flow and immersion • sorption
Natural Barrier System		
Geology	granite rock salt clay hard rock (sedimentary/volcanic)	• control water flow and immersion • sorption • control solubility

region is the nuclide leaching from waste material to pores in solidified material, and the second one is the nuclide migration through the pore toward the surface of solidified waste. The coupling release coefficient and solubility limit models were applied to the former region and the diffusion model was to the latter region.

For the corrosion of container many kinds of logical models are considered.

For the migration through buffer materials the diffusion model is moderate when the clay materials are used as buffer, then the time period of montomorillonite alternation should be estimated.

The many types od outer boundary condition of engineered barrier system can be considered. It is necessary to consider the condition of actual case.

These four items are belong to the ground water scinario. The gas generation is categirozed into the different scheme.

The main coueses of gas generation are the following three phenonena; 1) degradation of oraganic materials, 2)corrosion of metal, and 3)degradation by radiolysis. In the estimation by Sweden, the gas generation in isolation system will be controlled mainly by the corrosion of iron. No way was decided to treat gas generation in safety assessment.

2.3 Discussion with chemical behaviors of nuclides

Chemical behaviors of nuclides are bery important for the engineered barrier system. In this report, nuclides were categorized into six groups and the chemical behaviors of nuclides in each category were surveyed. Based on the results of survey, Table 2.3 showes the functions of engineered barrier. The main chemical functions are the decreasing abilities of solubility by precipitation and of migration velocity by adsorption.

Table 2.3 Grouping of nuclide and corresponding barrier function.

Group of nuclide		Function of barrier
TRU, U Np, Pu, Am, U, etc.		① Decrease of solubility (Reduction) ② Decrease of solubility (Hydrolysis) ③ Adsorption (Cation exchange) ④ Decrease of solubility (Compound reaction)
Transition metal	Co, Ni Fe, etc.	① Decrease of solubility (Reduction) ② Decrease of solubility (Hydrolysis) ③ Decrease of solubility (Compound reaction)
	Nb	① Same as Co, Ni, Fe (conservative case) ② Posibility of no barrier (low solubility)
	Tc	① Decrease of solubility (Reduction) ② Sorption (Mechanism unknown)
Alkaline metal Cs		① Adsorption (Cation exchange)
Alkali earth metal Sr, Ra		① Adsorption (Cation exchange) ② Decrease of solubility (Hydrolysis)
Halogen I		① Decrease of solubility (Compound reaction) (with Ag/Cu) ② Sorption (Mechanism unknown)
Etc. C-14		① Decrease of solubility (Compound reaction) (Formation of calcium carbonate under cementaneous circumstance)

Chapter 3 Classification of TRU waste and provisional engineered barrier system.

As described in Chapter 1, TRU wastes generated in the reprocessing plant are classified in 20 kinds and those in MOX fuel manufacturing plant are in 3 kinds at present state. It is not plactical to set the engineered barrier system for each 23 wastes, therefore these wastes were re-categorized inclusively.

For the re-categorized wastes nuclide components and concentraions were estimated and the important nuclides were selected. Based on the results the essential barrier concept for each waste were discussed.

3.1 Re-categorization of TRU waste

For the re-categorization, nuclides included, waste materials, solidified materials, and homogenity were considered.

Nuclides included were divided into two types. The first type are plutoniums and their chain nuclides, the second type are fission products and activated products (FP/AP). The latter includes the iodine and iodine should be divided into the different type.

The waste materials are solid wastes of combustible, incombustible, and noncombustible materials and liquid wastes. The combustible and incombustible wastes are incinerated before isolation, therefore the materials can be assumed as inorganic. The noncombustible wastes are composed of metallic or inorganic materials. The liquid wastes are treated and the main components are inorganic salts. Therefore the main components of TRU wastes are inorganic or metallic materials.

Solidified materials are mainly asphalt, plastic, and cement. Asphalt and plastic are suitable for some kinds of liquid waste.

From the point of view of homogenity, solidified waste are divided into homogeneous waste and heterogeneous waste. At present, the difinition of homogenity is not technically desided. It is necessary to make experiments of

Table 3.1 Classification of TRU waste

No	category	waste	nuclide	solidified material	homogenity
1	Hull/end piece	ﾊﾙ・ｴﾝﾄﾞﾋﾟｰｽ	Pu, FP, AP	cement*3	unknown *4
2	Inorganic miscellaneous solid waste	β γ combustible β γ imcombustible TRU combustible TRU imcombustible jag chemical sludge resin silicagel sand activated carbon	Pu, FP, AP	cement*3	unknown *4
3	Metallic miscellaneous solid waste	β γ noncombustible TRU noncombustible active solid	Pu, FP, AP	cement*3	unknown *4
4	Solidified organic liquid waste	solvent dilution agent	Pu	plastic	homogeneous
5	Iodine adsorbent	iodine adsorbent	I	unknown *2	unknown *4
6	Solidified liquid waste (asphalt)	low level very low level (MOX liquid waste)	Pu, FP, AP	asphalt	homogeneous
7	Solidified *1 liquid waste (cement)	liquid wastes	Pu, FP, AP	cement	homogeneous
8	Inorganic MOX waste	MOX combustible MOX imcombustible	Pu	cement*3	unknown *4
9	Metallic MOX waste	MOX noncombustible	Pu	cement*3	unknown *4

*1 not generated at present
*2 not desided the suitable solidified material
*3 solidification by cement is assumed
*4 leachability rsearch is expecting

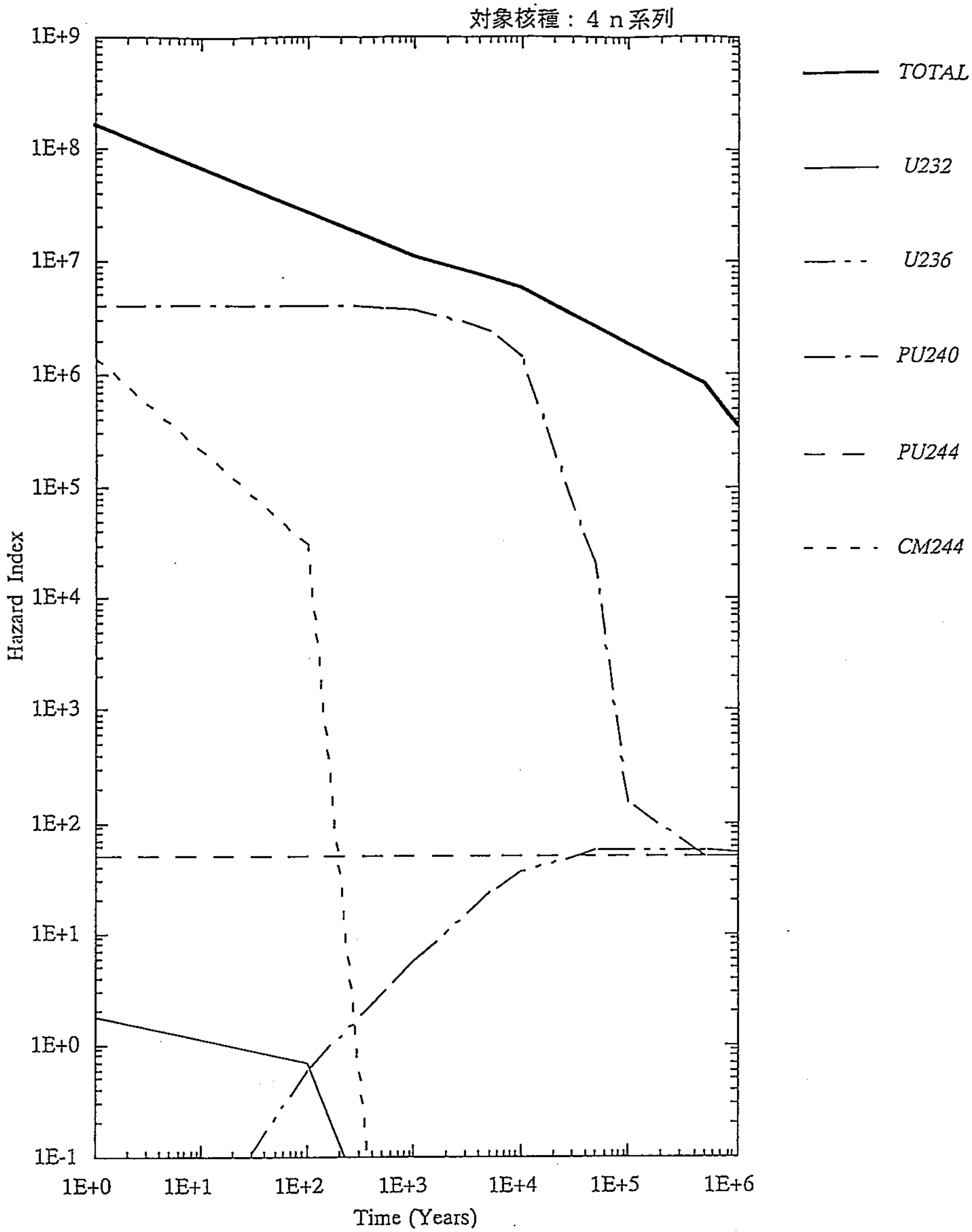


Fig. 3.1 Hazard index as a function of time in Hull/end piece waste

(4 n radioactive decay line)

Table 3.1 Classification of TRU waste

No	category	waste	nuclide	solidified material	homogenity
1	Hull/end piece	Hull/end piece	Pu, FP, AP	cement*3	unknown *4
2	Inorganic miscellaneous solid waste	$\beta\ \gamma$ combustible $\beta\ \gamma$ imcombustible TRU combustible TRU imcombustible jag chemical sludge resin silicagel sand activated carbon	Pu, FP, AP	cement*3	unknown *4
3	Metallic miscellaneous solid waste	$\beta\ \gamma$ noncombustible TRU noncombustible active solid	Pu, FP, AP	cement*3	unknown *4
4	Solidified organic liquid waste	solvent dilution agent	Pu	plastic	homogeneous
5	Iodine adsorbent	iodine adsorbent	I	unknown *2	unknown *4
6	Solidified liquid waste (asphalt)	low level very low level (MOX liquid waste)	Pu, FP, AP	asphalt	homogeneous
7	Solidified *1 liquid waste (cement)	liquid wastes	Pu, FP, AP	cement	homogeneous
8	Inorganic MOX waste	MOX combustible MOX imcombustible	Pu	cement*3	unknown *4
9	Metallic MOX waste	MOX noncombustible	Pu	cement*3	unknown *4

*1 not generated at present
*2 not desided the suitable solidified material
*3 solidification by cement is assumed
*4 leachability rsearch is expecting

Table 3.2 Critical nuclides in each TRU waste

Waste	Representative nuclide	Important nuclide calculated by radioactive decay *1 (Hazard index > 10^2 m³/MtU)		
		10^2～10^3 yr	10^3～10^5 yr	after 10^5 yr
Hull/end piece	U, TRU, FP, AP	^{241}Am ^{238}Pu ^{240}Pu ^{239}Pu ^{242}Pu ^{243}Am ^{244}Cm ^{237}Np ^{242m}Am ^{245}Cm ^{241}Pu ^{90}Sr ^{137}Cs ^{85}Kr ^{14}C	^{239}Pu ^{242}Pu ^{240}Pu ^{243}Am ^{237}Np ^{210}Pb ^{241}Am ^{245}Cm ^{227}Ac ^{231}Pa ^{226}Ra ^{14}C	^{242}Pu ^{210}Pb ^{229}Th ^{237}Np ^{210}Po ^{227}Ac ^{226}Ra ^{231}Pa ^{230}Th
Inorganic miscellaneous solid waste	U, TRU, FP, AP	^{241}Am ^{238}Pu ^{240}Pu ^{239}Pu ^{242}Pu ^{243}Am ^{244}Cm ^{90}Sr ^{137}Cs ^{85}Kr	^{239}Pu ^{242}Pu ^{240}Pu ^{243}Am	^{242}Pu
Metallic miscellaneous solid waste	U, TRU, FP, AP	^{241}Am ^{238}Pu ^{240}Pu ^{239}Pu ^{242}Pu ^{243}Am ^{90}Sr ^{137}Cs ^{85}Kr	^{239}Pu ^{242}Pu ^{240}Pu ^{243}Am	^{242}Pu
Solidified organic liquid waste	U, TRU	^{241}Am ^{238}Pu ^{240}Pu ^{239}Pu ^{242}Pu ^{243}Am ^{244}Cm	^{239}Pu ^{242}Pu ^{240}Pu ^{243}Am	^{242}Pu
Iodine adsorbent	I	^{129}I	^{129}I	^{129}I
Solidified liquid waste *2 (asphalt)	U, TRU, FP, AP	^{240}Pu ^{245}Cm ^{243}Am ^{242}Pu ^{241}Am ^{242m}Am ^{238}Pu ^{239}Pu ^{237}Np ^{236}U ^{241}Pu ^{234}U ^{90}Sr ^{137}Cs ^{129}I	^{240}Pu ^{242}Pu ^{239}Pu ^{241}Am ^{245}Cm ^{243}Am ^{210}Pb ^{226}Ra ^{237}Np ^{230}Th ^{227}Ac ^{231}Pa ^{236}U ^{234}U ^{129}I	^{210}Pb ^{226}Ra ^{227}Ac ^{231}Pa ^{242}Pu ^{230}Th ^{229}Th ^{237}Np ^{236}U ^{129}I
Solidified liquid waste *2 (cement)	U, TRU, FP, AP	^{240}Pu ^{245}Cm ^{243}Am ^{242}Pu ^{241}Am ^{242m}Am ^{238}Pu ^{239}Pu ^{237}Np ^{236}U ^{241}Pu ^{234}U ^{90}Sr ^{137}Cs ^{129}I	^{240}Pu ^{242}Pu ^{239}Pu ^{241}Am ^{245}Cm ^{243}Am ^{210}Pb ^{226}Ra ^{237}Np ^{230}Th ^{227}Ac ^{231}Pa ^{236}U ^{234}U ^{129}I	^{210}Pb ^{226}Ra ^{227}Ac ^{231}Pa ^{242}Pu ^{230}Th ^{229}Th ^{237}Np ^{236}U ^{129}I
Inorganic MOX waste	U, TRU	^{241}Am ^{238}Pu ^{240}Pu ^{239}Pu ^{241}Pu ^{242}Pu ^{237}Np	^{239}Pu ^{240}Pu ^{242}Pu ^{237}Np ^{210}Pb ^{226}Ra ^{229}Th ^{230}Th ^{236}U	^{229}Th ^{237}Np ^{242}Pu ^{210}Pb ^{227}Ac ^{231}Pa ^{226}Ra ^{230}Th ^{236}U
Metallic MOX waste	U, TRU	^{241}Am ^{238}Pu ^{240}Pu ^{239}Pu ^{241}Pu ^{242}Pu ^{237}Np ^{234}U ^{238}U	^{239}Pu ^{240}Pu ^{242}Pu ^{237}Np ^{210}Pb ^{226}Ra ^{229}Th ^{230}Th ^{236}U ^{227}Ac ^{234}U ^{231}Pa ^{235}U ^{241}Am ^{238}U	^{229}Th ^{237}Np ^{242}Pu ^{210}Pb ^{227}Ac ^{231}Pa ^{226}Ra ^{230}Th ^{236}U ^{223}Ra ^{233}U ^{235}U ^{234}U ^{238}U

*1 Criteria of selection; Hazard Index≧100, Radioactive half life≧1y
*2 Same inventory

leachability.

Based on the above discussion, TRU wastes were re-categorized into nine kinds shown in Table 3.1. The following discussion are based on this categorization.

3.2 Selection of inportant nuclide

The estimation of nuclide components and concentrations in each solidified waste was carried out. The index used was hazard index (HI)described as the following equation.

$$HI = \frac{\text{Nuclide concentration in solidified waste} \quad (Bq/m^3)}{\text{Perrmitted nuclide concentration in water} \quad (Bq/m^3)} \quad \cdots 3.1$$

The time dependance of HI was calculated by ORIGEN 2 then radioactive decay was considered. Calculation results were exhibited as five figures of 4n, 4n+1, 4n+2, 4n+3 series and FP/AP. Fig. 3.1 shows the results for 4n series of Hull/end piece. A provisional criteria of that HI is higher than 100 and radioactive half life is higher than 1 year were used to select the important nuclide. Table 3.2 shows the selected nuclides of each TRU waste. After 10^5 years passed the important nuclides are only TRUs and their decay chain nuclides except for I-129. I-129 is found in the iodine adsorbent and the solidified liquid waste.

3.3 Discussion with engineered barrier system of each TRU waste

The function of the engineered barrier was arranged and the suitable barrier system for each waste was discussed.

The main functions of engineered barriers were categorized in the following three items; hydrological barrier, chemical barrier, and physical barrier. hydrological barrier and physical barrier are necessary for the all types of radioactive waste isolation. Chemical barrier, however, reflects the nuclide behaviors contained in the TRU wastes. The functions of chemical barrier are precipitaion to decrease solubility llimit and adsorption to decrease migration velocity. In the former case, one of the useful method is to use materials with high ion exchange capacity like as bentonite. In the latter case, one is to use cementaneous materials to make hydration products of TRU and/or FP/AP nuclides. Therefore the essential components of chemical barrier were considered as hybrid usage of cementaneous materials and bentonite. Cementaneous solidified materials work as chemial barrier. Table 3.3 shows the essential components of engineered barrier system for each TRU waste.

Table 3.3 Esscential engineered barrier system for each waste

No.	Waste	Main Nuclide	Solidified material	Hydrological barrier	Chemical barrier
1	Hull/end piece	Pu,FP,AP	Cement	Bentonite	Cement*
2	Inorganic miscellaneous solid waste	Pu,FP,AP	Cement	Bentonite	Cement*
3	Metallic miscellaneous solid waste	Pu,FP,AP	Cement	Bentonite	Cement*
4	Solidified organic liquid waste	Pu	Plastic	Bentonite	Cement
5	Iodine absorbent	I	Cement	Bentonite	Cement*
6	Solidified liquid waste (asphalt)	Pu,FP,AP	Asphalt	Bentonite	Cement
7	Solidified liquid waste (cement)	Pu,FP,AP	Cement	Bentonite	Cement*
8	Inorganic MOX waste	Pu	Cement	Bentonite	Cement*
9	Metallic MOX waste	Pu	Cement	Bentonite	Cement*

* Function by solidified material

Chapter 4 Comparison with safety performance of engineered barieer systems.

The relative safety assessment was carried out on the engineered barrier system of each TRU waste. Furthermore bentonite alternation from sodium type to calcium type was also calculated.

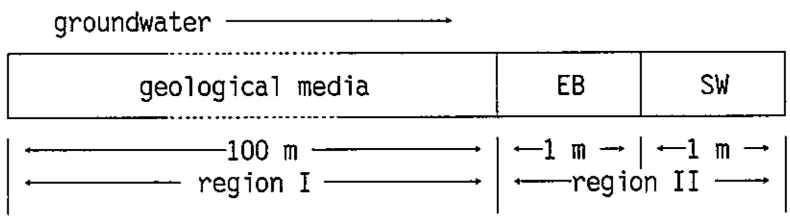
4.1 Relative safety assessment of engineered barrier system

4.1.1 Safety assessment model

The calculations were carried out under one dimensional system. Index used was flux of HI at the outer surface of engineered barrier. Code was FRONT which calculate one dimensional nuclide migration with radioactive decay chain.

The outline of calculation system is shown in Fig. 4.1. The calculated point was the upper stream side of engineered barrier. The upper stream side has the highest concentration slope. The length of geological media is 100 m and ground water velocity is 1 m/y.

The zone inside of engineered barrier is diffusion controlled zone because outter part of engineered barrier is bentonite. The total length of engineered barrier or solidified waste is fixed as 1 m to neglect the influence of barrier length on calculation results.



EB; Engineered barrier
SW; Solidified waste
region I ; water flow controlled zone
region II; diffusion controlled zone

Fig. 4.1 System for relative safety assessment

The boundary condition at the edge of upstream and downstream is $\partial c/\partial x=0$, that at boundary between geological media and engineered barrier or between engineered barrier and waste form is conservation of flux.

The model which describe the source term is the hybrid model of release coeficient and solubility limit. At first nuclides in the waste materials are released into the pore of solidified waste. If the concentration in the pore is higher than the solubility limit, the concentration is limitted at solubility limit. The solubility limit model should be carefully used when the isotope of a element is not unity. In the model used here the isotopic ratio is simplly assumed that the initial ratio is conserved.

4.1.2 Parameter for calculation

Table 4.1 shows the parameters of media. Table 4.2 shows distribution coefficients of nuclide onto media. Table 4.3 shows thr release coefficients calculated by distribution coefficients, density, and porosity. These values were extracted from technical papers. The data with no moderate paper were assumed by means of chemical analogy.

The data of solubility limit were calculated by PHREEQE under the alkaline atomosphere by cementaneous materials and reductive atomosphere by the corrosion of iron container. The database used was PHREEQE original database for minerals and OECD/NEA database for TRU nuclides. The chemical composition of groundwater was undesidable, therefore the pure water was assumed. Base on the PHREEQE calculation results, the following chemical behaviors were identified; Carbon and iodine have no solid phase which limit the solubility limit. Strontium concentration is much lower than solubility limit. Curium and uranium have lower solubility under oxidation atomosphere. Plutonium and neptunium have lower solubility under reduction atomosphere. Americium, protactinium, thorium, and lead are insensitive to redox condition. The starting time of nuclide release from waste materials assumed just after the isolation.

Table 4.1 Parameters of media for calculation

media	effective diffusivity (m^2/s)	porosity (−)	density (kg/m^3)
bentonite barrier	1.00E-10	0.33	2.7E+3
concrete barrier	1.00E-10	0.5	2.7E+3
concrete waste form	1.00E-10	0.5	2.7E+3
asphalt waste form	1.00E-9	0.33	1.0E+3
plastic waste form	1.00E-9	0.33	1.0E+3
geological media	1.00E-9	0.33	2.7E+3

Table 4.2 Distribution coefficients for calculation

element	bentonite	concrete	asphalt	plastic	geological media
Sr	1.09E-2	5.00E-3	6.57E-2	6.57E-2	5.00E-3
I	9.23E-4	5.00E-3	1.90E-2	1.90E-2	0.00E+0
Pb	* 5.53E-1	* 5.00E-1	* 6.67E+0	6.67E+0	1.00E-2
Ra	* 1.09E-2	* 5.00E-3	* 6.57E-2	6.57E-2	5.00E-3
Th	* 5.53E-1	* 5.00E-1	* 6.67E+0	6.67E+0	1.00E-1
Pa	* 5.53E-1	* 5.00E-1	* 6.67E+0	6.67E+0	1.00E-2
U	* 5.53E-1	* 5.00E-1	* 6.67E+0	6.67E+0	1.00E-2
Np	5.53E-1	* 5.00E-1	6.67E+0	6.67E+0	1.00E-2
Pu	* 1.00E+0	1.00E+0	6.67E+0	6.67E+0	1.00E-1
Am	* 1.00E+0	5.00E-1	6.67E+0	6.67E+0	1.00E-1
Cm	* 5.53E-1	5.00E-1	* 6.67E+0	6.67E+0	1.00E-2
data source	PNC data	TJL	IAEA/TECDOC 401	same as asphalt	Kato Shouhei

* ; assumed value with chemical analogy

Table 4.3 Release coefficients for calculation

element	concrete	asphalt	plastic
Sr	1.38E-1	3.00E-2	3.00E-2
I	1.38E-1	1.00E-1	1.00E-1
Pb	1.48E-3	3.00E-4	3.00E-4
Ra	1.38E-1	3.00E-2	3.00E-2
Th	1.48E-3	3.00E-4	3.00E-4
Pa	1.48E-3	3.00E-4	3.00E-4
U	1.48E-3	3.00E-4	3.00E-4
Np	1.48E-3	3.00E-4	3.00E-4
Pu	7.40E-4	3.00E-4	3.00E-4
Am	1.48E-3	3.00E-4	3.00E-4
Cm	1.48E-3	3.00E-4	3.00E-4

4.1.3 Results

At first the calculation was carried out for Hull/end piece because their inventory is the basic case of TRU waste. Fig. 4.3 (1)~(5) and Fig. 4.4 (1)~(5) show the nuclide release flux as a function of time at oxidation condition and reduction condition respectively.

The flux of FP/AP decreases linearly after the peak point appered because they have no radioactive decay chain. The couse of Sr-90 decrease is radioactive disintegration, that of I-129 is inventory decrease by release, and that of C-14 is composite of redioactive disintegration and inventory decrease. In case of TRU nuclides and their chain nuclide, the flux increases with time and it reachs the constant value.

The same calculations were carried out for the other wastes. Table 4.5 shows the nuclide with the highest peak and the order of peak value. Except for the iodine adsorbent the highest flux was obtained for 4n+2 series and the nuclides were Pu-242 or Ra-226.

The factors affecting to the nuclide release were considered. To control the Pu-242 solubility or Th-230 release rate is effective to decrease the highest flux peak.

Table 4.5 Nuclide with the highest release flux on each solidified waste and its order

waste form	oxidation	reduction
Hull/end piece	Pu-242 ④	Ra-226 ⑥
Inorganic miscellaneous solid waste	Pu-242 ⑦	Ra-226 ⑦
Metallic miscellaneous solid waste	Pu-242 ⑨	Ra-226 ⑨
Solidified organic liquid waste	Pu-242 ⑧	Ra-226 ⑧
Iodine absorbent	I -129 ①	I -129 ①
Solidified liquid waste (asphalt)	Ra-226 ⑥	Ra-226 ⑤
Solidified liquid waste (cement)	Ra-226 ⑤	Ra-226 ④
Inorganic MOX waste	Ra-226 ②	Ra-226 ②
Metallic MOX waste	Ra-226 ②	Ra-226 ②

4.2 Calculation of bentonite alternation from Na type to Ca type

When bentonite and cementaneous materials are used at the same time in the engineered barrier system, Na-bentonite will be altered to Ca-bentonite and the properties of bentonite will change. Calculation was carried out for the alternation of bentonite with precipitation, ion exchange, and migration of chemical components in the engineered barrier system. The results show that the Ca dominant bentonite will appear within several hundred years. Many of parameters used in the calculation were, however, uncertainly therefore the future research are necessary.

Chapter 5 Extraction of research theme typical to TRU waste.

Many of research theme were extracted from the discussion from Chapter 1 to Chapter 4. Those are arranged as hierarchical item structure as shown in Fig. 5. 1.

- engineered barrier system
  - migration through engineered barrier system
    - nuclide release from waste form
      - data acquisition
        - inventory (component concentration)
          - PNC waste
            - transfer ratio
              - variation by operation processes
              - variation by operation cycles
          - the others (ex. return waste)
        - nuclide release baehaviors
          - release rate
            - physical properties
            - chemical properties
            - apparent diffusivity
              - effective diffusivity
              - solubility
              - adsorption
              - pore structure
            - degradation
              - reaction with groundwater
              - microoragnism
              - radiolysis
              - corrosion of waste
              - compatibility of cement and bentonite
            - gas generation
              - microoragnism
              - radiolysis
              - corrosion of waste
            - homogenity
      - research of safety assessment
    - migration through engineered barrier system (inside buffer and/or structural materials)
      - data acquisition
        - physical properties
        - chemical properties
        - apparent diffusivity
          - effective diffusivity
          - solubility
          - adsorption
          - pore structure
        - degradation
          - reaction with groundwater
          - microoragnism
          - radiolysis
          - corrosion of waste
          - compatibility of cement and bentonite
        - gas generation
          - microoragnism
          - radiolysis
          - corrosion of waste
        - colloid
          - true colloid
          - pseudo colloid
        - homogenity
      - research of sfety assessment
  - research of natural barrier
    - data acquisition
      - hydrology
      - physical property
        - rock
      - chemical property
        - rock
        - ground water
    - model research
  - isolation ability
  - execution ability
  - economical efficiency

Table 5.1 Items about research of designing TRU waste isolation system