Demonstrating Feasible Disposable Concepts for Transuranic (Tru) Wastes in Japan – An Overview Of Project Tru-2 –

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ABSTRACT

In Japan, the Federation of Electric Power Companies (FEPC) and the Japan Atomic Energy Agency (JAEA) have been collaborating with relevant organizations to promote generic research and development (R&D) for the safe geological disposal of transuranic (TRU) waste based on the technical achievements in Japan's high-level radioactive waste (HLW) disposal program. A result of this collaborative effort was the production of a recent progress report on the R&D for TRU-waste disposal in Japan (project TRU-2). This paper is an overview of TRU-2 describing the key results and some unique methodologies developed. It is estimated that ca 140,000m^3 of TRU-waste will be generated in Japan by 2050. TRU-2 has demonstrated that depending on the alpha, beta and gamma contents, 63% of TRU-waste is suitable for near surface disposal (a few tens of meters deep), 18% for intermediate depth disposal (50-100m) and 19% for deep geological disposal (several 100m). Radionuclide migration analyses were performed with realistic near-field models and databases taking into account the specific design components in a TRU-waste disposal. This was complemented with a unique top-down sensitivity analysis for handling remaining uncertainties. I-129 and C-14 gave the dominant dose but this was much lower than background levels in Japan and regulatory levels in overseas countries. Furthermore, through application of alternative technologies, it was shown that higher containment of I-129 and C-14 could produce even more robust repository concepts over a wide range of geological environments. Finally, it was demonstrated that feasibility and cost could be improved through the concept of co-locational disposal with HLW; a concept that has already been demonstrated in many other countries (e.g. Belgium, France, Germany, Switzerland).

INTRODUCTION

Background and definition of TRU-waste in Japan

The reprocessing of spent fuel (SF) from nuclear power plants (NPP) is an essential requirement for maintaining the nuclear fuel cycle. This is especially important in a country such as Japan which lacks energy resources and is dependent on overseas imports to sustain its energy needs. Components of the nuclear fuel cycle are shown in Fig. 1.

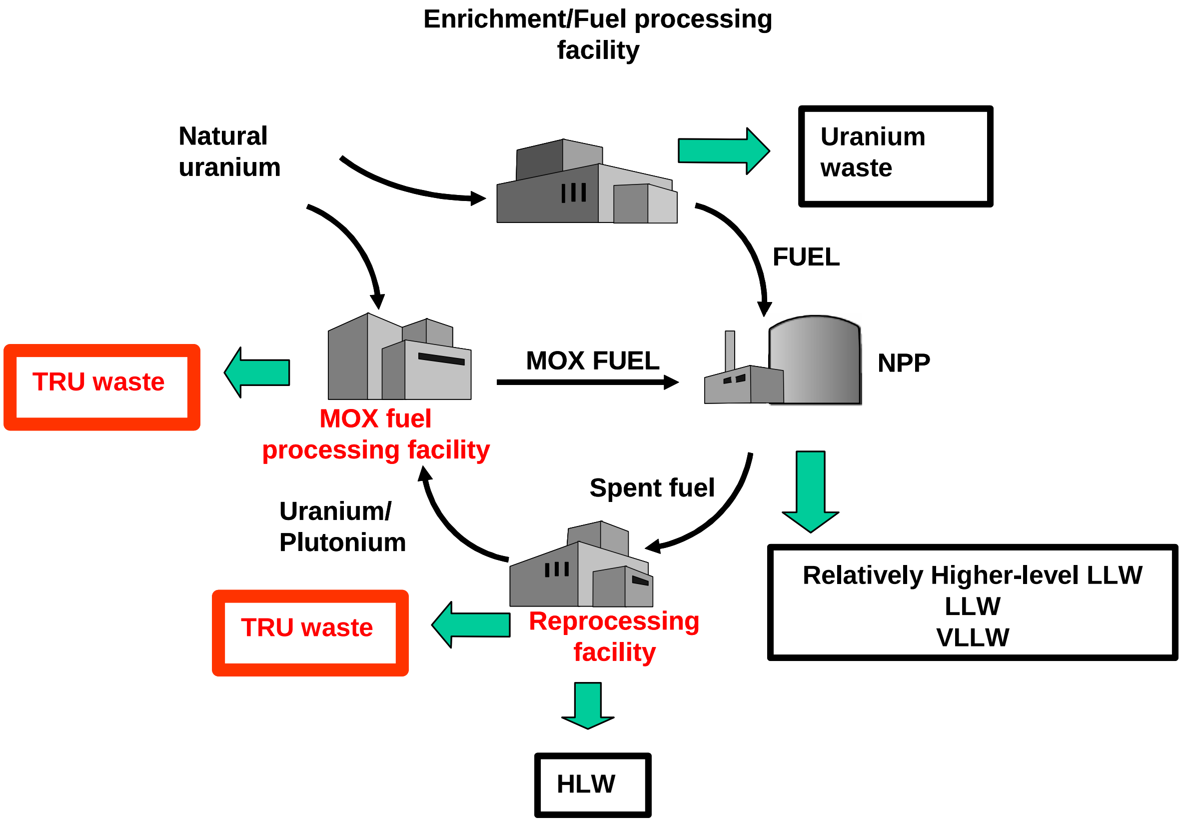

Fig. 1. Simplified diagram showing the generation of TRU and other radioactive wastes in Japan (excluding medical, research and industrial wastes). (NPP, Nuclear Power Plants; HLW; High-Level Waste; LLW, Low-Level Waste; VLLW, very Low-Level Waste)[2].

Large volumes of waste containing long-lived radionuclides (e.g. Pu-239), fission products (e.g. I-129) and activation products (e.g. C-14) will be generated during the operation and dismantling of reprocessing plants and MOX fabrication plants. These kinds of wastes are defined as TRU-waste in Japan. The Japanese TRU-waste definition also includes non-HLW returned from overseas reprocessing plants (British Nuclear Group (BNG) Sellafield (formerly part of British Nuclear Fuel Limited (BNFL)) and AREVA NC (formerly COGEMA)). Fig. 2 shows the distribution of Japan's TRU-waste on a plot of alpha activity versus beta and gamma - although this should be considered as the technical definition.

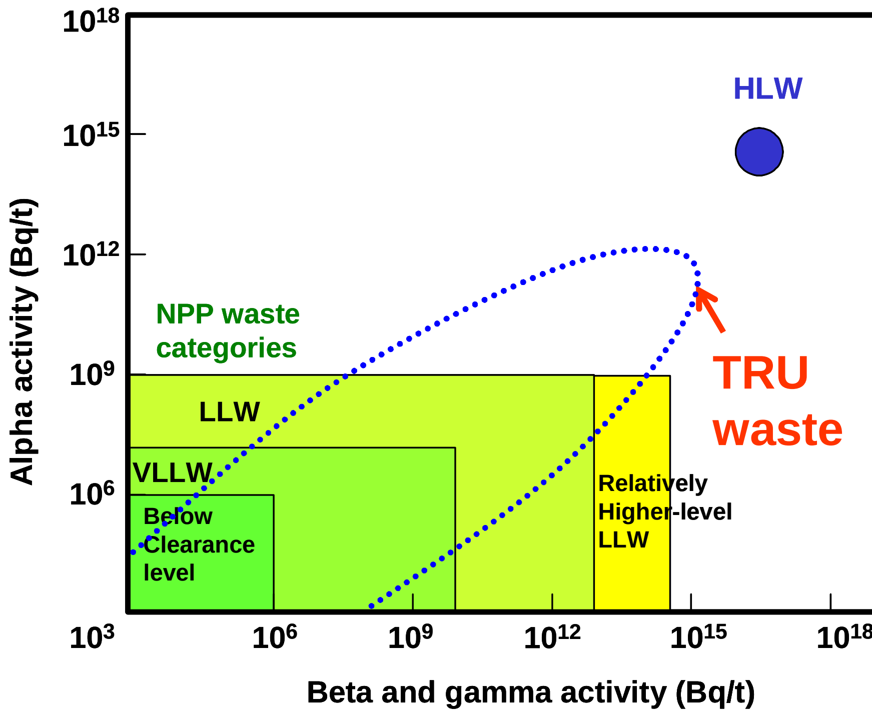

Fig. 2. The radionuclide concentration of TRU-waste (see text for formal definition) and comparison with NPP wastes (green and yellow). Note that VLLW, LLW and relatively higher-level LLW (green and yellow parts) refer to waste generated from the decommissioning of NPPs [2].

TRU-2 report

The Federation of Electric Power Companies in Japan (FEPC) and the Japan Nuclear Fuel Cycle Development Institute (JNC (now JAEA)) published a milestone progress report on the disposal concept of TRU waste in Japan in March 2000 (abbreviated here as TRU-1 [1]). After the publication of TRU-1, a TRU sub-committee was set-up by the Atomic Energy Commission of Japan (AEC) with the task of evaluating the feasibility of TRU waste disposal in Japan. Following guidance from this sub-committee, FEPC and JAEA have been collaborating with Japan Nuclear Fuel Limited (JNFL), the Radioactive Waste Management Funding Research Center (RWMC) and the Central Research Institute of Electric Power Industry (CRIEPI) to expand the generic R&D for TRU-waste and to study the feasibility of TRU-waste disposal in Japan. A result of this large scale collaborative effort was the production of a second progress report on the R&D for TRU-waste disposal in Japan (abbreviated here as TRU-2 [2]).

The objectives of TRU-2 were:

- To build on the R&D of TRU-waste disposal (performed up to 2000) using more realistic boundary conditions and up-to-date databases.
- To describe the progress on Japanese R&D related to TRU-waste disposal.
- To illustrate R&D priorities namely: bentonite-cement interaction models; methods to reduce effects of nitrates which are particularly voluminous in Japan; optimized

approaches including co-locational disposal with HLW, which is new in Japan; alternative designs to immobilize and confine key radionuclides in order to improve on safety robustness.
- Provide a technical basis in order to promote the establishment of a TRU regulatory framework and an implementation body in Japan (currently Japan has no implementor for the geological disposal of TRU-waste).
- Highlight outstanding technical and managerial issues.

This paper is an overview of TRU-2 and highlights the key results and some unique methodologies developed.

EXPECTED TRU WASTE INVENTORY IN JAPAN BY 2050

The volume of TRU waste generated from the operation and decommissioning of domestic reprocessing and MOX fabrication plants, considering a full operation period, and returned wastes from the overseas reprocessing plants (BNG Sellafield, UK and AVENA, France) up to around 2050 is estimated to be ca 140,000 m^3, assuming application of treatment methods currently planned. TRU-2 demonstrated that depending on the alpha, beta and gamma activities in TRU-wastes and properties, 63% would be suitable for near surface disposal (several meters deep), 18% for intermediate depth disposal (50-100m) and 19% for deep geological disposal (300 - 1000m). Although the nuclide composition of the LLW originating from NPPs is different to that of TRU waste, revised criteria set for specific nuclide concentrations for TRU waste was formulated for illustrative purposes using the same calculation methods as for the LLW from the NPPs (Table I).

Table I. Volumes of TRU waste for the various disposal concepts. TRU waste designated for deep geological disposal is further sub-divided into four groups [2].

Disposal concept	***Volume, m^3***	***Group***	***Volume, m^3***
Geological disposal (depth greater than 300 m)	26,641	Group 1	318
		Group 2	6,732
		Group 3	6,175
		Group 4	13,416
Intermediate-depth disposal, (depth 50-100m)	25,205		
Near surface disposal (several meters depth).	88,431		
Total	140,276		

TRU wastes for geological disposal were grouped into 4 types. Group 1 includes weak-sorbing nuclides, such as I-129. Group 2 includes hulls and end-pieces, which generate heat and contain high concentrations of C-14. Group 3 includes chemical substances such as sodium nitrate, which have to be further analysed in terms of impact on radionuclide migration. Group 4 consists of other miscellaneous wastes.

Although all disposal depths are covered in project TRU-2, the report itself has focused mainly on R&D and feasible implementation associated with TRU-wastes destined for deep geological disposal.

ENGINEERED BARRIER SYSTEM (EBS) DESIGN AND LAYOUT

Since the heat generation is small, TRU-wastes for deep geological disposal can be emplaced together in tunnels with large cross-sections. However there is a wide variety of waste materials such as metal, nitrates and organics. For this reason in the basic TRU repository layout, each of the waste groups will be emplaced in separate disposal tunnels. In other words one disposal tunnel or drift will contain waste from one group only (see Table I for associated volumes). Moreover the design specification of each disposal tunnel will depend on the characteristics of the waste group and the surrounding host rock. Since no candidate site exists in Japan yet, generic datasets for soft rocks (ca. sedimentary rock) and hard rocks (ca. crystalline rocks) were used in numerical analyses to model the performance of the EBS designs. Owing to the larger concentration of highly soluble and low sorbing radionuclides in TRU-waste in Groups 1 and 2, it was shown that disposal tunnels for these wastes required a buffer consisting of mixed compacted bentonite and sand. On the other hand, disposal tunnels for Groups 3 and 4 did not require a bentonite buffer.

The effects of thermal stress (originating mainly from Co-60 as gamma heat in Group 2), gas generation and extrusion of buffer material had only a small effect on the mechanical stability of the near-field according to numerical evaluations performed. However, interaction between creep behavior of the host rock and deformation of the EBS could be considerable depending on rock type. Disposal tunnels with horseshoe shaped cross sections and circular cross sections were shown to be mechanically stable in the hard rock dataset but in soft rock dataset only circular disposal tunnels could be used. An advantage of horseshoe shaped tunnels is that they allow more efficient and cost effective techniques for the emplacement of buffer and waste packages.

Note that the above results were based on generic environments. When a site is eventually selected it is planned that safety assessments and the design of the TRU-waste repository layout will be carried out iteratively, so that the design can be modified to take into account the specific host rock boundary conditions. This is particularly important when deciding the design of structural support and layout of disposal tunnels for each waste group. For example Group 3 wastes are likely to form a nitrate plume so the disposal tunnels for these wastes need to be located downstream and at a sufficient distance from the disposal tunnels of the other groups; it was shown that a nitrate plume could have adverse effects on metal supports of the disposal tunnels (although not bentonite) and/or the pathways by which the nuclides migrate from these wastes.

One of the key concerns in the design of the disposal tunnels for Group 1 and Group 2 wastes was alteration of Na-bentonite to Ca-bentonite which has undesirable swelling properties. Based on the evaluation of the spatial variation in chemical properties in the cement-buffer system it is expected that sufficient amounts of montmorillonite will remain after 100,000 years. It was found that Na montmorillonite is dominant for several 10,000 years but then alters to Ca montmorillonite after several 1,000 years. This means that a loss in the impermeability function of the buffer material cannot be ruled out and was taken into account in the safety assessment described below.

Finally detailed planning for construction, operation and closure of a TRU-waste repository in both soft rocks and hard rocks was presented and shown to be feasible with current technology.

SAFETY ASSESSMENT

Since a repository site has not yet been selected and the geological environments in Japan vary widely, in order to demonstrate safety, a conservative disposal analysis that takes into account many types of uncertainty was considered necessary (ca. JAEA's H-12 report on HLW [3])

FEPs

In the safety assessment of the deep geological disposal of TRU-waste, important FEPs that could potentially affect nuclide migration in the repository environment were identified, taking into account Japanese geological environments and NEA's international FEP list [4] as follows:

- Chemical composition of groundwater
- Cement/bentonite interactions
- High-pH plume effects on rock
- Hydraulic conditions in the near-field
- Effect of colloids
- Effects of organic materials
- Effects of microbes
- Effects of radiation
- Effects of sodium nitrates
- Effects of gas generation

Although Japan is a tectonically active country located on or near the boundaries of four converging plates, FEPs related to volcanism and new fault activity were excluded in FEP screening since it was considered that such natural phenomena could be avoided by site selection. FEPs that could be avoided through engineering measures were considered to have little influence on the disposal system, or were considered to be low probability events were also excluded from the safety assessment calculations.

Evaluations of the FEPs showed that cement-bentonite interactions and effects of sodium nitrates were phenomena that contained the largest uncertainties:

Cement-bentonite interactions

It is a well known fact that owing to the large amount of cementitious materials that will be used in a TRU-2 (or I/LLW) repository, interaction between bentonite and hyperalkaline fluids from the cementitious materials can be problematic due to the close proximity of these materials in repository designs. The interaction could cause the alteration of mineralogy and associated change in hydraulic property of bentonite and have a deleterious influence on the function of bentonite as a hydraulic barrier, and may lead to adverse impacts on the long-term safety of the repository. Many studies on such interactions (e.g. [5]) have been carried out in order to establish a bentonite alteration scheme, however, uncertainty in current understanding of the precise alteration scheme, especially mineral paragenetic sequences during alteration, still remains. This is largely attributed to insufficient knowledge about the chemical scheme, thermodynamics and kinetics. Other uncertainties also arise from non-linear relationship between, chemical, mechanical and mass-transport events, and in evaluating how the mineralogy and the hydraulic property of bentonite evolves subject to alteration by hyperalkaline fluids. For this reason, multiple scenarios for mineralogical alteration of bentonite were developed [6] and multiple sets of assumptions were employed for numerical simulation, in order to bound the uncertainties in the evaluation of chemical, mineralogical and hydraulic evolution of EBS over the long-term.

The conditions of bentonite buffer in a radionuclide migration analysis were determined on the basis of the following conclusions drawn from analyses of EBS evolution.

- A realistic case: The impermeable characteristics of the bentonite are expected to be maintained for at least 100,000 years. Moreover, because the precipitation of pore-filling secondary minerals occurs near the interface between cementitious materials and bentonite, nuclide migration is significantly prevented in this area. From the results, it is considered that the alteration of the engineered barrier has a positive effect on the radionuclide retention function. However, it is necessary to note that there may be effects from fractures formed by external factors and inhomogeneous reactions.

- A conservative case: It is assumed that the pore filling by secondary minerals in the interfacial altered layer will not restrict diffusion because of mechanical instability of altered zone. This means that the initial effective diffusion coefficients of the cementitious material and bentonite will be nearly constant in future. In this case, the impermeable characteristics of the bentonite will be guaranteed for at least 100,000 years since sufficient montmorillonite remains.

- A hypothetical case: The following conditions are considered to have low probabilities: montmorillonite dissolution is promoted by constantly unsaturated conditions because the equilibrium constant for montmorillonite solubility is larger than that present in the standard database; the dissolution of montmorillonite is promoted since the dissolution rate law for montmorillonite under highly alkaline conditions follows the instantaneous equilibrium model. In these hypothetical cases, the impermeable property of bentonite will be lost after several 1,000 years.

Effects of sodium nitrates

In Japan, the spent fuel from NPPs is reprocessed and uranium and plutonium are recovered by the PUREX method. This method produces spent nitrate salts (mainly $NaNO_3$) which would be brought into a TRU waste disposal facility (Group 3) if special pre-treatments are not applied. The total amount of nitrate salts to be emplaced in Group 3 disposal tunnels was estimated to be about 3.25×10^6kg. It was found that nitrates and its reduction product (NH_3) could affect the behavior of radionuclides. In a radionuclide migration analysis for Group 3 disposal tunnel, an oxidizing condition and formation of ammine complexation of B-type metal ions were assumed. Moreover NO_3^- might be reduced to N_2 by microbes. However the impact of N_2 gas generation was estimated to be insignificant.

Scenario classification

Scenarios related to safety assessment were constructed from FEPs that were not excluded by screening. The two main scenarios were the groundwater and the isolation failure scenarios. In the former, radioactive materials may be transported to the biosphere by groundwater and gas. In the latter, radionuclides reach the biosphere through a decrease in physical distance between the waste and the biosphere. In contrast with HLW, some radionuclides in TRU-waste (e.g. I-129) may be volatile and a scenario describing the migration of these isotopes in a gaseous form is classified as on of the groundwater scenarios. The groundwater scenarios are further divided into a base scenario, where it is assumed that the present geological conditions and surface environment will continue in the future and perturbations scenario where future changes are considered. The base scenario consists of a reference scenario and alternative scenario. The reference scenario is based on FEPs connected with initial conditions and FEPs connected with the assumed/specified safety function of the disposal system. Alternative scenarios are based on FEPs that could possibly influence the safety function of the assumed/specified disposal system such as the effects of gas and colloid formation (including microbe colloids).

Reference case

Based on the results of the FEP evaluation, nuclide migration analyses were performed with realistic near-field models and databases taking into account the specific design components and phenomena encountered in a TRU-waste disposal system. In the reference case it was assumed that temperature did not exceed 80 degrees centigrade (below which no mineral alteration in cementitious materials is anticipated), cement materials have a pH > 12.5 allowing formation of Ca-type bentonite, re-saturation and radionuclide release was instantaneous, organic material in Group 2 waste affects the EBS, nitrates effect both EBS materials of Group 3 disposal tunnels and sorption partition coefficients of radionuclides. The assumed geological environment is freshwater type groundwater in granite (i.e. hard-rock dataset) with fractures. Dissolution/precipitation, sorption, diffusion and advection dispersion were considered in the nuclide migration analysis in the geosphere. The results of the dose released to the biosphere is shown in Fig. 3 (the same biosphere methodology used in H-12 [3] was applied).

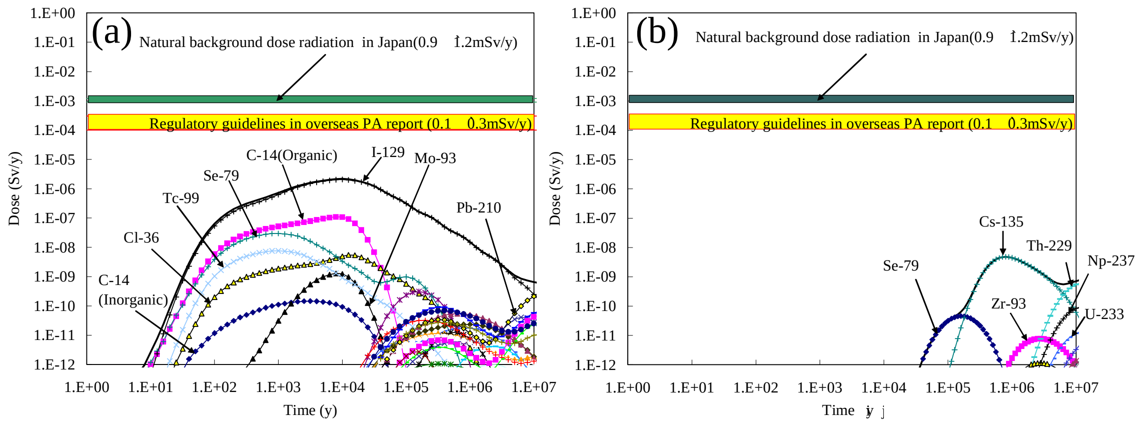

Fig. 3. Calculations of dose rate (reference case) for (a) TRU-waste [2] and (b) HLW [3]. For TRU-waste the maximum dose is ca 2 μSv/y and the dominant radionuclides are I-129 from Group 1 and Organic C-14 from Group 2.

I-129 in group 1 gave the dominant dose but this was much lower than natural background dose radiation in Japan and regulatory levels in overseas countries. However dose was higher than that obtained from HLW waste using the same analyses although no overpack is used in the disposal concept for TRU-waste.

Perturbation scenario

In the perturbation scenario, analytical cases for uplift and erosion, climate and sea level change, initial defects in EBS components, and formation of new migration pathways were evaluated. In many of the cases, the dose did not exceed 10μSv/y. In cases such as the climate and eustatic sea level change, which is assumed to imply relatively fast groundwater flow, or the scenario for the formation of a new transport path by well drilling and water sampling, the maximum dose was 100μSv/y. However using risk theory, it was shown these are low-frequency events.

Isolation failure scenarios

As an isolation failure scenario, uplift/erosion resulting in the repository approaching the ground surface and the accidental penetration of the repository by drilling was evaluated. The former was smaller than the latter. In the case of penetration by drilling, after dose conversion, the risk was shown to be below 10^{-6} - 10^{-5} /y which is the target safety standard in several overseas countries.

Top-down sensitivity analysis

In order to gain confidence in the safety assessments performed, it was necessary to consider the effects of diverse uncertainties in the current generic R&D phase of TRU-waste disposal were also considered. These uncertainties could be evaluated using existing deterministic techniques by increasing parameter ranges and the number of parameter combinations etc. However a comprehensive deterministic evaluation of all uncertainties would result in an unmanageable large number of cases in order to give adequate combinations of uncertainty. To deal with this some national programs (e.g. project Opalinus Clay [7]) have used probabilistic approaches derive a risk estimate for the whole system considering the variation of parameters using probability density functions to model possible parameter variations and the parameter combinations of those in order to complement the adequacy of the deterministic evaluation. However, in addition to such probabilistic estimates on the influence of uncertainty, there is a need in uncertainty analyses to identify the key parameters that have a dominant control on the safety of the system and to extract quantitatively the conditions that would yield a robust system.

To this end, a new top-down sensitivity analysis method was developed for TRU-2. The top-down sensitivity analysis is essentially a stochastic approach and was developed to play a new role in the consequence calculation of uncertainty. The overall methodology consists of: (1) a nuclide migration model that as far as possible comprehensively incorporates the various phenomena that occur within the repository, (2) a stochastic approach used to sample independent parameters randomly and to identify parameters that have a large impact on dose and then to extract threshold values of parameters and/or combinations of parameter thresholds that yield a condition whereby maximum dose does not exceed a target value.

This approach was applied in the safety assessment of the disposal concept considered in TRU-2. It was found that hydraulic parameters significantly affect the maximum dose. If transmissivity, hydraulic gradient, host rock matrix porosity, distance of radionuclide transport, fault length and EBS design parameters were assumed to have reference values intended to be moderately conservative, even if the uncertainties of all other parameters are taken into account, the maximum dose was less than several 10s of μSv/y and safety was not significantly compromised. From the obtained threshold condition, it was confirmed that, if a maximum acceptable target dose of 10μSv/y is assumed, safety is not compromised by the effects of engineered barrier degradation, colloids, gases and initial oxidation. It was also confirmed that the system’s safety would not be compromised in the case that the geological environmental conditions were not as favorable as the reference conditions, if improvements in the performance of waste packages were included.

Alternative technologies

In the groundwater migration scenario, it was shown that the performance of the TRU repository is highly sensitive to the hydraulic conductivity of the geological environment and sorption coefficients of the key nuclides I-129 and C-14 which are both high soluble. On the other hand, the alkaline alteration of bentonite and long-term effects of nitrates are still controversial issues. In case there is a need to deal with these issues, TRU-2 examined two alternative concepts which are still being developed and not currently integrated into the basic disposal concept:

- Improving safety margins for a wide range of geological environments by developing alternative engineered barrier technologies for immobilizing I-129 (e.g. in quartz) and confining C-14 (e.g. in ultra-low-permeable concrete);
- Minimising uncertainties by developing low-pH cement to reduce the alkaline alteration of bentonite and developing nitrate decomposition techniques to reduce the long-term effects of nitrate/bituminized waste.

In all eight immobilisation techniques were studied for immobilizing I-129 in group 1 and two kinds of advanced container for confining C-14 in Group 2 were assessed. This research is related to up-grading engineered barrier technology with the prospect of achieving nuclide retention performance for several tens of thousands of years.

It is expected that in future these alternative technologies will contribute to a greatly improved confidence in the geological disposal of TRU wastes with significant optimized disposal concepts.

OPTIMIZATION MEASURES

The co-location concept of TRU-waste and HLW repositories

In order to reduce costs and the burden on siting, a concept of co-locating HLW and TRU-waste disposal facilities (Fig. 4) was examined. Heat, high pH constituents, nitrates and organic matter were identified as the important factors that have been determined as the critical reciprocal influences from HLW and TRU-waste disposal facilities. Knowledge of these influences was summarized and the effect of the separation distance between the two disposal facilities was analyzed. It was shown that these mutual influences could be avoided by establishing a separation distance between the two disposal facilities of several 100 meters. Additionally, to establish the technology of co-locational disposal, the layout and engineering measures were evaluated (Fig. 4).

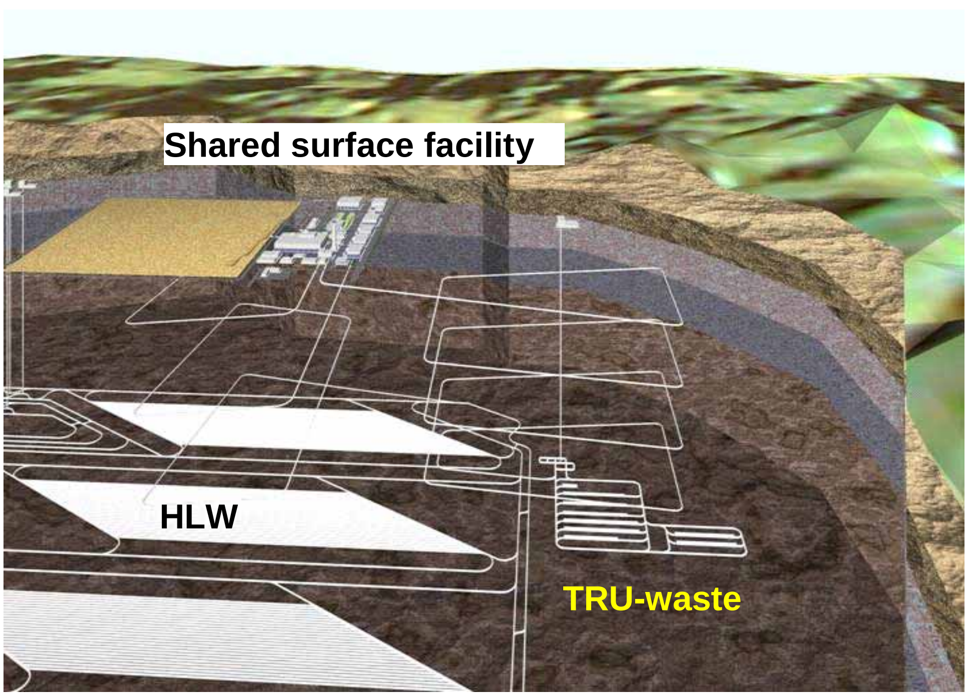

Fig. 4. Example concept for co-located HLW and TRU-waste repositories in a single complex [2].

CONCLUSION

The main outcomes from project TRU-2 can be summarized as follows:

- I-129 from group 1 gave the dominant dose in the reference case but this was less than regulatory limits in countries and natural radiation exposure in Japan.
- The results of evaluations of the perturbation/isolation failure scenario analyses, showed risks below 10^{-6}-10^{-5} /y, which is the target safety standard used in several foreign countries.
- Much uncertainty surrounds cement-bentonite interactions especially regarding the types of cement used, thermodynamic stabilities/kinetics of bentonite, mineral precipitation/dissolution rates etc.
- There is also much uncertainty regarding the behaviour of nitrates in the deep geological environment particularly with respect to transitions in geological media, radionuclide solubility in solutions with high concentration of nitrates.
- Uncertainty could be reduced through the development and application of alternative technologies for immobilizing and confining the key radionuclides (I-129 and C-14), development of low-pH cement to reduce the alkaline alteration of bentonite, and development of nitrate decomposition techniques to reduce the long-term effects of nitrate/bituminized waste.
- TRU-2 has demonstrated the feasibility of disposing of TRU-waste at shallow, intermediate and deep geological depths.
- Feasibility could also be improved through the concept of co-locational disposal with a HLW disposal facility; a concept that has already been demonstrated in many other countries (e.g. Belgium, France, Germany, Switzerland).

Whilst uncertainties and outstanding issues were identified, these were not considered as insurmountable obstacles for making a safety case.

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