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Disposal of low- and intermediate-level waste in Switzerland: Basic aspects of potential relevance to microbial effects

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Summary. Current projects for the disposal of low- and intermediate-level radioactive waste in Switzerland are based on the concept of a horizontally accessed repository under a hillside. Most of the waste to be disposed of in such a repository is operational and decommissioning waste from nuclear power plants and wastes from medicine, industry and research. This waste is generally solidified in cement and placed in steel drums or concrete containers. Once the repository caverns have been filled with waste, they will be backfilled with a porous mortar to allow gases, produced by the anaerobic corrosion of steel and by microbial degradation of organic material, to escape from the near field. Valanginian marl, which is one of three envisaged options for the host rock, is characterized by a high carbonate content, up to 75% in some locations. The organic content of the marl is between 1 % and 2 %, while pyrite is present in concentrations up to 5%. The groundwater is reducing, and its pH tends to lie in the neutral to slightly alkaline range. Potentially important microbial effects on the long-term performance of the system are microbial degradation of barrier materials and organics, the effect of microorganisms on sorption, and their role as catalysts.
Key words. Nuclear waste; microbial effects; cement; marl; ion exchange resins.

General repository design

The present disposal concept in Switzerland calls for two repositories – a type 'C' repository for high-level waste (HLW) and some of the longer-lived components of intermediate level waste (ILW) and a type 'B' repository for the less radiologically significant low/intermediate-level waste (L/ILW). The criterion used to define the limitations for ILW nuclide inventories acceptable for the type B repository will be defined on the basis of the characteristics of the site eventually selected. The current priority for Swiss projects is to find a suitable site for the large volumes of LLW and short-lived ILW; the lower volumes of long-lived ILW are less urgent and can, in principle, be handled along with the HLW if necessary.

The present paper focuses on the L/ILW repository, for which the reference concept is characterized by the following:

- Disposal in underground rock caverns with access through horizontal tunnels. The reception area is also underground.
- A system of engineered safety barriers comprising the waste solidification matrix (cement, bitumen, polymers) inside drums (steel); possible grouting of the waste drums with liquid cement in a concrete container; backfilling of remaining empty space with special concrete; concrete lining of the disposal caverns and sealing of access tunnels on closure of the repository. The waste is delivered in conditioned form, i.e., solidified in drums. All remaining engineered barriers are provided during construction, operation and closure of the repository.
- There exists the possibility of dividing the waste into several toxicity classes in order to maximize the barrier potential of the repository, e.g., by placing waste with higher toxicity in caverns with better barrier properties.

One possible basic configuration for the L/ILW repository is shown in figure 1. A curved access tunnel path has been incorporated into the design to prevent short-cuts of direct groundwater flow from the repository caverns to the biosphere.

For Swiss projects, three potential host rocks have been identified: Valanginian marl, crystalline schists/gneisses and anhydrite. This paper will consider marl only, firstly because it has a high priority at present and secondly because more reliable data are available for this formation.

In common with the approaches in most countries, the Nagra concepts for the disposal of highly radioactive wastes and for the disposal of low-/intermediate-level wastes in Switzerland rely on natural barriers (host rock formation and overlying geological formations) as well as engineered (man-made) barriers. The paper deals first with the waste itself and its solidification matrix, then the other engineered barriers, and finally the natural barrier Valanginian marl. It emphasizes those aspects considered to be of relevance to microbial effects.

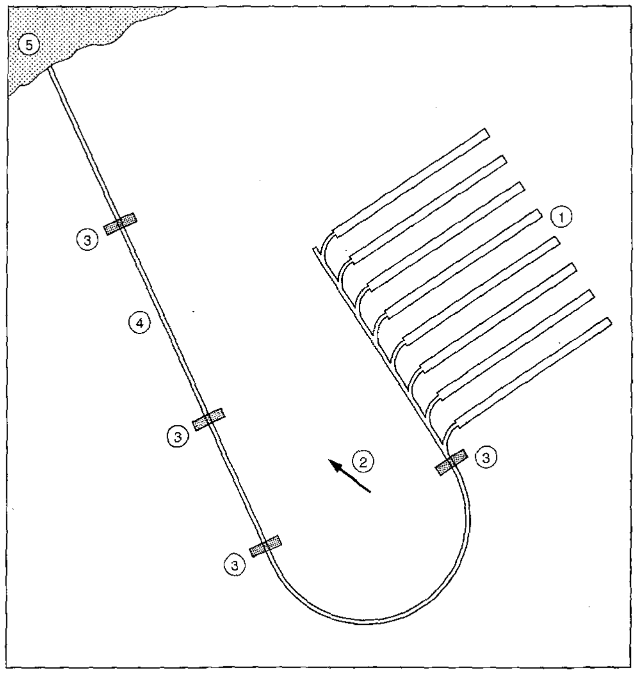

Figure 1. A possible configuration for a Swiss L/ILW repository with horizontal access tunnel (adit). 1) Disposal caverns; 2) Direction of groundwater flow; 3) Plugs restricting groundwater flow through the (loosely backfilled) adit (4); 5) Adit mouth. It should be noted that the overburden above the adit increases from zero in the top left-hand corner to typically several hundred meters in the bottom right-hand corner.

Waste inventory and waste form

General

In this section the waste inventories and waste forms used or envisaged for low-level and short-lived intermediate level waste will be briefly described, and orders of magnitude for the total repository activities are given. According to present planning for nuclear waste disposal in Switzerland, long-lived intermediate-level waste (mainly waste arising from spent-fuel reprocessing) will be disposed of either in deep caverns specially designed in the same geological formation as the other L/ILW, or in another facility. This type of waste will, therefore, not be further discussed in this paper. Further information on all types of wastes can be found in references 1 and 2.

Operational waste

Approximately 80% by volume and over 90% by activity of solidified operational waste consists of ion-exchange resins solidified in a cement (or, in a few cases, polymer) matrix contained in 200-l steel drums. Typically, the ion-exchange resins consist of polystryrene skeletons crossed-linked with divinyl-benzene-sulphite (cation exchanger, acidic, about 70% of the total), or divinyl-benzene-trimethylamine (anion exchanger, basic, about 30% of the total). The cement matrix is, in general, a pozzolanic cement, i.e., a cement with a much smaller concentration of free portlandite ($Ca(OH)_2$) than ordinary Portland cement, because of the addition of highly reactive silica fume or fly ash. Thus the pH in the cement pore-water would be expected to lie within the range 10.5–12, compared to values between 12.5 and 13.5 for an ordinary Portland cement. The 200-l drums containing the solidified waste are made of plain sheet carbon steel. Other operational wastes are typically concentrates and slurries (with cement or bitumen as a matrix), filters, non-incinerable solid waste, incinerated waste and fuel elements casings, all with cement as a matrix.

The total beta/gamma activity of these wastes will be typically in the order of 10^{17} Bq and be dominated initially by relatively short-lived nuclides such as Cs-137, Sr-90, Co-60 etc. The alpha activity will be many orders of magnitude lower, in the order of 10^{11} Bq.

Decommissioning waste

By contrast to operational waste, decommissioning waste is spread evenly among several waste types, for all of which a cement solidification matrix as well as large concrete containers (total volume 22 m^3) are currently foreseen. It should be pointed out, however, that a final choice of materials and sizes will only be made when the nuclear power plants in Switzerland are decommissioned, i.e., according to present planning not until well into the next century. Although the waste volumes will be spread among several types, the activity is largely concentrated in waste from those parts of the reactor which, during the plant operation, were subjected to a significant neutron flux. These comprise the reactor pressure vessel and pressure vessel internals (high strength ferritic steel) and the biological shield (baryte concrete). In addition, there is so-called contaminated secondary waste (resins, concentrates etc.) arising from the cleaning/dismantling of those parts. This implies that the main nuclides will be steel and concrete activation products such as Cl-36, Fe-55, Co-60, Ni-63 and Eu-152. The total beta/gamma activity will be, as in the case of the operational waste, approximately 10^{17} Bq. The total alpha activity will be in the order of 10^{10} Bq.

Low alpha reprocessing waste and waste from medicine, industry and research

Low alpha reprocessing waste is mentioned here for the sake of completeness: it arises from the operation of the reprocessing plants. In the standard waste form in the current Swiss disposal concept, this type of waste is to be solidified in cement inside asbestos-cement containers. While the total beta/gamma activity in this type of waste is significantly lower than that from reactor plant operational waste, the total alpha activity would be typically 10^{14} Bq. Although low in absolute value, this activity would represent more than 98 % of the alpha activity of the L/ILW repository.

Waste from medicine, industry and research institutions should also be mentioned here. The standard waste form is here again steel drums containing the waste solidified with cement. This waste is particularly heterogeneous, with a more variable and less precisely defined radionuclide inventory than the waste from nuclear power sources.

Total volumes

The total volume of the wastes discussed in this section is about 120,000 m^3, approximately 60 % of which is low-level waste.

Engineered barriers

General

As indicated in section 2, the total activity in a L/ILW repository would, for Swiss conditions, initially be approximately 1 % of that in the HLW repository. Furthermore, the activity concentration in L/ILW is generally considerably lower than in HLW. This implies that the engineered barrier system for L/ILW can be less conservatively designed to achieve an equivalent expected performance. In particular, overpacks are not required, and thus, for a large proportion of wastes, only two engineered barriers are foreseen: waste matrix and backfill. One requirement that is of importance for the choice of backfill material is that it must take into account the considerable quantities of steel present in the L/ILW (steel drums containing cemented operational waste, steel reinforcements in the decommissioning waste, etc.). This steel will corrode under anaerobic conditions, and therefore hydrogen gas will be produced by the concomitant reduction of water. In addition, microbial degradation of organic materials can produce gas – mainly methane and CO_2 (a microbial effect quantitatively considered in current safety analyses).

In order to ensure that this gas does not accumulate in the near-field (potentially leading to pressures of up to about 100 MPa), it is necessary to select a backfill material that has a sufficient gas conductivity. This, in turn, implies that the requirement for a hydraulic conductivity as low as possible must be relaxed. Depending on the type and toxicity of the waste and on the hydraulic conductivity of the host rock, it may be necessary to envisage a third barrier (hydraulic barrier) at the interface between backfill and host rock. Since this third barrier would, of necessity, have a low gas permeability, it may be necessary to provide (small) gas vents through the barrier at the top of the disposal cavern. This would result in the typical design for a repository cavern shown in figure 2.

Backfill

In Switzerland, as in Sweden, attention is at present focused on Portland cement/pumice and air-entrained mortars as potential backfill materials for L/ILW repositories in order to provide the required gas conductivity. As more data on the gas generation rates become available from corrosion and microbiology studies, it will be possible to optimize the mortars with respect to their hydraulic conductivity, sorption capacity, and flow

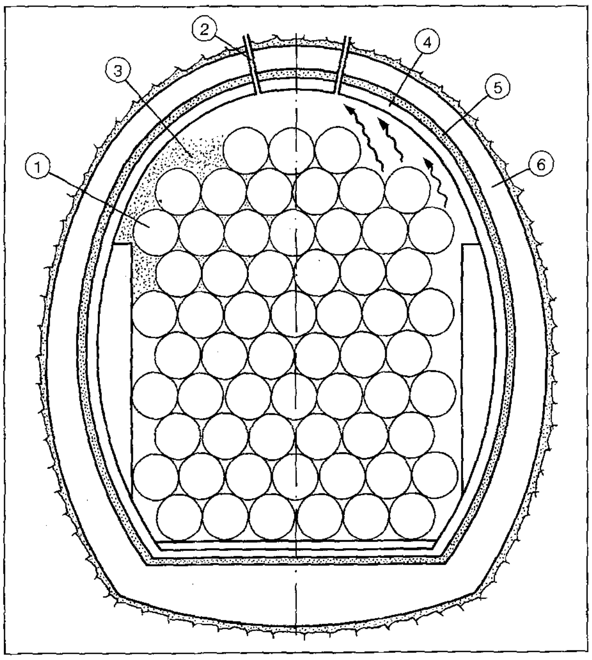

Figure 2. A possible design for a L/ILW disposal gallery. 1) Drums with cemented waste, generating gas at a (slow) rate, 2) Gas vents, 3) Porous backfill, 4) and 5) Outer barrier (only for certain types of wastes) here a 2-layer (concrete-clay) combination, 6) Gallery liner (for structural purposes only, no barrier function).

properties. Experimental programs are being implemented at the present time to achieve these aims.

A few points of relevance to microbial effects should be mentioned here. Firstly, a high gas conductivity also implies a high hydraulic conductivity, so that the backfill cannot be a significant water flow barrier. Because cementitious materials and aggregate materials have a good sorption capacity for many nuclides, the backfill will, however, act as a fairly good chemical barrier to solute transport. Secondly, in contrast to the waste matrix, where pozzolanic cements are widely used, the backfill will be based on ordinary or, more likely, on sulphate-resistant Portland cement. This implies a pH range above 12.5 for considerable periods of time (i.e., until all alkali hydroxides and portlandite have been leached out of the backfill by the groundwater). Thirdly, the organic content and content of other nutrient elements (e.g. N, P) of the backfill will be very low (organics will, however, probably be present in low concentrations, as a result of their being used as fluidizing additives).

Outer hydraulic and/or diffusion barrier

At the present stage of design, and pending detailed site investigations and site-specific safety analyses, it is expected that only a small proportion of the wastes (e.g. low alpha reprocessing wastes) will require an outer barrier. The design of that barrier will require long-term host rock creep data, since the barrier has to withstand significant stresses over hundreds of years to be effective. This means that the barrier material(s) can be selected and the barrier dimensioned only on a site-specific basis. Cementitious materials and smectite-type clays (as well as combinations of them) are expected to be prime candidates and engineering studies are being started at the present time.

Geosphere

Geological background

As indicated previously (section 1) the reference repository is located in a hillside within a marl formation. The overburden of rock will be at least several hundred meters. Due to the topography, the water flow path from higher areas of the hill will pass through the repository with eventual exfiltration in the neighboring valley, giving expected flow paths through the host formation on the scale of several hundred meters.

The marl itself is an agrillaceous rock with a high carbonate content (23–75%). The balance is mainly made up of clay minerals (e.g., kaolinite and illite) and quartz. Of importance from the point of view of microbiology is the organic carbon content, which lies between 1.2 and 1.7%. Also potentially relevant is the presence of pyrite, which may be a biologically usable reductant, at concentrations of 1–5%.

Waters within the marl are generally fairly dilute ($\leq$ 500 µS/cm electrical conductivity), with Ca, Na, Mg, HCO_3 and SO_4 the predominant components. Localized zones have, however, been identified with highly mineralized (electrical conductivity $>$ 1,200 µS/cm) NaCl waters. In both cases the waters tend to lie in the neutral to slightly alkaline pH range and, despite the presence of sulphate, would be expected to be reasonably reducing. Further details are given in reference 3.

Of considerable relevance is the finding of natural gas (mainly CH_4) in this formation. This gas may be biogenic and indicate low levels of continuing methanogenesis in this formation. The gas itself also means that, at least locally, the formation is not totally water-saturated.

The geosphere barrier

The general hydraulic conductivity of the marl is rather low and, in intact zones, very low water flow rates would be expected. The rock is fractured, however, and flow occurs predominantly in such fractures, and in large-scale disturbance (or shear) zones. After degradation of the near-field barriers and the release of nuclides to the far field, transport is assumed to occur in a network of fractures. During this transport, concentration of radionuclides will decrease due to dilution (in space and time) and decay. Of key importance for the safety assessment, therefore, is the nuclide transport path and time – the latter being affected by interactions between the dissolved radionuclides and the rock. Many such interactions are often empirically grouped under the collective term 'sorption', which includes all processes distributing radionuclides between the aqueous phase and the rock surface. Two further important factors contributing to retardation must, however, be distinguished: matrix diffusion and precipitation.

Matrix diffusion is the process by which nuclides diffuse from the main advective flow paths into the stagnant porosity of the bulk rock, where they may then be sorbed. This mechanism has been shown to be very important in that it greatly increases the volume of rock which can cause retardation of the migration nuclides. Precipitation may arise owing to changes in the water chemistry along the flow path which cause a decrease in the solubility of particular nuclides. The performance of the geosphere barrier is thus established by the hydraulic characteristics of the rock (defining the flow path and flow velocity) and the effciency of the various retardation mechanisms.

Safety analysis

Base case and perturbing factors

The basic description of the performance of the repository assumes leaching of the waste and subsequent transport of dissolved radionuclides through a well-specified geological system. Current safety assessment models of this system are kept as simple as possible and, by selection of conservative parameter values, a reasonably quantitative 'base-case' assessment can be carried out. Limitations in available data can be studied by parameter variations and uncertainty analysis of the base case. Additional factors could be significant which are not represented in current models and which, therefore, have not yet been fully evaluated. Of most current relevance are inorganic colloids, organics and microbes.

In geological systems it is generally difficult to separate these factors. The term 'colloid' as applied to groundwaters is simply based on size and includes a wide range of inorganic and organic material as well as microbes. The 'organic' and 'inorganic' components, indeed, are often intimately associated and do not reflect distinct populations. Organic carbon is, of course, an additional source for microbical life and a product of biological metabolism. Work, at present, is focussed on characterizing these components in the marl with a view to later evaluation of their role in nuclide release and transport.

Incorporation of microbiology into safety assessment

Many papers in the present multi-author review consider particular aspects of microbiology in a repository environment. On a global scale, the main emphasis within current Swiss safety analysis projects is to identify and quantify processes which could degrade repository performance. It is quite possible that the net effect of microbial activity will be to improve repository performance (e.g., by immobilizing radionuclides or ensuring favorable chemical conditions); in order to demonstrate safety, however, a high priority requirement is to show that unfavorable processes do not lead to unacceptable decrease in performance.

As indicated, the engineered barriers present a hyperalkaline, oligotrophic environment which, even if not sufficient to ensure sterility, will considerably constrain microbial activity. Although there is some indirect evidence of biological activity in the host rock, metabolic activity levels would be expected to be very low. Generally, therefore, the issue is one of defining the consequences of the presence of low numbers of chemolithotrophes.

Important processes which may be influenced by such activity, apart from the gas generation, are associated with barrier biodegradation and radionuclide release and transport. Biodegradation of various relevant materials is considered in the papers by Bock, Christofi, Langomazino, Roffey and Wolf (see Microorganisms in nuclear waste disposal, Part II, scheduled for publication in Experientia in 1991.). In terms of radionuclide release and transport, key factors are the extent of uptake by microorganisms (see e.g. the paper by Gadd, this issue), the interaction of such microorganisms with rocks (see e.g. the papers by Bar-Or and Shilo, Lappin-Scott and Costerton, this issue), degradation of organic materials in the waste and in the rock (potentially leading to increased concentrations of complexing agents) and the effects of the organisms and of their byproducts on radionuclide sorption (West et al., Microorganisms in nuclear waste disposal, Part II). Finally, the role of microorganisms as global geochemical catalysts should not be ignored – particularly their catalysis of slow redox reactions in the groundwater S, N and C systems – which can indirectly affect both barrier performance and radionuclide speciation/solubility.

1 Nagra: Project Gewähr 1985, Nuclear Waste Management in Switzerland: Feasibility Studies and Safety Analyses; Nagra Gewähr Report Series NGB 85-09, Baden (Switzerland), Nagra, 1985.

2 Nagra: Inventar und Charakterisierung der radioaktiven Abfälle in der Schweiz; Nagra Technical Report Series NTB 84-47, Baden (Switzerland), Nagra, 1984.

3 Nagra: Berichterstattung über die Untersuchungen der Phase I am potentiellen Standort Oberbauenstock; Nagra Technical Report Series NTB 88-18, Baden (Switzerland), Nagra, 1988.

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