Integration of TRU disposal studies in Switzerland

Ian G. McKinley, W. Russell Alexander
Nagra, 5430 Wettingen, Switzerland

ABSTRACT

Long-lived intermediate-level waste ("TRU") is recognised as one of the major waste categories in the Swiss national inventory. Disposal studies are relatively far advanced and involve extensive international collaboration.

An important milestone in the Swiss programme will be the publication of Project *Entsorgungsnachweis* in 2002 which will aim to demonstrate the feasibility of siting a repository for co-disposal of SF (spent fuel), HLW (vitrified high-level radioactive waste) and cementitious TRU in Switzerland. Thereafter, work on TRU is expected to continue focused on the underground test sites and collaborative projects until a decision is made to implement a repository in Switzerland (or join an international project) in 20 - 30 years time.

BACKGROUND

In Switzerland, Nagra is responsible for developing repository projects for all types of radioactive waste resulting from a nuclear power programme (which supplies ~40 % of the total national electricity requirements) and from medicine, industry and research (MIR). Although Switzerland is a small country and the nuclear power programme totals only ~3 GW(e), the spectrum of wastes produced is very wide. This results not only from reprocessing of about 1/3 of the expected spent fuel arisings (for a reference reactor lifetime of 40 years) but also from the great diversity of MIR sources.

All radioactive waste in Switzerland is planned to be disposed of in one of two deep repositories (Figure 1). Most low- and intermediate-level waste (L/ILW) will go to an "in-mountain" repository featuring horizontal access to emplacement caverns located several hundred metres below the surface. A site for such a repository has been identified at Wellenberg in Central Switzerland but further steps towards implementation (involving construction of an exploration tunnel) will be dependant on the results of a Cantonal referendum expected in Summer 2002. The waste inventory for this repository will be constrained by the results of a performance assessment using site-specific data, but wastes containing significant concentrations of long-lived radionuclides would be excluded.

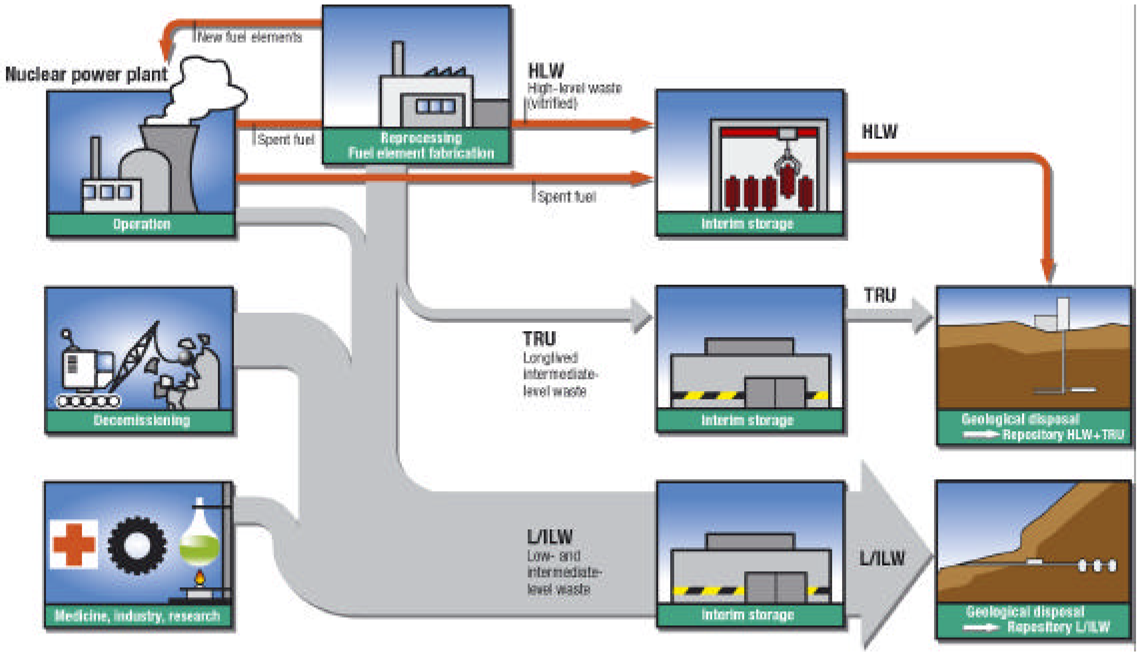

Fig. 1. Swiss waste source / repository types

Long-lived intermediate-level waste (often referred to as TRU – even though transuranics may not be the most problematic radionuclides present) are planned to be co-disposed with vitrified HLW and SF (both conventional UO_2 and MOX). Two host rock options are presently being considered – crystalline basement and Opalinus Clay – both located in the Northern part of Switzerland, close to the border with Germany. During 2002, a major project to demonstrate the feasibility of siting such a repository in a specific area of Opalinus Clay (Zürcher Weinland) will be published (the Entsorgungsnachweis, or EN, project).

INVENTORIES

The repository of HLW/TRU is not required before about 2050 because of the availability of adequate interim storage capacity. Even the decision of whether to implement a repository in the crystalline or sedimentary host rock – or even to go for a possible multi-national option – need not be made in the next couple of decades. Over this period, important decisions will be made on the future of nuclear power (e.g. whether or not to extend the life of or replace existing nuclear power plants) and the back end of the nuclear fuel cycle (e.g. whether to reprocess or not; if waste substitution will be allowed) which will greatly influence the inventory of TRU waste which has to be disposed. Equally important are future developments in the technology of waste treatment and conditioning, which will influence waste volumes, properties and packaging.

To guide repository programme development, therefore, model inventories have been developed in Nagra for different future scenarios and these are an important complement to the detailed inventories of existing wastes.

Both the inventory of activity (or radiotoxicity) and that of key long-term, safety-relevant nuclides are dominated by returned TRU from reprocessing. In general, however, such waste is relatively well characterised.

More problematic from the point of view of inventory specification (even if not from the aspect of long-term safety following disposal) are some long-lived ILW from reactor operation / decommissioning and from MIR. An example from the former category would be highly activated reactor internals and from the latter would be materials activated by exotic particles in high-energy physics research facilities (e.g. particle beam dumps). In many of these cases, direct characterisation of all different waste types would be extremely difficult and costly. Nagra has thus developed in-house modelling capacity to predict nuclide inventories in a wide range of activated wastes. Model calibration / reality checks can then be restricted to a few easily measured γ-emitters.

Inventory development is a continuous process which requires not only sophisticated calculational tools / databases and experienced manpower but also extensive, open contact with the waste producers. Nagra experience in development of inventories of the Swiss national programme has been expanded by collaborative projects on Italian (with Ente per le Nuove tecnologie l'Energia e l'Ambiente – ENEA) and Japanese (with the Japan Nuclear Cycle Development Institute – JNC) wastes.

DISPOSAL CONCEPTS

Nagra concepts for disposal of TRU depend somewhat on the repository host rock. The waste itself is predominantly immobilised in steel or concrete containers by a cementitous grout. Earlier designs for crystalline rock considered such containers to be loaded into a concrete silo, which was then backfilled with cement. The silo itself is surrounded by a thick layer of compacted bentonite.

More recent designs for a Swiss sedimentary host rock considered horizontal emplacement in concrete lined caverns within standard-sized concrete packages which are infilled with a special gas-permeable grout. In this case, the caverns require no external bentonite layer.

As discussed further below, a recent performance assessment [1] clearly indicated that potential long-term releases from a TRU waste repository are dominated by only a few components of this heterogeneous waste category and hence segregation of waste into different sub-categories can allow more extensive engineered barriers to be included for the more problematic waste.

The very high performance of the natural geological barrier for the case of the Opalinus Clay host rock reduces the need for such strengthened engineered barriers. Nevertheless, Nagra, in collaboration with a range of Japanese partner organisations, has evaluated a range of concepts for optimisation of TRU EBS design.

At the most fundamental level, the role of external bentonite layers to enhance performance of disposal silos / caverns has been re-assessed. Highly compacted bentonite can form an effective diffusion barrier which additionally provides mechanical protection (due to its plasticity) and acts as an effective colloid filter. Potential degradation scenarios must be considered, however, such as bentonite alteration by hyperalkaline cement / concrete leachates, disruption by repository-generated gas and erosion by flowing groundwaters. An alternative approach which can also ensure diffusive release from the emplaced waste is the use of a high permeability "hydraulic cage". Although the colloid filtration function is lost, this option may prove much more robust with respect to chemical or physical perturbations and be much easier (and cheaper) to actually implement in a quality assured manner.

The choice between very high and very low permeability barriers depends very much on the detailed characteristics of the waste and the host rock. Here it is important to consider not only long-term performance under expected evolution, but also perturbation scenarios and the practicalities of implementation.

Apart from segregation of wastes between caverns / silos with different barrier systems, conceptual studies have also highlighted advantages of structured placement of different types of waste within a single disposal area (Figure 2). Such designs are only practicable for the case that inventories are well established and standardised emplacement packages are used. Although the performance assessment modelling is made more complex, this may be more than compensated by the great increase in performance resulting from using less problematic wastes as additional barriers for the most safety-critical TRU components.

Potential optimised TRU disposal design using a thick outer layer of gravel (high permeability and hydraulic conductivity) acts as a hydraulic cage around the inner part of the EBS.

Note: high toxicity waste (red) can be surrounded by lower toxicity waste (yellow) to increase diffusion distances for problematic nuclides.

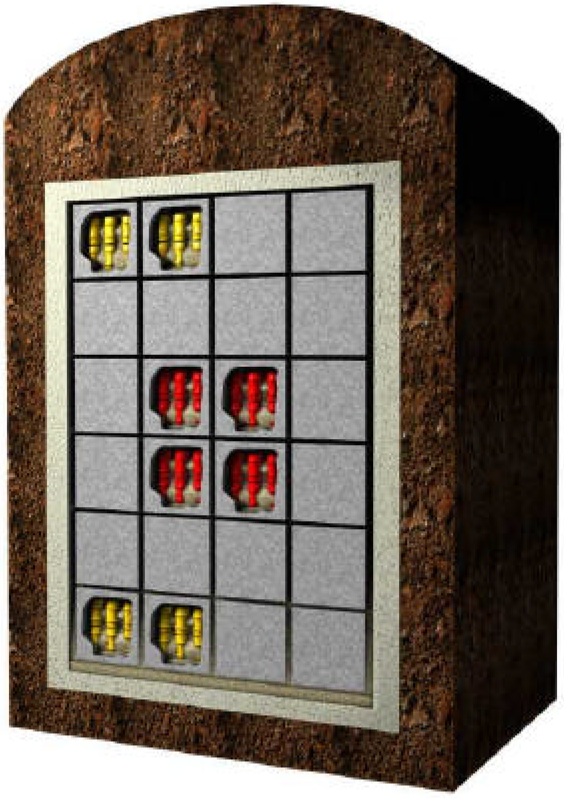

Fig. 2. Example of an optimised TRU disposal concept (collaborative study with Obayashi, Japan [2])

PERFORMANCE ASSESSMENT MODELS AND DATABASES

In Switzerland, there is no regulatory time cut-off and hence, theoretically, safety has to be demonstrated for all time. In practice, performance assessment calculations are carried out to the point where the dose maximum has passed – for deep geological disposal of HLW / TRU this being ~10^7 years. Shading on graphical presentations of performance assessment results and associated discussion emphasises that results for very long times (>~10^5 - 10^6 years) must be considered as illustrative only. Although the total radiotoxicity of TRU is much lower than that of HLW / SF, the key nuclides and the assessment timescales are effectively the same.

Compared to either HLW or SF, the performance assessment model chain components (near field, far field and biosphere) are similar and require the same input from site geological synthesis and associated models (e.g. groundwater fluxes and flow system details, groundwater chemistry, etc.). In detail, however, the near- and far-field models are more complicated for TRU.

Compared to the very simple case of vitrified HLW (see, for example, [3]), the assessment of the TRU disposal system has to consider:

Larger physical size

- Many more components and materials
- Heterogeneity within / between waste categories
- Chemical system far from equilibrium with host rock
- Many potential sources of gas
- Many potential sources of organics / substrates for microbial growth
- Many actual (through corrosion) and potential (chemical gradients) sources of colloids
- Large number of scenarios for mechanical / chemical degradation
- Higher concentrations of "problematic" nuclides (long-lived / anionic speciation).

Of course, many of these TRU characteristics are common also to L/ILW, but the relatively low toxicity and the short half-life of most components in the latter allow a much simpler approach to performance assessment to be justified.

The differences in the far-field models used for HLW and TRU are less extreme but nevertheless challenging – resulting predominantly from the relative complexity of chemical leachates from TRU and their potential for interaction with – and alteration of – the host rock.

Over the last 2 decades, the models available to evaluate TRU performance have improved dramatically, resulting from improved system understanding, more extensive databases and better computational tools. Such performance assessment models require an infrastructure of an integrated programme of laboratory studies, field experiments, natural analogue projects and research (process) model development in many key areas. Due to the generic nature of much of the work involved, such studies have formed a natural focus for many international collaboration projects.

A good example which illustrates both the need for an integrated R&D programme and the potential for international collaboration is provided by work aimed to support assessment of the consequences of the interaction of hyperalkaline cement leachates and the repository host rocks.

Cement / concrete leachate is initially dominated by Na/K OH, giving rise to pH values >~13. Thereafter, leaching of portlandite ($Ca(OH)_2$) and then a range of CSH phases maintains hyperalkaline (pH >~11) conditions for a much longer time period. The evolution of pH depends on the waste, repository design and host rock but models indicate the duration of highly alkaline conditions could extend through performance assessment relevant timescales ($>\sim10^5$ years). Models of cement leaching are complemented by static and dynamic laboratory tests and analogue studies of archaeological cements / concretes.

The minerals within potential host rocks and, more particularly, those in flow paths which contact advective groundwater, are thermodynamically unstable at such very high pH values and will undergo alteration. Chemical thermodynamic models can give indications of the direction of such alteration (e.g. [4]), but are greatly constrained by the limitations of databases for high pH conditions and the poor availability of kinetics data. Such limitations are critical as illustrated by the common formation of very long-lived metastable phases under relevant, low temperature (<50 °C) conditions.

A large number of laboratory studies have been carried out to extend databases and test models – both for single minerals and whole rocks (e.g. [5]; [6]; [7]). Slow reaction kinetics mean, however, that more realistic flow-through systems are often not feasible and reaction must be artificially enhanced in batch reaction systems using either finely crushed material (increased available surface area) or increased temperature (e.g. [8]; [9]). In the few cases where long-term experiments are run in the laboratory, maintaining the required high pH is a considerable technical challenge.

An alternative to laboratory studies, which is both more realistic and less vulnerable to perturbations over long timescales, involves tests in underground test facilities (Figure 3). At present, such tests are running as international collaborations in both underground test sites in Switzerland – at Grimsel in the crystalline rock of the Swiss Alps (Project HPF; see www.grimsel.com) and Mont Terri in the Opalinus Clay of the Jura Mountains (Project CW; see www.mont-terri.ch).

To further extend the timescales, a number of studies have examined the interface between archaeological concrete structures and surrounding rock (e.g. see the reviews of [10]; [11]). The advantage of longer timescales is generally balanced, however, by less relevant geological settings and less well defined boundary conditions.

To approach the timescales of performance assessment directly, the only possible option involves the study of natural analogues. The particular case of Maqarin in Jordan, where spontaneous combustion of a bituminous marl has provided a natural deposit of cement minerals (see [12]), is an unusually close simulation of typical TRU repository conditions (Figure 4). Over a period of ~10 years, this project has allowed some of the processes assumed to be associated with hyperalkaline plume development to be confirmed by observations (e.g. sealing of flow porosity by secondary minerals; presence of microbes, etc.) and allowed the strengths and weaknesses of more detailed models (e.g. predictions of sequences of secondary minerals) to be determined directly (for details see [13]; [14]; [15]).

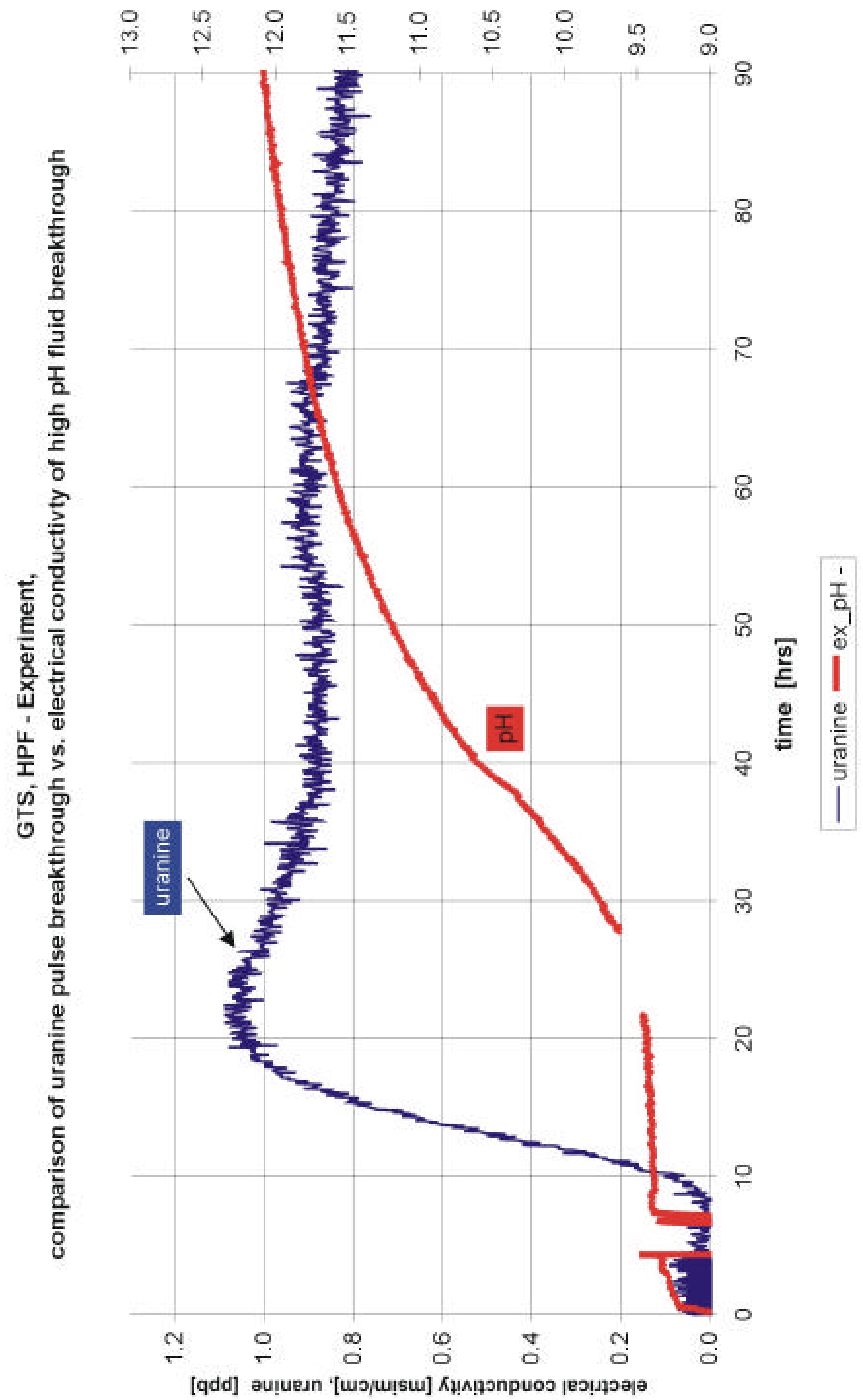

Fig. 3. **Example of results from studies of hyperalkaline plume migration in fractured crystalline rock from the Grimsel test site. Here, hyperalkaline cement leachates are injected into a fracture and interactions between the leachates and the host rock are studied. This plot represents breakthrough compared to the conservative tracer uranine. The "HPF" project is a collaboration between Nagra (CH), ANDRA (F), US-DOE / Sandia National Laboratories (USA), JNC (J) and SKB (S)**

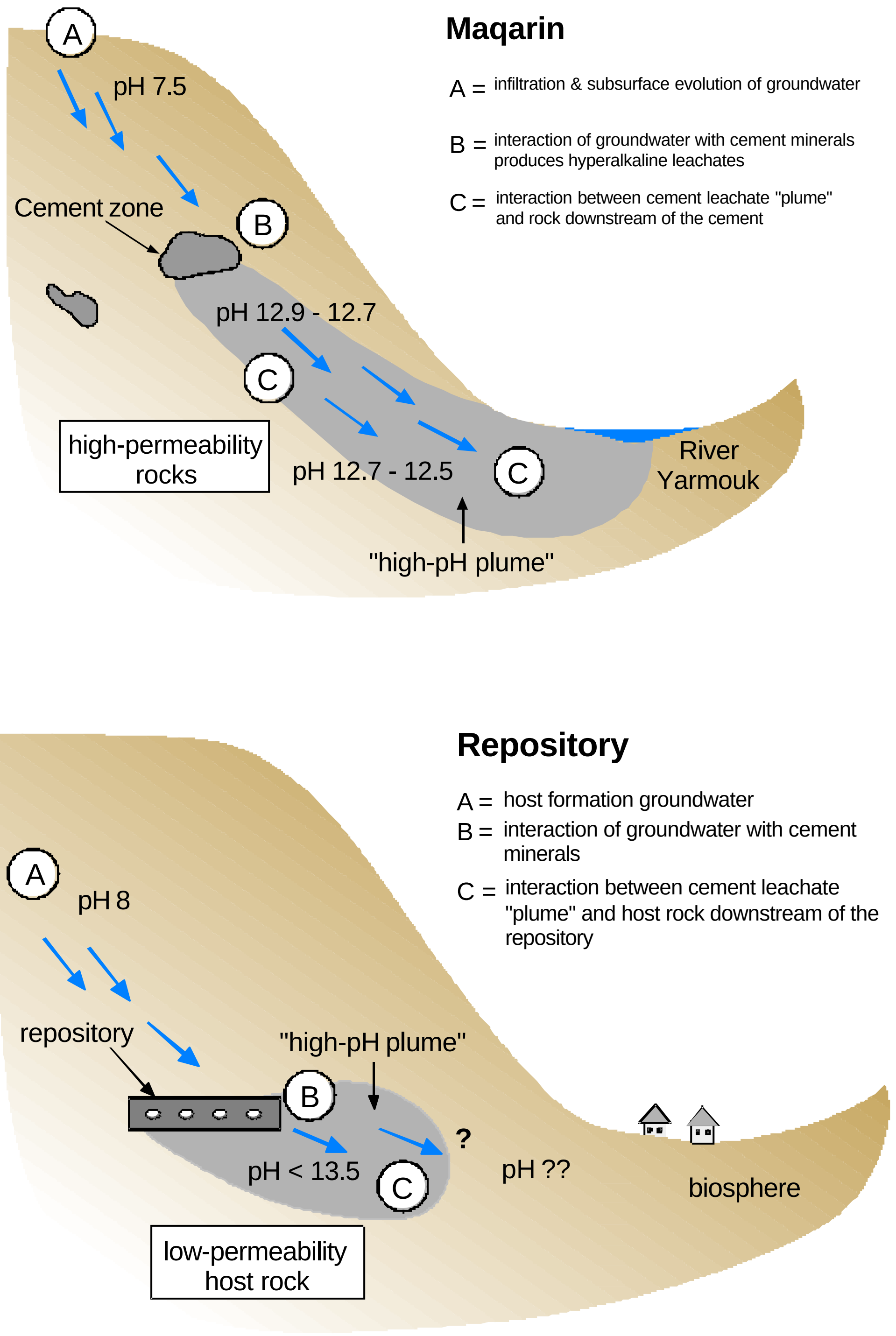

Fig. 4. Shematic outline of hyperalkaline plume development under natural conditions at Maqarin and as expected in a potential repository site. The Maqarin project is a collaboration between Nagra (CH), ANDRA (F), SKB (S), CEA (F) and Nirex (UK). JNC (J) have recently joined the group and Ontario Hydro (CDN) was previously involved.

PERFORMANCE ASSESSMENT RESULTS

The full performance assessment of TRU disposal within the EN project is almost complete and will be published by the end of 2002. Preliminary results are consistent with other studies (e.g. Japanese 1st TRU Progress Report – [1]) in that the final calculated releases to the biosphere are dominated by long-lived, anionic, fission products which are concentrated in only a restricted number of waste types. Indeed, the perspective provided by EN – for co-disposal of HLW, SF and TRU – clearly illustrates that a few mobile nuclides result in the dominant releases from the wastes. These dominant releases are seen from directly disposed spent fuel and it is clear that reprocessing only moves them into the TRU waste stream. It must be emphasised, however, that the radiotoxicity of such nuclides is very low and resultant doses lie well below regulatory guidelines. Their apparent significance can be attributed to the very high efficiency of deep repositories to completely retain more toxic radionuclides for geological periods of time.

CONCLUSIONS

There is now good evidence that many deep geological disposal options could safely contain TRU wastes. Nevertheless, the complexity and heterogeneity of this waste type and its content of long-lived radionuclides make development of a convincing safety case particularly challenging. Options exist both to strengthen the EBS and / or make it more robust and also to improve the sophistication of the safety case assessment and increase confidence in the results via more observation data from the laboratory, in situ studies and natural analogues. As much of the work involved is of general interest to many national programmes, existing international collaboration in this area can be expected to continue or even expand in the future. A potential future topic for future collaboration would be long-term demonstration projects in underground test sites (possibly lasting decades, using real waste) which would focus on what is probably the major challenge in nuclear waste management – public acceptance.

REFERENCES

[1] TCO (TRU Coordination Office), 2000: Progress Report on the TRU waste disposal concept in Japan, JNC / FEPC report, JNC TY 14000 2000-002, TRU TR-2000-02

[2] McKinley, I.G., Kawamura, H., Neall, F.B., Ando, K. (in press): Repository Design Optimisation for Long-Lived ILW; Sci. Basis Nucl. Waste Manag., **24**.

[3] Alexander, W.R. and McKinley, I.G. (1999): The chemical basis of near-field containment in the Swiss high-level radioactive waste disposal concept in Chemical containment of wastes in the geosphere (eds R. Metcalfe and C.A. Rochelle), Geol. Soc. Spec. Publ. No. 157, 47-69.

[4] Steefel, C.I. and Lichtner, P.C. (1994): Diffusion and reaction in rock matrix bordering a hyperalkaline fluid-filled fracture. Geochim Cosmochim Acta, 58, 3595-3612.

[5] Chermak, J.A. (1996): Interactions of the Palfris and Opalinus Shales with high pH NaOH and KOH solutions at 75°C. Unpublished Nagra Internal Report. Nagra, Wettingen, Switzerland.

[6] Adler, M. (2001): Interaction of claystones and hyperalkaline solutions at 30oC: A combined experimental and modelling study. PhD Thesis, University of Berne, Berne, Switzeland.

[7] Adler, M., Maeder, U.K. and Waber, H.N. (1999): High-pH alteration of argillaceous rocks: Experimental study. Schweiz. Mineral. Petrogr. Mitt. 79, 445-454.

[8] Chermak, J.A. (1992): Low temperature experimental investigation of the effect of high pH NaOH solutions on the Opalinus Shale, Switzerland. Clays and Clay Minerals 40: 650-658.

[9] Chermak, J.A. (1993): Low temperature experimental investigation of the effect of high pH KOH solutions on the Opalinus Shale, Switzerland. Clays and Clay Minerals 41: 365-372.

[10] Thomassin, J.H. and Rassineux, F. (1992): Ancient analogues of cement-base materials: Stability of calcium silicate hydrates. App. Geochem. Supplementary Issue, 1, 137-142

[11] McKinley, I.G. and Alexander, W.R. (1992): A review of the use of natural analogues to test performance assessment models of a cementitious near field. Waste Manag. 12, pp253-259.

[12] Khoury, H.N., Salameh, E., Clark, I.D., Fritz, P., Bajjali, W., Milodowski, A.E., Cave, M.R. and Alexander, W.R. (1992): A natural analogue of high pH cement porewaters from the Maqarin area of Northern Jordan – I: Introduction to the site. J. Geochem. Explor. 46, pp117-132.

[13] Alexander, W.R. (ed), (1992): A natural analogue study of cement buffered, hyperalkaline groundwaters and their interaction with a repository host rock – I: Definition of source terms. Nagra Technical Report Series NTB 91-10, Nagra, Wettingen, Switzerland.

[14] Linklater, C.M. (ed), (1998): A natural analogue study of cement buffered, hypedralkaline groundwaters and their interaction with a repository host rock – II. Nirex Science Report S/98/003, Nirex, Harwell, UK.

[15] Smellie, J.A.T. (ed), (1998): A natural analogue study of cement buffered, hyperalkaline groundwaters and their interaction with a repository host rock – III: Further definition of the hyperalkaline plume. SKB Technical Report TR 98-04 (Vol. I and II), SKB, Stockholm, Sweden.