STUDY ON SAFETY OF TRU-CONTAINING WASTE DISPOSAL IN JAPAN

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ABSTRACT

This paper describes the present status of TRU-containing waste disposal in Japan from the safety standpoint. Because of large amounts of such waste generated and the variety of its characteristics, TRU-containing waste should be classified into categories according to radioactivity and disposed of within different types of disposal concepts. For the time being, three disposal alternatives have been proposed in Japan. They are shallow land disposal within concrete pits, mid-depth disposal and deep geological disposal. Focusing upon the deep geological disposal of some TRU-containing waste, radionuclides migration analysis and dose evaluation have been conducted under base scenarios and perturbation scenarios. It has been found that safety for geological disposal of TRU-containing waste could be ensured under assumed hypothetical conditions.

INTRODUCTION

Radioactive wastes containing transuranic elements, generated mainly from several types of fuel cycle facilities, such as reprocessing and MOX fuel fabrication (hereinafter referred to as TRU-containing waste), are now regarded as one of key issues to be carefully considered within the context of radioactive waste management in Japan.

With regard to regulatory concerns, the Advisory Committee on Nuclear Fuel Cycle Backend Policy, a subsidiary of the Atomic Energy Commission of Japan (AEC), prepared recommendations titled *"Fundamental Considerations on Conditioning and Disposal of Radioactive Wastes Containing Transuranic Elements"* [1]. In order to support these recommendations, the Japan Nuclear Cycle Development Institute (JNC) and electricity utilities jointly published a comprehensive technical report in March 2000, called TRU report [2], which highlighted and demonstrated alternative design concepts for their geological disposal. In the above report, it was concluded that TRU-containing waste can be classified and disposed of within different kinds of facilities, and most of it is ready to be disposed of in shallow land repositories.

In this study, based on disposal concepts envisaged in the TRU report, the present status of TRU-containing waste management is discussed from the viewpoint of safety.

DEFINITION OF "TRU-CONTAINING WASTE"

Terminologically, "TRU-containing waste", used in Japan, is similar to "long-lived intermediate level wastes", which is in wider use in the world. "TRU-containing waste" discussed here is defined as follows.

Many kinds of radioactive wastes are generated during operation and decommissioning of several facilities on nuclear fuel cycle. These are precisely "TRU-containing waste" which consist of the following kinds of radioactive wastes.

- Waste generated from reprocessing plants during their operation, except for vitrified HLW
- Waste generated from reprocessing plants during their decommissioning
- Returned waste from COGEMA, except for vitrified HLW
- Waste generated from MOX fabrication plant during its operation
- Waste generated from MOX fabrication plant during its decommissioning

It is obvious that the concept of TRU-containing waste covers a wide range of radioactive wastes; therefore, characteristics of the wastes are quite different from one another. Radioactivity varies in several orders of magnitudes. Some are solidified by cement, others by bitumen. Some generate heat while others do not. Some contain certain chemical agents, such as sodium nitrate and organic materials, which might enhance nuclide migration because of their chemical activities in potential. Moreover, it should be pointed out that amount of TRU-containing waste is estimated very larger, as compared with that of vitrified HLW.

CLASSIFICATION OF TRU-CONTAINING WASTE FROM VIEWPOINT OF WASTE MANAGEMENT

Alternative Disposal Concepts for TRU-containing Waste

The fact that TRU-containing waste has a large variety of characteristic allows us to classify it into several types, which are to be disposed of according to different types of repository. For the time being, the following three concepts of disposal are proposed for TRU-containing waste:

- ☐ Shallow Land Disposal (a few meters deep)
- ☐ Mid-Depth Disposal (several tens of meters deep)
- ☐ Deep Geological Disposal (hundreds of meters deep)

The repository of Shallow Land Disposal is composed of concrete pits surrounded by bentonite-sand mixture, constructed several meters below the earth's surface. Although the repository of Deep Geological Disposal is now under investigation, the concept is envisaged as engineered barriers made of cement and/or bentonite-sand mixture hundreds of meters underground the largest part of TRU-containing waste is not significantly exothermic, so that large cavities could be applied for repository for Deep Geological Disposal.

Of the above concepts, the concept of Mid-Depth Disposal is the newest and the basic design is under way. The repository will be constructed from 50 to 100 meters underground. According to the present study, many types of TRU-containing waste could be disposed of in the repository.

Classification of TRU-containing Waste

In classifying TRU-containing waste into three disposal concepts, two kinds of regulatory standards are available at the moment. One is a tentative guideline regarding the total alpha concentration recommended by the AEC [3] and the other is the upper limit of some important nuclides for Shallow Land Disposal [4].

In 1994, the AEC published "Long Range Programme for Research, Development and Exploitation of Nuclear Energy", in which it recommended that TRU-containing waste should be classified according to the tentatively proposed guideline of 1 GBq/ton of total alpha concentration. The AEC published "Basic Criteria Regarding

Safety Regulation for Land Disposal of Solidified Low Level Radioactive Waste" in 1992, in which it determined the upper limit of concentration of relevant nuclides in waste package to be disposed of in concrete pits for shallow land disposal.

In this study, these two regulatory standards are applied for the classification of TRU-containing waste into three types of disposal concept. It should be noted, however, that much attention must be paid to these two standards when applying them. As the AEC commented, a guideline of 1 GBq/ton is a tentative indicator to be further discussed before finalization. There was not yet a concept of Mid-Depth Disposal, when the AEC offered the guideline of 1 GBq/ton. The upper limits for Shallow Land Disposal are applied to the solidified low-level waste from nuclear power stations. Some new upper limits of radionuclides should be required for TRU-containing waste, because those from nuclear fuel cycle facilities have quite a different composition of radionuclides from those of nuclear power station

Fig.1 shows a flowchart of how to classify TRU-containing waste in this study. A tentative guideline of total alpha and the upper limits of nuclide concentration are used as the first step to classification. Then, preliminary safety assessments are carried out for Shallow Land Disposal and Mid-Depth Disposal, respectively. The dose rate for Mid-Depth Disposal fully satisfies the safety guideline of 0.01 mSv/y for all cases. Some types of TRU-containing waste may feasibly be disposed of in a Shallow Land Disposal. The safety assessment for Deep Geological Disposal is described in the latter half of this paper.

Tentative Guideline of Total Alpha Concentration

As already indicated, the guideline of 1 GBq/ton of total alpha concentration recommended by the AEC is a tentative upper limit between Shallow Land Disposal and Deep Geological Disposal. A value higher than 1 GBq/ton is expected to apply for a new upper limit of Mid-Depth Disposal.

Table I shows disposal volumes of TRU-containing waste when A guideline of 1 GBq/ton of total alpha concentration is changed to 5 GBq/ton, 10 GBq/ton and 50 GBq/ton, respectively. It is clear that balance of waste volumes between Mid-Depth Disposal and Deep Geological Disposal can vary accordingly as the guideline of total alpha concentration changes. It suggests that the guideline of total alpha concentration will become one of the most important factors in optimising efficiency and economy for management of TRU-containing waste.

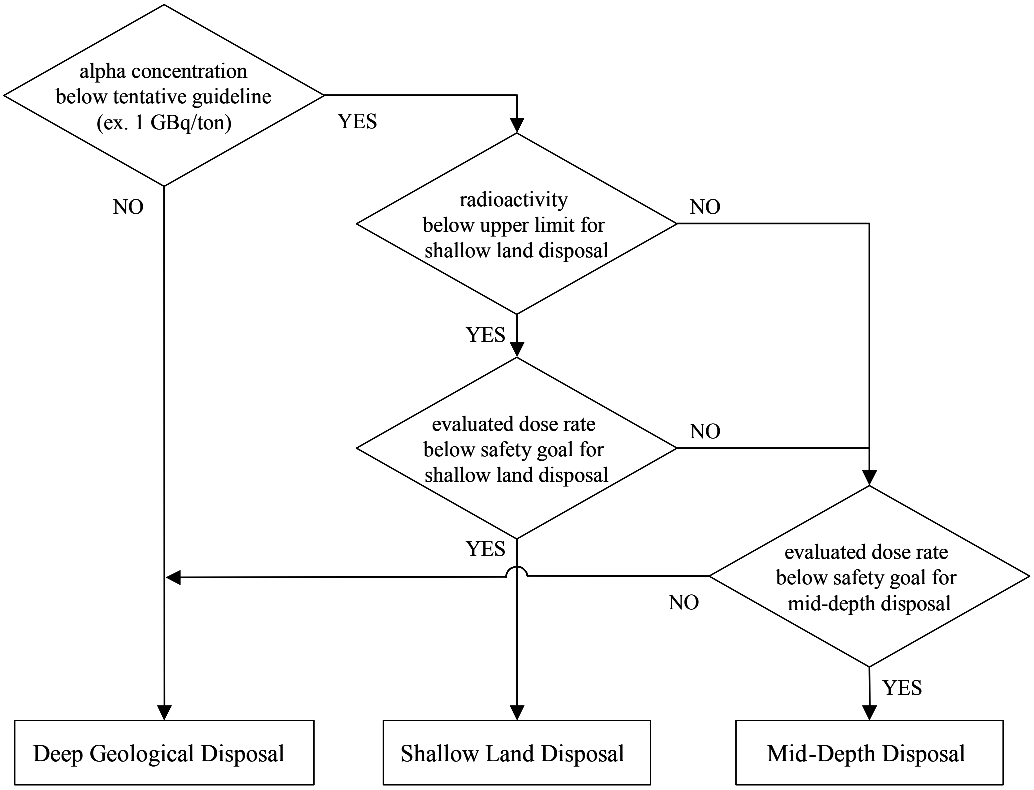


Fig.1 Flowchart on classification of TRU -containing waste

Table I. Allocation of TRU-containing Waste with Varied Guideline for Classification

	Guideline of total alpha concentration			
	1 GBq/ton	5 GBq/ton	10 GBq/ton	50 GBq/ton
Shallow Land Disposal	13,000 m^3	13,000 m^3	13,000 m^3	13,000 m^3
Mid-Depth Disposal	25,000 m^3	28,500 m^3	32,000 m^3	35,000 m^3
Deep Geological Disposal	18,000 m^3	14,500 m^3	11,000 m^3	8,000 m^3
Total [a]	56,000 m^3	56,000 m^3	56,000 m^3	56,000 m^3

[a] waste generated up to 2035

SAFETY ASSESSMENT FOR DEEP GEOLOGICAL DISPOSAL OF TRU-CONTAINING WASTE

After classification and allocation of TRU-containing wastes, the preliminary safety assessment of Deep Geological Disposal of some these is discussed here to confirm its feasibility. As described above the amount of TRU-containing waste to be subjected to Deep Geological Disposal depends upon the total alpha guideline. Assuming a total alpha concentration of 50 GBq/ton as a guideline here, of the total volume of 56,000 m^3 up to the year 2035, the waste volume of 8,000 m^3 is estimated as the volume to be disposed of in a deep geological repository.

TRU-containing Waste for Deep Geological Disposal

TRU-containing waste subject to Deep Geological Disposal still has a wide range of properties and it is proposed, therefore, that it be sub-classified further into four groups, based upon their characteristics, such as heat generation, inventory of key nuclides (^{129}I and ^{14}C) and contents of deteriorates (organic materials and sodium nitrate), as defined in Table II.

Table II. Grouping of TRU-containing Waste for Deep Geological Disposal

	Typical Waste	Characteristics
Group 1	spent iodine filter (cement solidification)	large content of ^{129}I, one of the key nuclides
Group 2	hulls and ends (compacted)	relatively high heat generation large content of ^{14}C, one of the key nuclides
Group 3	process concentrated liquid (bituminised or cemented)	large content of asphalt and sodium nitrate which may affect water chemistry and/or nuclide migration
Group 4	all others	relatively low activity

Basically, the above Groups are different from one another in the design, size and pitch of the disposal unit for emplacing each of the Groups of waste. Nevertheless, for all of the groups, cementituous materials are used as fillers in the voids between waste packages and structural materials. Installing bentonite buffer materials outside should enhance the containment function of the engineered barrier system for Group 1 and Group 2. The disposal unit for Group 3 should be located the furthest downstream from groundwater flow in order to prevent any effects of deteriorates discharged from Group 3 on water chemistry and/or nuclide migration of the other groups.

Modelling

The migration of nuclides released from waste form, through the engineered barrier system and the geosphere to the biosphere, is numerically analysed. One-dimensional advection-dispersion equations are solved with a combination of two mathematical schemes, a finite differential method for the engineered barriers and an analytical solution using the Talbot method [5] for the natural barriers. Optionally, however, the same finite differential method can be chosen for the natural barrier as well. The code used here can handle time-dependent parameters both in the engineered barrier system and for natural barrier, such as groundwater velocity, solubility, sorption coefficient and diffusion coefficient. In order to reflect the evolution of the multiple barriers system appropriately, it is particularly important for the repository of TRU-containing waste, for example, cement alteration, etc. Details of the model are available from the literature [2].

There is no candidate site for Deep Geological Disposal in Japan yet. Therefore, the natural barrier should be considered as a rather generic condition in safety assessment. Although many kinds of geological formation are available in the Japanese geological context, two types of rock have been chosen for representing sites, one is sedimentary rock, such as clay, and the other is crystalline rock, such as granite. From the modelling point of view, sedimentary rock is simulated as porous media and crystalline rock as parallel plains of permeable fracture.

Scenario Development for Base Scenarios

Prior to carrying out analysis of nuclides migration, scenario analysis has been conducted to identify what should be analysed for Deep Geological Disposal of TRU containing waste in the case of base scenarios, which can be regarded as "normal evolution scenarios". First, important features, events and processes (FEPs) are listed, and then their potential combinations are integrated. Finally, their relative importance is comprehensively interpreted and is implemented by the same Systematic Approach Method introduced by SKI [6]. For details of methodology in presenting base scenarios, reference should be made to the literature [2].

As a result, following preconditions should be addressed firstly regarding the natural barrier.

☐ The characteristics of the natural barrier do not change with time, included in these characteristics is the distance between repository and the prevailing groundwater flow, also the disposal environment is chemically reducing.
☐ Volcanic activity is ruled out at the site selection stage.
☐ A low groundwater flow rate, as expected from selected Japanese geological repository environments, is assumed.

Secondly, the following phenomena should be regarded as key issues for TRU-containing waste disposal in potential, based upon the scenario development mentioned above.

☐ Chemical and hydraulic evolution of cementituous materials due to cement alteration
☐ Cation exchange of bentonite from Na rich type to Ca rich type
☐ Expulsion of contaminated pore water due to gas generation
☐ Effects of nitrate on sorption and precipitation
☐ Alteration of bentonite and rock by hyper alkaline water of cement

Finally, in consideration of the above phenomena, the following calculation cases have been set up.

☐ REFERENCE CASE
Including cement alteration and bentonite calcification using time independent parameters with a conservative setting.
☐ GAS CASE
Simulating continuous expulsion of contaminated pore water by accumulated gas pressure using time dependent parameters of the engineered barriers system. There are two variations, depending on whether retardation by sorption or precipitation during expulsion is ensured or not.
☐ NITRATE CASE
A scoping calculation assuming the effects of nitrate will propagate not only within the engineered barrier system but also to the natural barrier. Conservative values for sorption coefficients and solubility limits are chosen both in the engineered barrier system and in the natural barrier.
☐ HYPER ALKALINE CASE
Assuming mineralogical alteration of bentonite other than calcification and alteration of rock as well as CSH formation due to hyper alkaline cement pore water for long periods. Conservative values are chosen for permeability of bentonite, sorption coefficients and solubility limits both in the engineered barrier system and in the natural barrier system.

Results of Base Scenarios

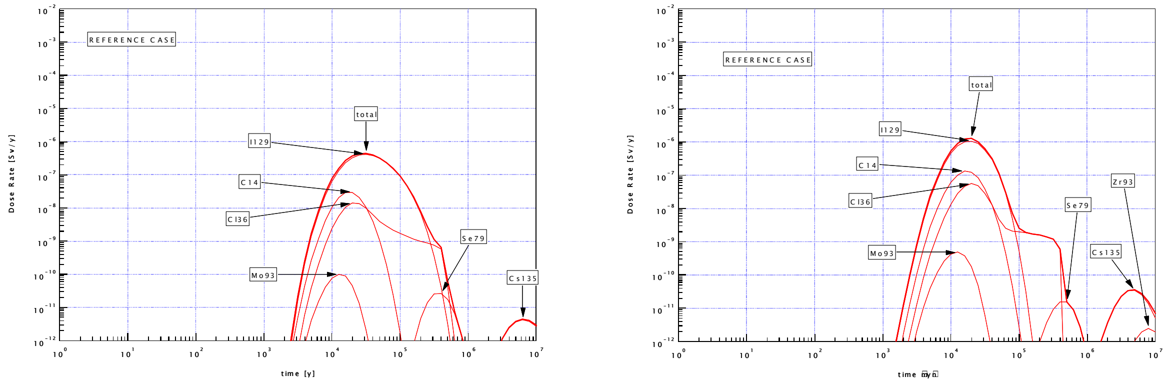
Fig.2 (a) and (b) show profiles of the calculated dose rate with the time for the REFERENCE CASE for sedimentary rock and crystalline rock, respectively. ^{129}I, contained mostly in group 1, is the dominant nuclide because it has a very long life (half life : 15.7 million years) and is less sorptive and highly soluble. ^{14}C, much involved in the hulls and ends of group 2, is the second most significant contribution, one order of magnitude less than ^{129}I. Although it is not clearly shown in the figure, other scoping calculations varying hydrological conditions in a natural barrier suggest that ^{14}C is rather sensitive to migration time due to its half life of 5,730 years, which may induce decay during migration up to the biosphere. Contrarily and paradoxically, actinides, typical nuclides which characterize "TRU-containing waste", do not show any significance on the dose rate profile because they tend to adsorb or precipitate under a reducing environment and do not have much inventories in waste originally, compared to vitrified HLW.

Profiles of calculated dose rates for each case defined as base scenarios are shown in Fig 3 (a), (b).

Regarding the GAS CASE, there is variation whether sorption and precipitation in the engineered barrier system is established or not during expulsion of contaminated pore water by accumulated gas pressure. Obviously, however, the difference is not significant and both cases are very similar to the REFERENCE CASE.

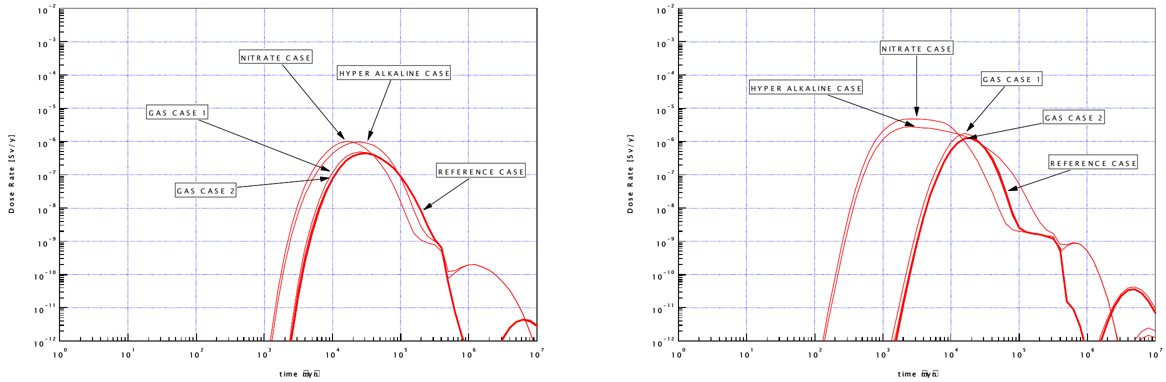
Contrary to the GAS CASE, both the NITRATE CASE and the HYPER ALKALINE CASE give several times higher maximum dose rate than the REFERENCE CASE. This is because more conservative parameters for natural barriers are adopted in these calculation cases.

Nevertheless, it should be pointed out that calculated maximum dose rates do not exceed 0.1-0.3 mSv/y, which is the safety standard used in foreign countries, even if a hypothetically conservative case such as the NITRATE CASE is taken into account. For the time being, there is no regulatory safety criterion in Japan for Deep Geological Disposal of TRU-containing waste. But it would be possible to conclude at least that Deep Geological Disposal of TRU-containing waste is feasible from the viewpoint of safety as far as assuming base scenarios, namely normal evolution scenarios.



(a) Sedimentary Rock (b) Crystalline Rock

Fig.2 Contributions of Key Nuclides on Dose Rate Profile



(a) Sedimentary Rock (b) Crystalline Rock

GAS CASE 1 : sorption and precipitation are not established during expulsion
GAS CASE 2 : sorption and precipitation are established during expulsion

Fig.3 Calculated Dose Rate Profile for Each Cases Defined as Base Scenarios

Setting of Perturbation Scenarios

Perturbation scenarios are characterized by natural phenomena, initial defects in installing engineered components of repository and future human activities. Contrary to the above base scenarios, probabilities of events of the perturbation scenarios vary with site selection and level of quality control for geological disposal.

For the perturbation scenarios, almost the same sets of scenario identification are employed, as demonstrated by JNC in the second progress report on the research and development for the geological disposal of HLW in Japan [7]. The internal process of the repository is quite different between vitrified HLW and TRU-containing waste, but the external events that initiate perturbation scenarios are the same.

According to the second progress report, the perturbation scenarios are as follows :

☐ UPLIFT AND EROSION
It is assumed that the depth of the repository gradually decreases as a result of continued uplift and erosion. Moreover, possible impact on flow around the repository and the effects of oxidizing surface water should be duly considered once the repository approaches the ground surface.

☐ CLIMATIC AND SEA-LEVEL CHANGES
It is assumed that the annual average ambient temperature will continues to fall gradually and reach its lowest level in approximately one hundred thousand years due to glaciations. Accordingly, sea level is assumed to continue falling and the saline groundwater in coastal areas may turn fresh due to movements of the interface between fresh groundwater owing to changes in the GBI (Geosphere Biosphere Interface).

☐ POOR BACKFILLING OF TUNNELS AND DEFECTS IN PLUGS
Incomplete sealing of the emplacement tunnels and access tunnel due to poor backfilling and defects in plugs are assumed to result in relatively fast radionuclide transport pathways along these tunnels.

☐ DRILLING OF A WELL AND WATER EXTRACTION
Drilling of a deep well near the repository is assumed. Nuclides released from the repository may go directly into the biosphere. The biosphere model should be modified for well water use.

☐ BORING
It is assumed that boring shafts used for geophysical prospecting, etc., might accidentally reach the area of the repository. The near field environment of a repository would shift from being reduced to being oxidized due to the introduction of air. Moreover, it is possible that the boring shaft itself would become a migration pathway.

Table III shows details of the evaluation of the perturbation scenarios. The footnotes indicate differences in assumptions between the cases of TRU-containing waste and vitrified HLW.

Table III. Conditions and Assumptions for Perturbation Scenarios

Perturbation Scenarios		Remarks
Natural Phenomena	Uplift and Erosion Evolution of ground water flow and water chemistry	☐ Uplift rate = erosion rate : 0.1mm/y, 1mm/y ☐ No retarded migration through fault ☐ Oxidized groundwater less than 100m depth (varied diffusion coefficient, solubility limit and sorption coefficient) ☐ Flow rate 10 times larger less than 100m depth ☐ Exposure to the surface is not accounted for
	Climatic / Sea Level Change Periodic evolution of water chemistry	☐ Saline water composition at initial ☐ No retarded migration through fault ☐ Periodic alternation between saline water and fresh water (varied diffusion coefficient, solubility limit and sorption coefficient) ☐ Biosphere model accounting for sea water use ☐ Corrosion rate 10 times faster (hulls and ends) [a]
Initial Defects of Engineered components	Failure of Sealing	☐ One gallery [b] ☐ Flow rate at EDZ 10 times larger ☐ Access shaft regarded as migration path [c] (100m long, flow velocity 10 times faster than rock, sorption the same as rock)
Human Activity in Future	Deep Well	☐ Biosphere model accounting for well water use
	Boring Oxidized groundwater	☐ One gallery near boring [d] ☐ Oxidized groundwater in the engineered barriers system (varied diffusion coefficient, solubility limit and sorption coefficient) ☐ Corrosion rate 3.3 times faster (hulls and ends) [a]
	Boring Change of migration pathway	☐ One gallery near boring [d] ☐ Event 1,000y after closure of repository [e] ☐ Flow rate at EDZ 10 times larger than REFERENCE CASE ☐ Simultaneously discharged to biosphere through EDZ

[a] not relevant to vitrified HLW

[b] 200 packages of vitrified HLW

[c] simultaneously discharged to biosphere through EDZ

[d] 4 packages of vitrified HLW

[e] event soon after closure of repository

Results of Perturbation Scenarios

Fig.4 (a) and (b) show the calculated maximum dose rate for the perturbation scenarios and also the results of REFERENCE CASE in base scenarios for comparison purposes.

In the case of uplift and erosion, a new tendency of dose contribution of key nuclides is observed. ^{99}Tc and ^{237}Np, which are not significant in the REFERENCE CASE, provide a comparable dose peak to those of ^{129}I, the dominant nuclide in base scenarios. Technetium and neptunium are the redox sensitive elements and their valences are assumed to shift from reduction to oxidization as Tc(IV) -> Tc(VII), Np(IV) -> Np(V), respectively. They are well confined within a multi-barriers system in reducing environment, but, once an oxidizing environment is established, they will suffer chemical form change to become more mobile. However, the maximum dose rate in the case of uplift and erosion is practically at the same level as that in the REFERENCE CASE as shown in Fig-4

In the case of climatic and sea level change, in order to identify consequences of each key feature of the scenario explicitly, the following four sub-cases have been set up.

Table IV. Sub-cases Setting for Climatic and Sea Level Change

	Engineered barriers system and geosphere	Biosphere	Metal corrosion rate (hulls and ends)
Sub-case 1	time-dependent parameters due to periodic change between fresh and saline water	river model	same as REFERENCE CASE
Sub-case 2	time-dependent parameters due to periodic change between fresh and saline water	periodic change between river model and sea water model	same as REFERENCE CASE
Sub-case 3	time-independent parameters relevant to saline water	river model	same as REFERENCE CASE
Sub-case 4	time-independent parameters relevant to saline water	river model	10 times faster than REFERENCE CASE

Generally, the calculated dose rates are several times higher than that of the REFERENCE CASE due to the conservative choice of parameters relevant to saline water. If the seawater model is employed as a biosphere in sub-case 2, this effect is fairly mitigated to provide a lower dose rate, even less than the REFERENCE CASE for sedimentary rock, due to the high dilution effect available in seawater model.

The enhanced corrosion rate in a saline environment is potentially important especially for nuclides trapped inside hulls and ends, such as activated products, because they are released much faster from metal. It is obvious in Fig.4 that this effect has certain impact on the dose rate, but not a critical one.

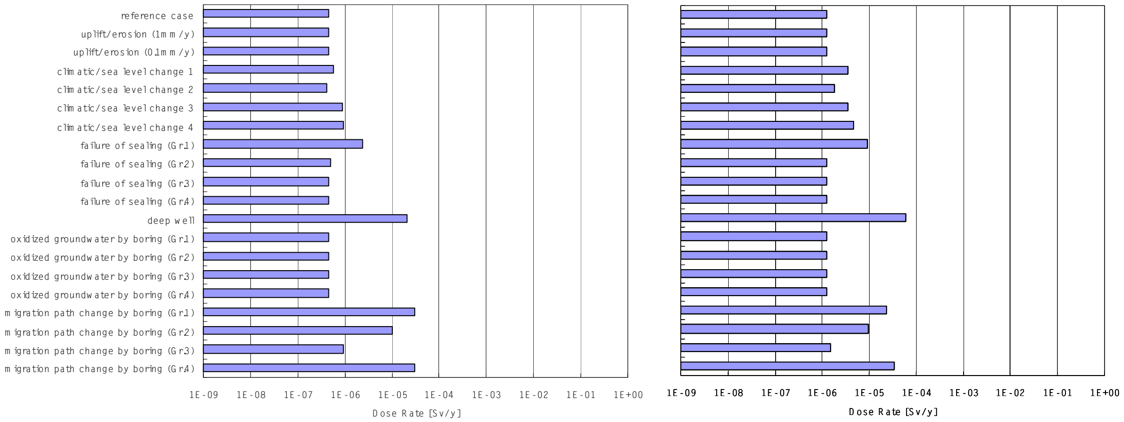
There will be several points where plugs or other sealing will be facilitated in a repository. It is assumed that incomplete sealing will occur at the access shaft; hence it will become a main migration pathway. This means that the natural barrier is substituted for by less effective barrier, the access shaft of 100m long, is much more permeable than rock, from the viewpoint of modelling. According to this assumption, the dose in this case is several times higher than that of the REFERENCE CASE.

The dose rate in the case of a deep well is one of the highest. The only difference between the case of a deep well from the REFERENCE CASE lies in the use of well water, instead of river water in the biosphere. The volume of dilution of well water is much smaller than that of river water. The higher dose conversion factors assigned for the well water use model are reflected directly in the results of the dose rate.

As observed in the case of uplift and erosion, a similar tendency toward enhanced importance of new key nuclides, redox sensitive elements, such as technetium and/or neptunium, also appears in the case of oxidized groundwater due to boring. Contrary to the previous case, however, intrusion of oxidized groundwater is limited within the engineered barriers system and does not extend to natural barriers. Hence, this redox impact is of less importance. Therefore, the calculated dose rate is at the same level as that of the REFERENCE CASE or as that of the case of uplift and erosion.

One of the most significant increases in dose rate, which is approximately one hundred times higher than that of the REFERENCE CASE, is found in the case of migration path change due to boring. This can be explained by assuming the shortcut and/or change of migration pathways in the scenario, ignoring the performance of a natural barrier.

Another new tendency can be found in this case. The maximum peak dose rate of Group 4 is as high as that of Group 1 for sedimentary rock, which is a little higher than that of crystalline rock. The above maximum peak rate of Group 4 can probably be explained by a complete shift of key nuclides contributing to dose rate. In this case, ^{241}Am is identified as the most significant nuclide in Group 4, which is never seen in other cases because of its relatively short half life of 433 years and hence effective decays. Due to the absence of a bentonite layer surrounding disposal units of Group 4 and to ignorance of the performance of any natural barrier, in this case, ^{241}Am associated with its parent ^{241}Pu is released so quickly that it becomes much more significant than ever. This suggests that the results of perturbation scenarios are not always predictable as base scenarios, "normal evolutions", so that utmost care must be exerted in assuming and interpreting consequences for perturbation scenarios.



(a) Sedimentary Rock (b) Crystalline Rock

Fig.4 Calculated Maximum Dose Rate for Perturbation Scenarios

In the case of deep well drilling and of migration path change due to boring, the calculated maximum dose rates do not exceed 0.1 mSv/y, considerably below the safety standard used in foreign countries, namely 0.1-0.3 mSv/y. Taking the probability of perturbation scenarios into account in interpreting the consequences of the scenarios, as in the base scenarios, it can be concluded that Deep Geological Disposal of TRU-containing waste is expected to be feasible from the viewpoint of safety even assuming perturbation scenarios.

CONCLUSION

In this study, an alternative method to classify TRU-containing waste is proposed. Based on the classification, safety for deep geological disposal of some TRU-containing waste is assessed. Results of the assessment have led us to conclude that geological disposal of such types of waste is feasible from the viewpoint of safety, because the evaluated dose rates meet safety goal both for base scenarios and even for perturbation scenarios with conservative conditions hypothetically assumed.

We have prospects for the feasibility of geological disposal of TRU-containing waste from this preliminary assessment. But we must be aware that further studies are required in order to enhance our confidence, because the entire repository system is considerably complicated and remains further research and development for geological disposal of TRU-containing waste.

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