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REVIEW OF INTERNATIONAL PROGRESS IN TRANSURANIC AND LONG-LIVED INTERMEDIATE LEVEL WASTE DISPOSAL

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ABSTRACT

The disposability of transuranic (TRU) waste (also known as long-lived intermediate level waste, ILW) has not received the same degree of attention as vitrified high-level waste (HLW) or spent fuel (SF). In many respects TRU/ILW represent more complex wasteforms.

Two international workshops (in Switzerland in 1996 and Japan in 1999) have discussed the topic and several studies dedicated to such wastes have been undertaken, such as that by JNC of Japan in 2005.

More recently, a third workshop in the series was held in January, 2005 in the UK, with the aim of providing an update on recent international progress in the field and indicating areas of potential common interest for future collaboration.

This paper reviews the topics discussed at the workshop, presents the conclusions of the workshop and provides an update of the current status and the future direction of work in relation to TRU/ILW wastes.

Overall, the workshop concluded that whilst there were still uncertainties and a number of outstanding issues where further work was required, there were no insurmountable obstacles to making safety cases for the disposal of TRU/ILW waste.

INTRODUCTION

The disposability of transuranic (TRU) waste (also known as long-lived intermediate level waste, ILW) has not received the same degree of attention as vitrified high-level waste (HLW) or spent fuel (SF). In many respects TRU/ILW represents a more complex wasteform. Two international workshops (in Switzerland in 1996, hosted by Nagra, and Japan in 1999, hosted by JNC) have discussed the topic. Several studies dedicated to such wastes have been undertaken, such as that by JNC and other Japanese institutions in 2005 [1]) which continues on from earlier studies [2].

More recently, a third workshop in the series was held in January, 2005 in the UK, hosted by UK Nirex Ltd, with the aim of providing an update on recent international progress in the field and indicating areas of potential common interest for future collaboration. This paper reviews the topics discussed at the workshop, listed below, presents the conclusions of the workshop and provides an update of the current status and the future direction of work in these areas.

The workshop was attended by representatives of several national radioactive waste programmes, including Belgium (ONDRAF/NIRAS and SCK-CEN), France (Andra), Japan (JNC, RWMC and CRIEPI), Switzerland (Nagra) and the UK (Nirex, UKAEA, Serco Assurance and the Environment Agency). Discussions were focused on TRU/ILW issues in those countries, but it is acknowledged that the US and German programmes also deal with these waste types. However, they were not able to send representatives to the workshop.

Following presentations on TRU/ILW disposal strategies in each national programme, a series of topical presentations and discussions were held 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 participants identified the state of development that has been reached in understanding of system behaviour and modelling. Values of some modelling parameters should be verified and some models (e.g. for gas generation) needed better validation. In many cases this has led to over-conservative assumptions being made. It was clear that some of these limitations will be addressed by ongoing research programmes.

Knowledge on other areas, such as the impact of organic degradation products in the near field were now much better understood than previously, leading to more realistic assumptions on their behaviour. The behaviour of Pu in a repository was of interest to some countries and it was agreed that there should be further international co-operation on this subject.

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.

Overall, the workshop concluded that whilst there were still uncertainties and a number of outstanding issues where further work was required; there were no insurmountable obstacles to making safety cases for the disposal of TRU/ILW waste.

DEFINITION OF TRU WASTE

It should be noted at the outset, that there is no common definition of transuranic (TRU) waste; each country which uses the term either officially or unofficially appears 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), may 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, specific countries also had particular radionuclides to consider, such as Pu and U in a few cases, although ^{36}Cl and ^{129}I were common factors.

For example, TRU waste in Belgium included what was known as A3X waste, defined as a high Pu containing waste, arising mainly from operational and dismantling activities of the MOX production plant. In the French case, TRU would include B wastes representing some seven categories of waste arising from reprocessing operations, PWR maintenance and research. TRU waste in Japan is defined as waste generated from the operation and decommissioning of reprocessing and MOX fabrication plants, but excludes HLW; it does include non-HLW returned waste from BNFL and COGEMA. Japanese TRU thus encompasses many classes of waste from below clearance level to greater than LLW (i.e. ILW) and comes under four groupings: Group 1 predominantly ^{129}I; Group 2 hulls and ends containing ^{14}C; Group 3, nitrate and bitumen; and Group 4, other technological wastes.

Within Switzerland, long-lived ILW (or, in German, LMA) mostly arose from reprocessing operations at BNFL and COGEMA. The UK also did not have a TRU waste classification, but it was broadly equivalent to ILW, which, in the UK, is not segregated by half-life.

SUMMARY OF THE PREVIOUS WORKSHOPS

Baden workshop (1996)

The Baden workshop noted that TRU repository concepts discussed rely on the near field to provide a chemical barrier, with some including a physical, barrier such as bentonite or a bentonite plus sand mixture. Several phenomena had been identified that may perturb the near field, such as evolution of repository temperature, organic degradation products, microbes, and the "flushing away" of near-field materials.

All repository concepts described relied on low permeability host rock to ensure longevity of the chemical barrier and to provide retardation and dispersion of released radionuclides. Phenomena that had been identified that may perturb the repository performance included the presence of colloids and gas, the formation of a high-pH plume. The co-location of high level waste (HLW) and spent fuel (SF) with TRU waste could also present challenges.

The overall conclusion of the first workshop was that 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. Issues that were identified as requiring further work included the effects of microbes, colloids, high-pH plume, and gas migration and its effects.

Tokyo workshop (1999)

The second workshop discussed four "key issues": cement degradation, the long-term stability of bentonite, the migration behaviour of key radionuclides, and gas production and release. In general, modelling uncertainties in any of these areas was addressed by making conservative assumptions, but the workshop identified the most important areas which required further work. Such uncertainties included carbonation and near-field radionuclide release, the behaviour of a high-pH plume (for which natural analogue projects, such as the Maqarin project (see [3]), would help increase understanding), and cement-bentonite interactions.

The chemistry of the migration of key radionuclides posed several questions, noting there was a wide range of reported Kd values for the same radionuclides. Moreover, ^{129}I and ^{36}Cl were often assigned a value of Kd=0, but even a small non-zero value would have a high beneficial impact on doses. Gas generation was recognised as being dependent on waste composition and on individual repository conditions. Models for gas generation and behaviour were seen as conservative (particularly for microbial production).

SUMMARY OF CURRENT (2005) NATIONAL POSITIONS

The participating countries are at various stages in examining the feasibility of deep disposal for TRU/ILW and not all governments have an established radioactive waste management or 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 design concept.

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. A final report on the feasibility of deep disposal is due to be presented to the French parliament in 2005 for a decision in 2006.

Japan established the Nuclear Waste Management Organisation (NUMO) in October 2000 to implement geological disposal for vitrified HLW and a volunteer site selection programme has begun, with construction of a repository planned for the 2030s. However, a policy decision on implementation for TRU/ILW wastes was expected in 2006. Nagra of Switzerland has carried out a series of safety assessments over the last 30 years (the latest, Project Opalinus Clay, in 2002 [4]) 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.

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 countries would only have one go 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 ^{14}C, especially in the form of CH_4 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 discrepancies 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 which 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/ILW would have a cementitious barrier.

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 its models, although these erred 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 is needed 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 though, by using familiar 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.

NEXT WORKSHOP

It was agreed that TRU/ILW workshops would retain their high value to participants if held every two years. The next one would therefore be held in 2007, and the participants were pleased to accept the offer of Andra to host this in France.

ACKNOWLEDGMENTS

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