

Key natural analogue input required to build a safety case for direct disposal of spent nuclear fuel in Japan

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Abstract Natural analogues have been previously used to support the safety case for direct disposal of spent nuclear fuel, but the focus of such work was very dependent on the key barriers of specific national disposal concepts. Investigations of the feasibility of such disposal in Japan are at an early stage but, nevertheless, it is clear that building a robust safety case will be very challenging and would benefit from focused support from natural analogue studies—both in terms of developing/testing required models and, as importantly, presenting safety arguments to a wide range of stakeholders. This paper identifies key analogues that support both longevity and spread of failure times of massive steel overpacks, the effectiveness of buffering of radiolytic oxidants and the chemical and physical mechanisms retarding release of radionuclides from the engineered barriers. It is concluded that, for countries like Japan where performance needs to be assessed as realistically as possible, natural analogues can complement the

existing laboratory and theoretical knowledge base and contribute towards development of a robust safety case.

Keywords Deep geological disposal · Steel corrosion · Canister failure · Redox buffering · Radiolysis · Realistic assessment

1 Introduction

Direct disposal of spent nuclear fuel (SNF) is considered in the countries in which this material is considered as a waste, rather than a potential resource for recycling. Natural analogues (NAs) have been previously used to support development and presentation of the safety case for such an option in several advanced repository programmes. It is clear, however, that the focus of such analogue studies has been very dependent on the weighting placed on different safety barriers in specific national disposal concepts.

Thus, the Swedish and Finnish “KBS-3” concept depends very much on the barrier provided by a thick copper canister and hence NAs focus on Cu corrosion (e.g. native copper found in relevant geological settings, Marcos 1996, and the Kronan cannon as an example of massive copper-rich alloy surrounded by clay, Hallberg et al. 1987) and the longevity of the surrounding bentonite (e.g. considering the impact of the early thermal period, Reijonen & Alexander 2015). Because of the requirement to show low risk of rapid corrosion during the perturbed chemical conditions that could result from deglaciation, this has also been a topic for analogue studies.

The safety case is quite different for the Yucca Mountain site (USA), in which disposal would have occurred above the water table. To strengthen safety arguments, NAs of metal corrosion and uranium ore leaching under unsaturated conditions were examined using archaeological analogues at

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Santorini and U mineralisation in a similar geological setting at Peña Blanca (Murphy and Percy 1994).

A further contrast is provided by a proposed SNF repository in the Opalinus Clay of Switzerland, where the safety case is predominantly based on the massive diffusion barrier presented by the host rock. Here NAs have been used to build arguments that demonstrate that diffusion controls solute transport (Mazurek et al. 2008) and that this characteristic will not be perturbed by the presence of the repository (e.g. as a result of high pH fluids from the leaching of concrete, Alexander 1994).

Until recently, Japan was committed to a policy of 100 % reprocessing of spent nuclear fuel as part of a philosophy of recycling to minimise reliance on imported resources (JNC 2000). As a result of this, the highest activity wastes considered for geological disposal are the vitrified high-level waste (HLW) and longer-lived intermediate-level waste (termed TRU in Japan) resulting from reprocessing. Now, however, options for direct disposal of at least some SNF are under consideration as part of a wider debate on the future of nuclear power in Japan (Kawamura and McKinley 2013a).

Although a reference concept for SNF disposal in Japan has yet to be established, like the H12 concept for HLW (JNC 2000), any design will probably need to include a robust engineered barrier system (EBS), which compensates for the limited credit that can be taken for the geological barrier, especially early in the siting process (Kawamura and McKinley 2013a). Because of potentially more corrosive groundwaters and less stable tectonic settings, demonstration of extremely long lifetimes of Cu canisters is very difficult and, indeed, might even be inappropriate when the potential timescales of uplift and erosion in Japan are considered (discussed further below). An inherently more robust option might be in-tunnel emplacement of smaller steel canisters, similar to the Nagra concept (Nagra 2002) and the H12 horizontal option (JNC 2000). Even more robust might be cavern disposal of large steel casks: basically the CARE concept (Masuda et al. 2004) modified for disposal of SNF, possibly together with HLW (Kawamura and McKinley 2013b). It can be noted that the practicality of the H12 vertical concept has not been demonstrated for HLW and hence this option is best excluded for the much larger SNF packages.

For disposal concepts like these, it would be expected that analogue evidence will be needed to support EBS options that can provide the key roles of:

1. Assuring long enough complete containment for significant reduction in the total radiotoxicity of the inventory (say ≈ 10 ka).
2. Providing temporal dilution of “instant release fraction” (IRF) radionuclides, which are released as a

pulse when loss of complete containment occurs (see e.g. Nagra 2002). These are isotopes of more volatile elements that accumulate at gap and grain boundaries during reactor operation and, for post closure safety; the critical radionuclide is usually I-129, but Cl-36 and Cs-135 may also be important.

3. Assuring redox buffering of radiolytic oxidant to justify slow fuel matrix leaching and low solubilities of key radionuclides. Such radiolysis is dominated by alpha-emitting radionuclides after containment failure and is much more relevant for SNF than HLW.

In addition, for a tectonically active country like Japan, the impact of uplift/erosion scenarios may need to be considered; even though these may be less dependent on the actual disposal concept, they are more significant due to the longer duration of high toxicity for SNF as compared to HLW (Nagra 2002).

The following sections will examine how existing information from NAs can provide input to the concept development process. In addition, for options that appear feasible, further NA studies required to support a robust safety case will be outlined.

2 Complete containment and spreading releases of the IRF

There is an increasing body of evidence supporting the low rate of corrosion of massive steel overpacks (OPs) under reducing conditions. Although in early HLW disposal studies OP lifetimes in the order of 1 ka were assumed (e.g. JNC 2000), the analysis supporting this work is highly over-conservative and, especially when analogue studies are considered, there are strong arguments for containment times in excess of 10 ka for thick-walled (ca. 20–25 cm), mild steel canisters (McKinley et al. 2012). This would be sufficient for assuring decay of shorter-lived, more toxic radionuclides, but would have negligible impact on long-lived isotopes like I-129 or Cs-135 (half lives ≈ 16 Ma and ≈ 2 Ma, respectively).

Although it is rarely emphasised, dilution and dispersion are generally the most important processes in assuring negligible doses from long-lived IRF isotopes. To provide such dilution, the distribution of failure times of OPs is actually more important than the minimum containment time.

To date, ranges of analogue corrosion rate data (e.g. Johnson and Francis 1980; Yoshikawa et al. 2003) have been interpreted only with the intention of selecting maximum values that are demonstratively conservative. Although there is certainly a degree of measurement uncertainty associated with the error values quoted (both in the corrosion depth and the time over which corrosion

occurred), it is also reasonable to assume that such ranges also represent true variability—as would be expected in terms of both the composition of the metal involved and the environment in which corrosion occurred. For the case of steel OPs or emplacement casks, compositional variation would be relatively small, but the variation in the chemical environment may be significant in a repository which could spread over an area of several km² and which will vary with time due to processes like uplift/erosion and sea level change (Miyahara et al. 2009).

In order to assess the potential impact of variability in corrosion rates, we have collected derived average corrosion rates from analogue measurements under relevant conditions and expressed these in Fig. 1 as times to failure for conventional OPs (requiring penetration of ≈ 10 cm: the failure time for massive casks would be 2–3 times longer). It is emphasised that such data must be used with care: measurement uncertainties are in the order of \pm a factor of about 5 and some of the higher corrosion cases may be from conditions that are low oxygen/anoxic rather than truly reducing. Nevertheless, the dataset shows a roughly lognormal distribution, which is not unexpected for the corrosion of buried iron (e.g. Sadiq et al. 2004). Potentially, data from other sources could have been added but, when studies are carried out explicitly as analogues, they often explicitly bias the measurements by selecting greatest corrosion thickness on samples with variable corrosion (e.g. as in Yoshikawa et al. 2003) where the extrapolated lifetimes would spread around 10–100 ka).

If such data are representative and if failing OPs are distributed randomly over the entire footprint of the repository, this produces very significant dilution of IRF releases in both space and time compared to the simplistic approach used in H12 (JNC 2000), where all OPs were assumed to fail simultaneously (potential concentration reduction around 5–6 orders of magnitude, depending on

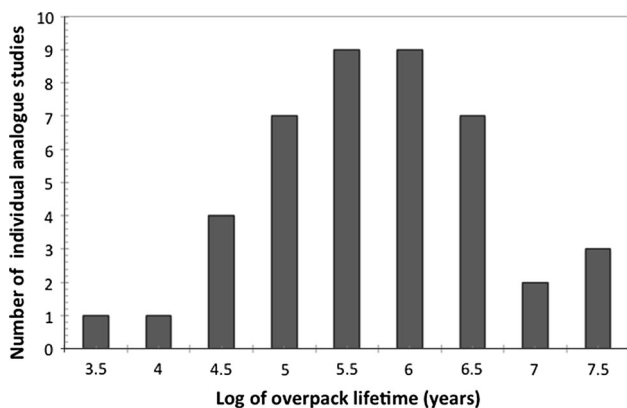


Fig. 1 Natural analogue corrosion data from Johnson and Francis (1980) used to derive a histogram of failure times for a thick-walled steel overpack

site characteristics). In reality, failure times may be grouped in particular repository zones with more similar chemical conditions, but even then release maxima would be considerably reduced relative to a simultaneous failure model.

It is important to note, however, that the lifetimes for ≈ 50 % of the cases would be >1 Ma, which is about the expected timescale of exposure of a repository in Japan due to uplift and erosion (JNC 2000; Miyahara et al. 2009). It would be anticipated that the corrosion rate would increase significantly as erosion brings a repository closer to the surface, but it might be worth considering if better performance—at least in terms of the IRF—might be obtained with a thinner OP that would ensure a spread of failures while the repository is still well below the surface (e.g. with a corrosion allowance of 1–5 cm).

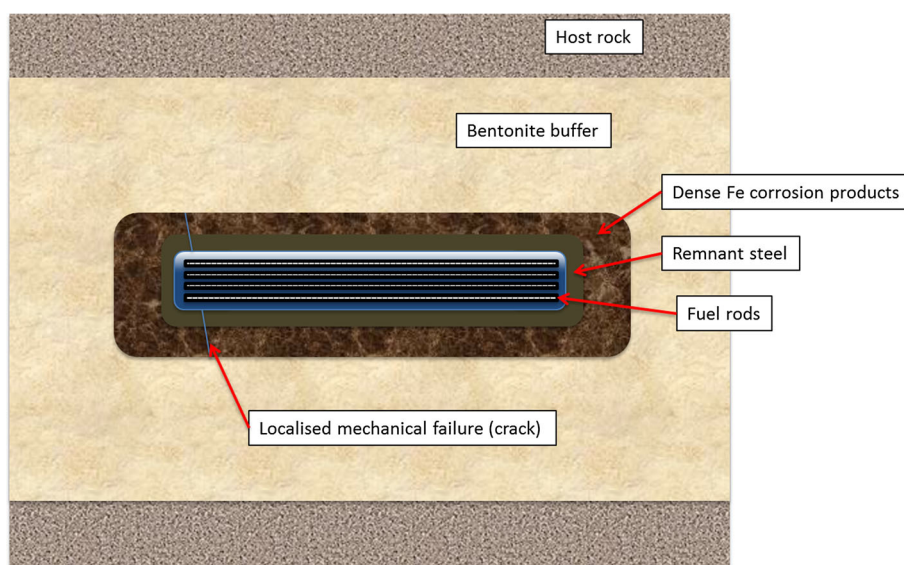
The impact of slow corrosion of massive steel containers on IRF releases is not, however, limited to the distribution of times of failure. As emphasised in McKinley et al. (2012), ignoring the barrier provided by the thick layer of massive corrosion products at the time of OP failure is grossly over-conservative. Even for poorly-sorbed IRF radionuclides, the transport resistance provided by diffusive transport through a failed OP within a bentonite buffer may significantly spread release fluxes (Fig. 2), although this has not been assessed to date.

It is assumed that mechanical failure (maybe at a weak point such as a weld) will allow water to penetrate the OP—but this will be a very slow process due to the low hydraulic conductivity of bentonite and any small crack(-s) which are likely to provide the only water access. Although no credit is taken for the fuel cladding or any other infilling material, it will take a significant time for all fuel within a single OP to come in contact with water. Indeed, corrosion of inner steel surfaces and cladding, alteration of fuel and radiolysis will consume water and produce hydrogen gas, further slowing contact with water and subsequent diffusion of IRF radionuclides to the failure point (a distance of up to 5 m). Taking credit for such processes might well spread releases from the outside of the buffer over periods of 1–10 ka.

Examination of Fig. 2 suggests that there is potential for increasing the extent of such release spreading by modifying the OP design to include some form of transport-resistant infill (normally infill is included with the main aim of improving resistance to crushing and hence provides only mechanical support—e.g. quartz sand). With the special aim of restricting water movement and, possibly, capturing I, some kind of clay/zeolite may well be a possibility worth considering.

In addition, Fe corrosion products have a renowned affinity for incorporation of trace elements and may incorporate even poorly sorbed/redox insensitive species into their structures. This is the basis for the use of Fe oxides

Fig. 2 Sketch of a failed steel overpack containing spent nuclear fuel. After failure, the overpack develops a thick zone of dense Fe corrosion products enclosing an inner zone of remnant steel



and oxyhydroxides in analytical techniques and environmental remediation (e.g. Laucht 2011). Although such scavenging can be considered as a form of co-precipitation, it may well be at least partially reversible, with trace species being released during Ostwald ripening, as initially amorphous/microcrystalline phases age into more stable macro-crystals. Nevertheless, the timescale of such aging may be in the order of thousands of years under ambient repository temperatures, providing another temporal dilution mechanism. To assess the credibility of this process, a simple mass balance is useful.

For the case of a simple steel overpack (typical mass ≈ 26 Mg) containing SNF (1.6 Mg), the inventories of I-129 (2.7 GBq) and Cs-135 (37 GBq) correspond to masses of about 300 g and 500 g, respectively. As the instant release fraction for I is about 10 % and that for Cs 5 %, in both cases the quantities released are at the ppm level compared to the mass of iron corrosion products. Although anionic species of iodine are generally considered to be poorly sorbed, uptake on iron oxides is well known (e.g. Couture and Seitz 1983) and, indeed, incorporation into Ga old iron formations at levels of 0.1–0.3 ppm have been reported (Cabral et al. 2011)—although the potential role of refractory organics here has to be clarified. Peaks of trace Cs associated with iron oxides forming at redox fronts have also been reported, with implied concentrations around 300–1000 ppm (MacKenzie et al. 1991; Akagawa et al. 2006).

Such considerations show significant potential for chemical processes that could spread the releases of key IRF radionuclides. However, it should be emphasised that such a comparison should not be over-interpreted: it shows only that the proposed mechanism is not unreasonable, but does

not give any good estimate of its impact under expected repository conditions.

From the summary above, it is clear that assuming that the OP provides no barrier roles after mechanical failure (apart from redox buffering—as in JNC 2000; Nagra 2002) is drastically over-conservative, especially for the IRF radionuclides which tend to dominate calculated releases from SNF. Although credible, developing assessments including spread of canister failure time, dispersion of releases from the near field and uptake of key radionuclides on corrosion products will require more support—in particular from NA studies.

3 Redox buffering within the failed OP

For a thick-walled OP, radiolysis is not a significant process until loss of integrity. Thereafter, when water contacts the fuel surface, α -radiolysis is particularly important. Because of the short range of α particles, the extent of this process is dependent on the area of surface contact between fuel and water. Although decay of shorter-lived α -emitters initially decreases the α activity of SNF, this is compensated at longer times by the ingrowth of α -emitting daughters in the actinide decay chains. Of particular significance is the ingrowth of U-238 daughters, at a rate set by the half-life of U-234 (about 250 ka).

The products of radiolysis depend very much on water chemistry, but can be simply represented as a reactive oxidant (e.g. hydrogen peroxide) and kinetically inert hydrogen, resulting in a net oxidising environment. Such radiolytic oxidant is a particular concern for SNF because the dissolution rate of the UO_2 matrix is very sensitive to redox

and may increase by many orders of magnitude if conditions become more oxidising (e.g. Mennecart et al. 2004). In the worst case, rapid dissolution of UO_2 may be coupled to precipitation of microcrystalline/porous secondary U (VI) minerals with higher specific surface areas, leading to further increase in the radiolysis rate in a form of autocatalysis. Low solubility of many radionuclides and potential redox buffering by remnant Fe in the OP may constrain radionuclide releases, but more rapid release of some mobile (e.g. I, Cs) or highly toxic radionuclides (e.g. Ra-226) cannot be precluded.

Because α -radiolysis is a surface process, strongly oxidising conditions may be very localised. The key question is, however, how these will be buffered to the reducing conditions found in the host rock. Several cases are, at least in principle, possible, with buffering:

- A. Very close to the fuel surface
- B. On/within OP
- C. Within the buffer
- D. Within the host rock

The consequences in terms of radionuclide releases may be dramatically different depending on which actually occurs or how these develop with time, as indicated in Table 1. The fluxes of radionuclides from the outside of the buffer would tend to increase $A < B < C \ll D$, with the final case having the additional complication that colloids are likely to form at the redox front and, if this is outside the buffer, the critical colloid filtration role of this barrier is short-circuited. For radionuclides with low solubility/high sorption, transportation in a colloidal phase could well increase resultant biosphere doses by many orders of magnitude (e.g. Gridrod 1993).

The situation is further complicated by the fact that redox buffering may either be inorganic or microbially catalysed. In the former case, at expected ambient temperatures, this requires a kinetically fast reductant to be supplied in solution [e.g. Fe(II)] or be present in an easily accessed solid phase. Microbially catalysed reduction can utilise a much wider range of substrates, including hydrogen gas and relatively refractory minerals (e.g. pyrite).

Indeed, utilisation of hydrogen as a reductant might assure that buffering is credible even in case A—utilising hydrogen produced from both radiolysis and anaerobic corrosion. However, this requires that active microbial populations exist within the buffer—which has been argued not to be the case, even though the supporting information base is rather ambiguous (Motamedi et al. 1996). Indeed, this issue is particularly complex as, while the existence of active populations within the EBS is hard to demonstrate for conditions in the far future, laboratory experiments carried out to examine “inorganic” redox buffering can rarely be proven to be sterile due to the ubiquity and robustness of extremophile microbes (West and McKinley 2002).

As has been previously noted (McKinley et al. 2012), developing arguments for the longevity of steel OPs tend to result in difficulties in assuring that they perform an effective redox buffering role. Because the systems involved develop only over millennia, the extent to which this problem is accessible with laboratory studies is inherently limited. Here again, a possible engineering solution might involve infilling the OP with some form of reactive iron mineral.

4 Communication

As discussed further elsewhere (Alexander et al. 2015), communication of complex arguments within a repository safety case is a key role of NAs. Most of the aspects noted above are, however, very technical and probably not very suitable targets for NA communication material. For the specific case of SNF, an issue where treatment is strongly linked to public perception involves uplift and erosion. It was previously mentioned that, for typical Japanese site conditions, direct exposure via denudation may occur within a period of about 1 Ma. In terms of public (and regulatory) concerns, this represents the distant future when it is not sensible to apply quantitative constraints or performance requirements. Nevertheless, the long duration of the toxicity of SNF and the potentially low rate of

Table 1 Impact of different redox buffering cases

Case	Rate of matrix dissolution	Initial release of radionuclides	Gradient within OP	Gradient over bentonite	Comment
A	Low	Low	Low	Low	Depends on effective, fast buffering
B	High	High	High	Low	
C	High	High	Variable	Variable	
D	High	High	Medium	High	Possible colloid formation at redox front

The cases are: *A* very close to the fuel surface, *B* on or within the steel overpack; *C* within the bentonite buffer, *D* within the host rock (see Fig. 2)

Table 2 Issues highlighted in this paper and potential NA support for required laboratory and modelling studies

Issue	Lab data requirements	Modelling requirements	NA requirements
Distribution of failure times for thick steel OP	Long-term (multi-decade) data under relevant conditions	Extension of release models to incorporate distribution of OP failures in space and time Assess potential increased performance for thinner wall OP variants	Re-analysis of archaeological analogue data to derive failure distributions Focused sampling of artefacts to determine potential role of environment and composition on corrosion rate
Internal IRF radionuclide transport within a failed OP	Scale model multi-decade simulations (maybe in URL)	Development of a 3D model for scoping calculations	Analysis of fractures in old steel containers or well castings (if any can be found)?
Uptake of key radionuclides by corrosion products	Accelerated, small-scale experiments on scavenging, Ostwald ripening	Development of a 3D model for scoping calculations	Re-analysis of analogue data on trace elements at redox fronts, possible sequential extraction of existing/new samples Data mining on trace element analysis on Fe oxides of different age Possible sampling of new sites where Fe oxides may contact relevant elements
Modify design to improve internal OP transport resistance	Lab studies of potential infilling materials (e.g. zeolites)	Extend scoping 3D model to allow the impact of such materials on performance to be assessed	Focused literature search of possible natural analogues of relevant materials
Extent of radiolysis at SNF surface	Re-assessment of existing experimental data: check for relevance to real systems	Extension of existing bulk surface models to consider partially saturated environments	Assessment of surface redox conditions in deep, rich ore bodies?
Mechanisms of redox buffering in relevant systems	Re-assessment of experimental data to rigorously determine extent of inorganic/microbially catalysed reactions	Extension of thermodynamic models to better represent kinetic constraints (including mass balances and supply of nutrients to microbes)	Focused analysis/new analogue studies of relevant redox fronts (especially within clay/in deep geological settings)
Modify design to improve internal OP redox buffering	Lab studies of potential redox buffers (e.g. Fe(II) minerals)	Extend scoping 3D model to allow the impact of such materials on performance to be assessed	Literature search of possible natural analogues of relevant materials
Development of uplift/erosion assessments for SNF	Accelerated/small scale simulation of SNF package nearing the surface	Development of new models to realistically represent system evolution in 3D	Synthesis of NA information to communicate timescales until exposure Focused NA to assess impact of erosion of a rich ore body

IRF instant release fraction, *NA* natural analogue, *OP* overpack, *SNF* spent nuclear fuel, *URL* underground rock laboratory

degradation of the UO₂ matrix (especially if redox is buffered in reducing conditions) could result in nominal doses far above standard regulatory guidelines. As this is the type of result that can be seized upon by opponents, it is important to have strong arguments for low impact of even such extreme scenarios.

As has been shown for other countries where erosive exposure cannot be excluded [e.g. Switzerland, after about 8 Ma for the reference repository in Opalinus Clay, Nagra (2002)], an eroding SNF repository is similar to a very rich U ore body lying close to the surface. If conventional

biosphere models used in performance assessment are applied, calculated doses are not only far above regulatory guidelines (McKinley and Chapman, 2009), but also above the natural radiation background. Strict application of such guidelines would thus prevent disposal of SNF almost anywhere in Japan—but in addition, if applied consistently, also prevent most other disposal activities (e.g. shallow low and intermediate level radioactive waste disposal, naturally occurring radioactive material, chemotoxic waste). In order to discuss such scenarios sensibly, therefore, it is important to use analogues to put the topics involved in context. This

can range from improving appreciation of what exactly a timescale of 1 Ma means for Japanese populations to more detailed understanding of the hazards from an eroding ore body.

5 Conclusions

In this paper, a number of important issues associated with the development of a safety case for the disposal of SNF in Japan have been identified. As indicated in Table 2, there is certainly useful input that can be derived from focused laboratory and modelling studies, but these will also require support from NAs—either in terms of re-assessing existing data, re-analysing appropriate samples or initiating tailored projects to fill gaps in knowledge.

Because of the active debate on the future of reprocessing in Japan, an assessment of the feasibility of SNF disposal is urgent, but it should be carried out as rigorously as possible. Demonstration of safety may be more challenging than in other countries, which requires development of a safety case that is as realistic as possible—avoiding over-conservatism, but clearly acknowledging any critical uncertainties. As illustrated in this paper, NAs may contribute both to developing a suitable disposal concept and also to defence of the resulting safety arguments.

The NA studies proposed will certainly be challenging, but similar problems may be faced by countries with similar geological settings (e.g. Korea, Taiwan) or those with a volunteer approach to siting (e.g. UK, Canada, possibly USA in the future) and hence it may be a good focus for international cooperation.

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