International Progress in Developing Cases for Long-term Safety of Repositories for Transuranic and Long-Lived Intermediate Level Wastes

Summary of the Third International Workshop

J. Mathieson, A. Hooper
Nirex
Curie Avenue, Harwell Didcot Oxon, OX11 0RH
UK

W. R. Alexander
Nagra
Hardstrasse 73, 5430 Wettingen
Switzerland

M. Shiotsuki, G. Kamei
JAEA
4-49 Muramatsu, Tokai-mura, Ibaraki 319-1184
Japan

ABSTRACT

In January 2005 an international workshop was held in Oxford, UK to discuss research progress in the disposability of transuranic (TRU) waste (also known as long-lived intermediate level waste). This was the third such workshop, with two previous ones being held in Switzerland in 1996 and Japan in 1999.

Whilst TRU/ILW represents more complex wasteforms than high-level waste (HLW) or spent fuel (SF), it is recognised that it has not received the same amount of attention.

This paper summarises the themes discussed at the workshop and the conclusions derived. Moreover, it provides a status report and future direction of work in relation to TRU and ILW wastes.

The overall conclusion of the workshop was recognition of the uncertainties surrounding the subject and outstanding issues where further work was required. However, there were no insurmountable obstacles to making safety cases for the disposal of TRU/ILW waste - safety assessment strategies were judged to be at a mature stage.

INTRODUCTION

In January 2005 an international workshop was held in Oxford, UK to discuss research progress in the disposability of transuranic (TRU) waste, also known as long-lived intermediate level waste. This was the third such workshop, with two previous ones being held in Baden, Switzerland in 1996 and Tokyo, Japan in 1999.

In summary, the Baden workshop noted whilst some TRU repository concepts relied on a near field chemical barrier, some others further added an additional hydraulic barrier, such as bentonite or a bentonite plus sand mixture. Effects that may perturb the near field were identified as including evolution of repository temperature, organic degradation products, microbes, and the leaching of near-field materials. All TRU concepts relied on a low-permeability host rock to ensure longevity of the chemical barrier and to provide retardation and dispersion of released radionuclides. Overall repository performance could be affected by the presence of colloids and production of gas, and the formation of a high-pH plume, and the co-location of high level waste (HLW) and spent fuel (SF) with TRU waste could also give rise to issues. Overall there was broad agreement on the most relevant issues including performance assessment methodology. Repository concept similarities were noted and the complexity of systems was recognised. Whilst not all phenomena were fully understood, no critical obstacles to repository safety were identified. Areas for further work included the effects of microbes, colloids, high-pH plume, and gas migration and its effects.

The second workshop covered cement degradation, the long-term stability of bentonite, the migration behaviour of key radionuclides, and gas production and release. In general, modelling uncertainties were addressed by making conservative assumptions, but the workshop identified the most important areas that required further work including carbonation and near-field radionuclide release, the behaviour of a high-pH plume (for which natural analogue projects, such as the Maqarin project would help increase understanding), and cement-bentonite interactions. The chemistry of the migration of key radionuclides raised several questions, relating to the wide range of reported Kd values for the same radionuclides. Models for gas generation and behaviour were seen as conservative (particularly for microbial production).

This paper summarises the themes discussed at the third workshop and the conclusions derived. Moreover, it provides a status report and future direction of work in relation to TRU and ILW wastes.

The workshop was attended by representatives of several national radioactive waste management programmes with an interest in TRU and ILW disposal:

- Belgium (ONDRAF/NIRAS and SCK-CEN),
- France (Andra),
- Japan (JNC (JAEA at present), RWMC and CRIEPI),
- Switzerland (Nagra) and
- the UK (Nirex, UKAEA, Serco Assurance and the Environment Agency).

Unfortunately, whilst the US and Germany also have an interest in this area, they were not able to send representatives to the workshop.

By way of introduction to the workshop, each country represented gave a summary of its TRU/ILW disposal strategy. This was followed by a series of topical presentations and discussions on:

- the long-term evolution of a range of TRU/ILW wasteforms;
- interactions between cementitious wastes and bentonite backfill;
- interactions between cementitious wastes and repository host rocks;
- potential impact and influence of nitrate-rich waste streams on a repository;
- gas generation and transport issues in the near field and geosphere;
- expected behaviour of cellulose and associated degradation products and their potential influence on radionuclide containment;
- examination of designs for the disposal of TRU/ILW waste streams on the same site as HLW and SF wastes;
- advanced encapsulation designs for very long-lived waste streams; and
- new performance assessment tools.

The presentations identified the state of development in the understanding of system behaviour and modelling. It was felt that the values of some modelling parameters should be verified and some models (e.g. for gas generation) needed better validation. In many cases over-conservative assumptions were being made but some of these limitations will be addressed by ongoing research programmes.

There was felt to be a better understanding in several other areas, such as the impact of organic degradation products in the near field, leading to more realistic assumptions on their behaviour. Of particular interest to some countries was plutonium behaviour in a repository and this was identified as an area for further international collaboration. It was further identified that by applying simple engineering solutions in repository architecture, many of the remaining uncertainties can be accommodated and some potentially expensive research could be reduced in scope, such as the behaviour of nitrate-bearing wastes. These would include the segregation of wastes types in different repository caverns, which applied equally in consideration of the possible co-siting of TRU/ILW disposal with that of HLW and SF. It was noted, however, that the waste inventory in some programmes was not detailed enough to allow segregation but it was felt that improving the inventory in these instances was the most appropriate solution.

There are clear differences in the strategies of programmes that have identified a specific host rock and those where the host rock remains an open issue, the latter requiring a greater degree of flexibility in the safety burden to be carried by the engineered system and the geosphere respectively.

The overall conclusion of the workshop was recognition of the uncertainties surrounding the subject and outstanding issues where further work was required. However, there were no insurmountable obstacles to making safety cases for the disposal of TRU/ILW waste.

DEFINITION OF TRU WASTE

It was noted at the outset of the workshop, that there was no common definition of transuranic (TRU) waste. Each country which used the term appeared to have a different definition and therefore care should be taken in reading this paper to avoid any misconceptions which may arise. Whilst countries such as Japan and the US, (and the IAEA), do have formal definitions of TRU, others do not. However, for the purposes of the workshops and for this paper, TRU broadly equates to long-lived intermediate level waste (ILW) and low-level waste (LLW) with "significant" alpha content. Further, a number of countries also had particular radionuclides to consider, such as Pu and U in a few cases, although Cl-36 and I-129 were also common factors.

Some examples of the interpretation of "TRU" are:

- In Belgium, A3X waste which includes high Pu content material and defined as arising mainly from operational and decommissioning activities of the MOX production plant.
- In France it included "B" wastes which cover seven waste categories arising from reprocessing operations, PWR maintenance and research.
- In Japan it is defined as waste generated from the operation and decommissioning of reprocessing and MOX fabrication plants, but excludes HLW. Moreover, it does include non-HLW returned reprocessing waste from BNGS and COGEMA. Therefore Japanese TRU encompasses many classes of waste from below clearance level to greater than LLW (i.e. ILW) (see Fig. 1) and comes under four groupings: Group 1 predominantly containing I-129; Group 2 hulls and ends containing C-14; Group 3, nitrate bearing waste (e.g. bituminized waste produced in JNC); and Group 4, other technological wastes.
- In Switzerland the term would cover long-lived ILW (or, in German, LMA) which mostly arise from reprocessing operations at BNFL and COGEMA.
- In the UK "TRU" was broadly equivalent to ILW, which, in the UK, is not segregated by half-life and to LLW containing long half-life material, notably Pu.

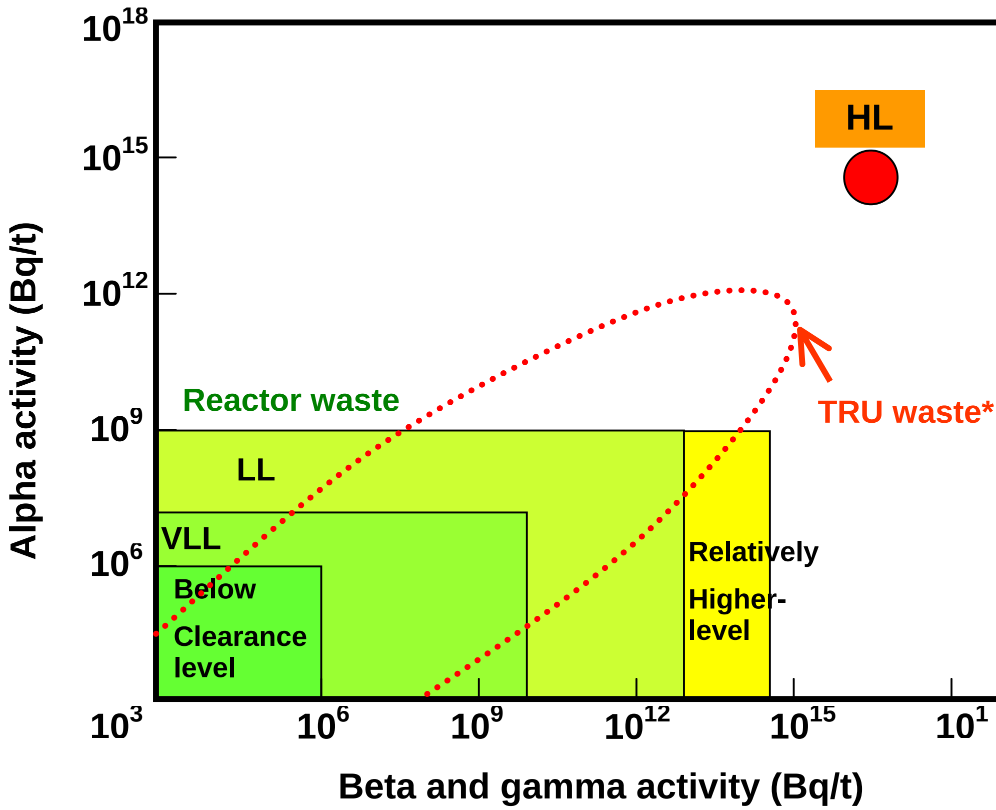

Fig. 1. TRU waste categorisation in Japan

*In Japan, TRU waste means wastes generated from operation and decommissioning of Reprocessing and MOX fabrication plants except HLW.

STATUS OF NATIONAL TRU PROGRAMMES

It was recognised that the national TRU programmes were at various stages in examining the feasibility the deep disposability of TRU/ILW and that not all governments (at the time of the workshop) had established a TRU/ILW specific policy. Several countries were examining the co-location of TRU/ILW wastes with HLW/SF, noting also that some classes of TRU/ILW (such as nitrate-bearing wastes) were also to be segregated within the same repository design.

The national programmes are summarised as follows (noting there have been developments in a number of them since the workshop):

- Belgium expects to begin construction of a repository in 2030, with operations from 2040 if the Boom Clay is confirmed as the reference host formation.
- France has identified a site in argillaceous sediment in the Meuse/Haute Marne area and began constructing an underground laboratory in 2001 at Bure. Since the workshop, a final report on the feasibility of deep disposal has been submitted to the National Evaluation Commission in preparation fro a Parliamentary debate in 2006 [1].
- Although Japan had established the Nuclear Waste Management Organisation (NUMO) in October 2000 this was only to implement geological disposal for vitrified HLW; construction of a repository was planned for the 2030s. Since the workshop, Japan had published a second progress report on TRU [2].

- Nagra of Switzerland had carried out a series of safety assessments over the last 30 years (the latest, Project Opalinus Clay, in 2002 [3]) which show that a co-located repository for TRU/ILW and HLW/SF/MOX would be possible in either crystalline or sedimentary host rocks. Following regulatory review of Project Opalinus Clay and a public discussion, a Swiss federal government policy statement is expected in 2006.
- In the UK, a Government policy decision is awaiting the outcome of ongoing public consultation and the deliberations of its advisory Committee on Radioactive Waste Management (CoRWM)' they will be making recommendations to the UK government in July 2006. At the time of writing this paper, they had shortlisted four long-term management options for consideration: storage, shallow disposal for short-lived wastes, deep geological disposal and phased deep geological disposal.

It was noted that many concepts grouped TRU wastes into different categories for separation within the repository. Repository architecture was therefore important, but the waste management organisations had to be clearer as to why separation was required. The inventory was a key tool for this, and should provide a comprehensive description of the radioactive, chemical and physical characteristics of the wastes.

Separation and co-location of HLW/SF and TRU/ILW was a feature of repository siting. Certain national programmes expressed concern that they would have limited chances at finding a repository site and thus the "criteria" for module separation would be very relevant. Segregation of ILW waste types was also critical and the reasoning for this would need to be explained.

"PROBLEMATIC" WASTES

Presentations in this session covered the effects of nitrate waste on chemical conditions in the near field and on engineered barrier systems (EBS), gas generation and transport in the post-closure phase, and cellulose degradation.

It was queried whether enough work had been done on nitrate bearing wastes, but it was recognised that potential issues could be resolved with engineering solutions for segregation and also re-treating the waste prior to disposal to minimise any issues. Different countries had different levels of nitrates in their wastes and the effects of this variability perhaps needed more discussion to better understand cement-bentonite interactions and redox potential. The behaviour of nitrates in conventional waste was an area where existing studies could be helpful in providing data. In addition, work on chemotoxic waste and from marine and soil studies may be relevant.

A TRU/ILW repository will produce gas through processes such as metallic corrosion, or microbial action and radiolytic degradation for organic wastes. Models for gas generation and behaviour were presented but it was noted that C-14, especially in organic form presented some uncertainties. Overall it was concluded that gas production rates are relatively low and that the gas is expected to be dispersed without disrupting the host rock and that this pathway would not contribute significantly to dose levels. However, different inventories of wastes, model assumptions and host rock characteristics could give rise to significantly different results in these conclusions.

Microbial degradation assumptions for the near field were thought to be over-conservative as the availability of nutrients here was questionable. Some work had shown that, theoretically, enough nutrients existed to support higher microbial populations than were observed. More work was required under actual repository conditions before microbial activity can be properly defined. Data on gas production rates in the EBS, particularly from microbial origin, were inconsistent and further effort to compare and evaluate datasets could be of value. A future large-scale gas experiment for both clay and fractured rock may be helpful here.

Wastes can contain significant quantities of cellulose in the form of paper, cotton cloth, etc. Degradation of this material gives rise to organic compounds that can complex with TRU/ILW nuclides. The most important degradation product is iso-saccharinic acid (ISA) as it increases the solubility of Pu^{4+} and decreases its sorption. Material presented at the workshop on the rate of degradation of cellulose and on the sorption of Pu and Am suggests that cement strongly sorbs ISA, whilst ISA sorption on Boom clay is negligible. It was concluded that small amounts of cellulose-containing waste are compatible with a cementitious repository in a clay-based host rock formation and that the understanding and confidence in this area was now sufficient for the development of a robust safety case. In the past, its importance had been overestimated but, recognising that, more confidence of the far-field behaviour would still be useful.

DISPOSAL STRATEGIES

This session heard specific updates on UK and Japanese progress on co-located repositories.

A "co-located" repository is designed to take a range of different wastes, such as SF, vitrified HLW and TRU/ILW within a single complex. However, it is envisaged that these different wastes would have separate vaults with different engineered barriers. For example, in the Japanese concept, HLW would be surrounded by bentonite whereas TRU classified into Group 1 and 2 would have a cementitious barrier in addition to bentonite barrier. Group 3 and 4 would have a only cementitious barrier. Fig. 2 shows Nagra's concept.

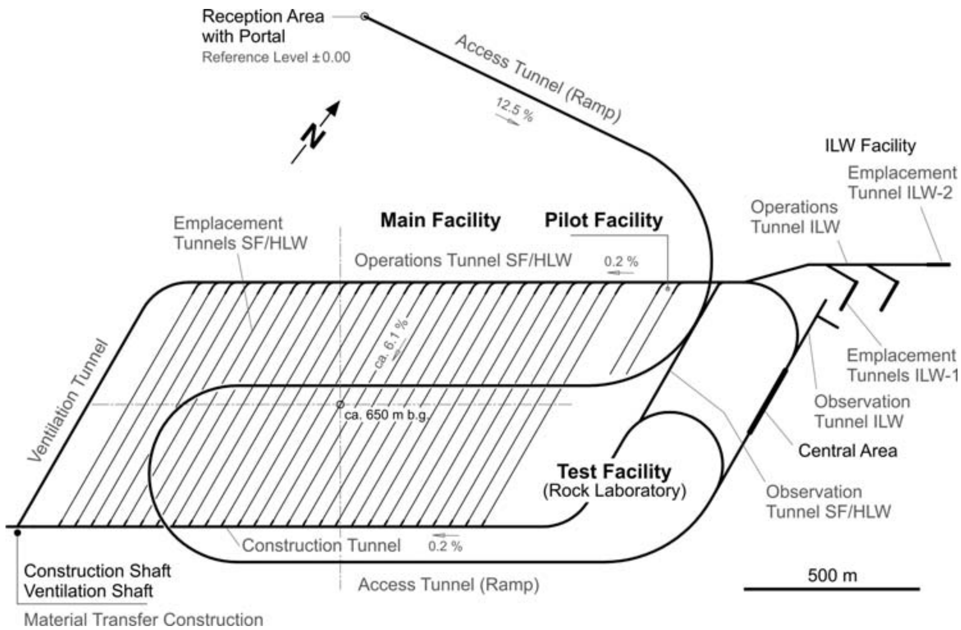

Fig. 2. Co-location of HLW/SF and ILW facilities

One issue in such a facility is that water with a high pH and/or high nitrate content leaching from the TRU/ILW wastes could adversely change the properties of the bentonite barrier (if present). Such effects though could be avoided by separating the two facilities by a distance of a few tens to a few hundred metres. Other work had concluded that heat transfer and not chemical interactions may be the most important factor in optimising the separation of HLW/SF and TRU/ILW disposal vaults.

For all participants, the issue of separation distances between the HLW/SF and TRU/ILW vaults was an important factor which would need to be considered if siting a co-located facility. There were common factors for all countries, but some properties were clearly host-rock dependent. Nevertheless, it was felt that most potential problems could be surmounted by appropriate, optimised repository designs and that no particular mechanism would rule out co-location.

The use of the term "co-disposal" was discussed. To some this could imply that both types of waste were actually co-disposed in the same vault. Given also that some types of TRU/ILW waste had to be separated from each other, preferred terms were "modular" or "segregated disposal" or, at the very minimum, wastes "co-located" at the same site.

Regarding the separation criteria used, some of these were clearly safety related and others were used to make the modelling easier. Further, some modelling aspects still needed to be developed, such as the time dependence of interactions, and corrosion and dissolution rate assumptions. In any event, it was recognised that cement/clay (i.e. bentonite) interactions were an issue for a HLW/SF repository in itself. Criteria had to be derived for how much cement would be allowed in these situations, noting that the Finnish ONKALO Project in support of the spent fuel repository is developing such criteria.

ENGINEERED BARRIER SYSTEM

Presentations in this session covered chemical evolution of the cement barrier, studies on a EBS design and advanced waste forms for iodine filters and containers for hulls waste, and a newly developed safety assessment tool for coupled processes (HMC: hydraulic, mechanical, chemical).

In general discussion on this session, various points were noted. The excavation disturbed zone (EDZ) does not "live forever". For example, experiments in the Mont Terri underground rock laboratory in Switzerland had shown that permeability in the EDZ approaches that of the undisturbed geosphere, through self-sealing, after only two years. For the case of diffusive systems, it was suggested also that results of Mont Terri could be supplemented with data from the Bure site and other, much older, tunnels (such as the Hauenstein railway tunnel in Switzerland).

If diffusion is the dominant method of radionuclide transport, then there would be no important phenomenological effects of the alkaline plume (apart, perhaps, from the effect of altered rock properties on gas migration). In the case of advective flow, the picture is much more complex and will need to be examined on a host-rock specific basis.

Worldwide, many groups have attempted to couple chemical reactions and transport of the hyperalkaline plume, but had difficulty in validating the models (although it should be noted that they presently err on the conservative side). Coupling was thought not to be useful for direct application in nuclide transport calculations, but was valuable for demonstrating phenomenological understanding.

It was felt that more needs to be done on basic thermodynamic data. The lack of good data made quantitative analyses difficult although it was adequate for general trend analyses. In many cases the use of engineering and design solutions to overcome the lack of good models was a more cost-effective method of dealing with the situation.

Studies emphasise that we have a good understanding of cement degradation and evolution. Where uncertainties still exist, parameters can be bounded to cater for this. The key point on TRU/ILW waste is that the approach to disposal policy and associated R&D has to be flexible. It contains many chemicals and a variety of wasteforms and, while the inherent uncertainties and difficulties caused by this should not be underestimated, TRU/ILW was not a 'problem' waste. Better consideration of decommissioning requirements and using an integrated approach to TRU disposal would further mitigate any outstanding issues.

Although it was generally agreed that the basic understanding of hyperalkaline leachate/clay interaction is appropriate, there is currently little confidence that the level of understanding is good enough to allow optimisation of the EBS. In the case of a bentonite buffer, over-conservative calculations, such as used in Project Opalinus Clay, will certainly cover any worst case conditions – but clearly point out the need for better mechanistic understanding to allow eventual optimisation of the designs. Alternatively, it could be argued that it is best to simply avoid the use of OPC in association with bentonite. Such difficulties could be avoided by using novel materials and establishing criteria for these.

With respect to safety assessment strategy, although this was a complex story to follow, we are arguably at a mature stage and this is therefore a valuable area for information exchange. Unfortunately, because of the complexity of most approaches, it does not appear transparent to all concerned. It was suggested that an internet forum should be created for exchanges of views. This may help people to focus their R&D programmes. These could be then brought together at the next TRU/ILW workshop.

CONCLUSIONS OF THE WORKSHOP

In summary, the workshop arrived at a number of conclusions and identified where further work would be advantageous.

There was no agreed definition of “TRU” although its meaning was broadly understood. However, care had to be taken if the term was used out of context as each country had different categories of waste types to consider. The key point on TRU waste is that the approach to disposal policy and associated R&D has to be flexible as it contains many chemicals and a variety of wasteforms. It was suggested that problematic wasteforms could be avoided by better consideration of decommissioning requirements and using an integrated approach to TRU disposal.

Many TRU disposal concepts sub-categorised TRU for separation within the repository, but there had to be a clearer appreciation of why such separation was required. There was a move towards a commonality of repository design regarding separation and segregation of TRU and HLW/SF/MOX. However, repository siting criteria may have to explicitly recognise this aspect, noting that cement- bentonite interactions were an issue for a HLW/SF repository in itself.

It was generally agreed that with reference to siting TRU and HLW/SF facilities, the terms “modular” or “segregated” disposal, “co-siting” or “co-locating” were preferred to “co-disposal”.

Studies emphasise that there is a good understanding of cement degradation and evolution. Where uncertainties still exist, parameters can be bounded to cater for this. It was agreed that there was a basic understanding of hyperalkaline leachate/clay interaction, but this may not be enough to allow optimisation of the EBS. Over-conservative calculations, will cover any worse case conditions or the use OPC in association with bentonite should be avoided.

Safety assessment strategies are at a mature stage and therefore a valuable area for information exchange. It was suggested that an internet forum should be created for exchanges of views on areas such as this and may help to focus R&D programmes.

Areas where more work was seen as advantageous are:

- nitrates waste variability and cement bentonite interactions and redox potential;
- modelling information for C-14 behaviour in the biosphere;
- gas production rates in the EBS, particularly a large-scale gas experiment for both clay and fractured rock;
- in relation to nitrates and bacteria:

➢ the reaction of microbial populations in host rocks to the disturbing presence of the repository;

➢ the effect on microbes in hyperalkaline systems;

- the use of data from the behaviour of nitrates in conventional waste, recognising some organisations were already looking at this;
- long-term experiments on new wasteforms and packages;
- although the understanding of cellulosic waste and ISA was felt to be in “good shape”, a better understanding of the effect of organic degradation products on bivalent radionuclides would be advantageous;
- sorption datasets for dehydrated and cements other than OPC. In addition, low pH cements, consideration of the loss of low radionuclide solubilities induced by hyperalkaline cement porewaters needs to be addressed.

NEXT WORKSHOP

It was agreed that TRU/ILW workshops would be held every two years with the next one hosted by ANDRA in France in 2007.

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