

Consideration of microbiology in modelling the near field of a L/ILW repository

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Summary. The long-term safety of proposed repositories for nuclear waste is demonstrated by the use of chains of mathematical models describing the performance of the various barriers to radionuclide mobilisation, transport, release into the biosphere and eventual uptake by man. Microbial contamination of such repositories is to be expected, and hence the extent and consequences of microbial activity must also be quantified. This paper describes a modelling approach to determine the maximum microbial activity in the near field of a repository, which can thus be related to maximum possible degradation of performance. The approach is illustrated by application to a proposed Swiss repository for low- and intermediate-level waste (L/ILW), which is immobilised in concrete and emplaced in a marl host rock.

Key words. Nuclear waste repository; near field; modelling; microbial activity; nutrient limitation.

Introduction

As described in detail elsewhere², a repository system can be conveniently divided into the near field, which consists primarily of the waste itself and the engineered components of the repository, and the far field which includes the geological barrier and the biosphere. In a Swiss repository for low and intermediate-level waste (L/ILW), the near field includes the waste and its solidification matrix (cement, bitumen or resins), canisters or drums (concrete or steel) and repository lining (concrete).

In order to model the near field in a safety assessment, the performance of each of the near-field components needs to be quantitatively assessed and the resulting radionuclide release rate to the far field (the source term) evaluated. The performance of the various engineered barriers depends not only on the retention of their physical integrity but also on their chemical rôle of buffering water composition and retarding radionuclide mobilisation.

The standard near-field model

For repository performance assessment, a very much simplified representation of the near field is used. For example, in the Swiss case as described in Projekt Gewähr 1985 safety analysis (Nagra, 1985)⁵ no rôle as a physical barrier is assigned to the metal drums and a continuous advective flux of groundwater (or free diffusion) throughout the repository is assumed. Radionuclide release is not considered to be constrained by elemental solubilities, and retardation during transport is only considered for a few radionuclides which are assumed to be moderately sorbed. The degradation of cement/concrete structures is considered only in terms of their performance as hydraulic barriers and is assumed to occur in distinct steps. The production of gas, mainly from anoxic corrosion of metals, could potentially give rise to perturbation of near-field performance but effects could be minimised by

using gas-porous concrete formulations and, possibly, vent systems in the liner.

Current effort is focussed on attempting to develop more realistic models of the near field. For example, some processes which could be either included or modelled better are: 1) Slow release of radionuclides from some matrices (e.g. bitumen, resin, activated steel); 2) elemental solubility limits; 3) variations in radionuclide sorption; 4) more realistic gas production rates from corrosion. In order to justify the use of such models, the absence of perturbing effects needs to be demonstrated, of which microbial processes could be particularly important.

Microbial perturbations

Several reviews have considered the constraints on microbial growth within a repository and the possible consequences of such growth (e.g., West et al.^{9,10}, McKinley et al.⁴). Of particular concern for the current generation of safety assessment models are processes which affect:

- radionuclide solubilisation and speciation (e.g., production of organic complexants as byproducts);
- radionuclide mobilisation (e.g., byproduct complexation or direct microbial uptake/taxis);
- gas production – influencing both engineered barriers, physical integrity and chemical degradation of cement (e.g., as a result of microbial degradation of organic materials).

It should be noted that microbial processes may also have positive effects on near-field performance due to:

- direct uptake and immobilisation of radionuclides;
- complexation of radionuclides by immobile macromolecular byproducts;
- catalysis of chemical reactions to provide a more favourable chemical environment (particularly by ensuring reducing conditions);
- hindering advective flow by pore clogging (with biomass or byproducts);

Table 1. Components of an average 1 m³

Component		Total repository with all waste types			Tunnel section with BA-1b waste		
		mass (kg)	% vol.	% mass	mass (kg)	% vol.	% mass
Organics	(w)	41	4.1	1.7	73	7.3	3.1
Steel	(s)	121	1.7	5.0	121	1.7	5.2
	(w)	110	1.4	4.5	0	0	0
Concrete/cements	(s)	1813	77.2	74.1	1813	77.2	77.7
	(w)	325	13.8	13.3	325	13.8	14.0
Salt concentrates	(w)	18	0.9	0.7	0	0	0
Other	(w)	18	0.9	0.7	0	0	0

w = waste/matrix; s = structural (canisters, backfill, lining).

– decreasing leaching/corrosion of surfaces due to biofilm formation.

The current Swiss research programme is, however, focussed on determination of the maximum possible detriment which could be caused by the presence of microorganisms in the near field. As such, potentially positive effects are conservatively ignored.

Modelling approaches

Given that microbial contamination of a repository is inevitable and that this could cause potentially significant perturbations, microbial activity in the near field must be quantitatively analysed. Three basic approaches are possible:

- Mass balance – the maximum biomass will inherently be limited by the inventory and/or supply rate of some essential element (e.g., C, N, P, S), which may represent a very significant constraint. If the biomass is very low, it may be possible to justify neglecting microbial processes in comparison with competing purely inorganic reactions.
- Thermodynamic – in addition to nutrients, constraints set by the available energy sources can be evaluated. Deep repositories tend to be extremely oligotrophic and hence this approach may be appropriate.
- Kinetic – the evolution of a population of organisms in a particular evolving chemical system is explicitly modelled using either empirical or mechanistically derived kinetic data.

For this work, approaches (a) and (b) have been used and are described in more detail in the following section. The mass balance involves simple accounting calculations while the thermodynamic approach is based on the seminal work of Thauer and co-workers^{7,8}. The kinetic approach would be complementary but has not been developed as yet due to lack of data, although a laboratory study has been initiated which may provide appropriate information.

Reference near field definition

An essential first stage in such modelling is defining a 'reference repository system' in detail. In first studies, an

average cubic metre of repository was defined based on the total repository inventories. More recently, an idealised section of a repository with a particularly relevant waste type has been analysed¹. The latter approach is inherently more realistic, but requires that several such sub-models be evaluated in order to quantify the influence on the repository as a whole.

The waste type chosen for further analysis ('BA 1b') consists of contaminated ion-exchange resins in steel drums backfilled with cement. This waste corresponds to a major part of the radioactivity produced during reactor operation and is relatively well defined⁵. It also represents a potentially significant nutrient source for microorganisms.

A reference section of emplacement tunnel is chosen so that the total contained volume is 1 m³. The material components in this unit volume are listed in table 1, together with the equivalent data for an averaged m³ of the total repository with all waste types. A further important part of the reference conditions are the flux and composition of groundwater flowing into this near field. The local hydrology and geometric arguments yield a flux of 0.3 l/year which, for the case of the marl host rock considered, has the composition specified in table 2.

From these basic data plus detailed chemical analysis of individual materials, elemental inventories in the near field can easily be calculated, as can the elemental fluxes in the groundwater. Although the chemistry of the near field is inherently very complex, the major components can be identified from the inventory data and their variation in time described in terms of basic reactions – e.g.,

Table 2. Marl groundwater chemistry and elemental flux

Species*	Concentration (kmol/m ³)	Flux* (kmol/a)
Na ⁺	1.3×10^{-1}	3.7×10^{-5}
Mg ²⁺	6.6×10^{-4}	1.9×10^{-7}
Ca ²⁺	2.7×10^{-3}	7.8×10^{-7}
Str ²⁺	9.2×10^{-5}	2.7×10^{-8}
SO ₄ ²⁻	2.6×10^{-2}	7.5×10^{-6}
NO ₃ ⁻	1.7×10^{-3}	4.9×10^{-7}
NO ₂ ⁻	1.1×10^{-4}	3.2×10^{-8}
Cl ⁻	7.6×10^{-2}	2.2×10^{-5}
C _{inorg.}	5.7×10^{-3}	1.6×10^{-6}
P	1.6×10^{-5}	4.6×10^{-9}

*) to reference 1 m³ of BA-1b for 10⁻¹⁰ m³/m²/s flow rate

Table 3. Nutrients in 1 m³ of tunnel section with BA-1b waste

Component		Content (kmol/m ³)	N	S	P
		C			
Ion exchangers	(w)	3.8	2.0×10^{-1}	2.0×10^{-1}	—
Steel	(s)	1.0×10^{-2}	8.6×10^{-4}	—	—
Concrete	(w)	6.6×10^{-2}	1.8×10^{-3}	4.1×10^{-2}	$*(5.2 \times 10^{-4})$
	(s)	2.0×10^{-1}	—	7.4×10^{-2}	$*(2.9 \times 10^{-3})$
Total		4.1	2.0×10^{-1}	3.2×10^{-1}	$*3.4 \times 10^{-3}$

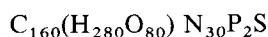
w = waste/matrix; s = structural (canisters, backfill, lining); *assumed value – see text.

oxidation, reduction, hydrolysis etc. Some of the most significant reactions are:

- Corrosion of steel to form iron oxyhydroxides with associated consumption of oxidants and, under anoxic conditions, production of H₂;
- Leaching of alkali hydroxides from the cement with an associated decrease in pore water pH; formation of carbonate and sulphate phases causing the depletion of these anions in pore water and degrading the physical properties of the cement;
- Hydrolysis of the ion-exchange resins providing a source of potentially mobile organic carbon.

Essential nutrient limitations on biomass

A possible representation for the relative requirements of major elements is given by the typical microorganism composition⁶:



The inventories of these nutrients in the reference tunnel section are given in table 3, from which it can be seen that P would undoubtedly be limiting, constraining the maximum possible biomass to 7 kg (dry weight) in 1 m³ assuming 100% P utilisation. In terms of groundwater supply, P would also be limiting with the maximum biomass from this source being very low ($\leq 10^{-5}$ kg dry weight/year). The results of such mass balance calculations must be used with care. Firstly, the calculations are only for 100% utilisation and do not take into account that much of these inventories may be totally refractory or released in a usable form only at very low rates. Secondly, the data themselves are very uncertain – particularly for P which is either not measured or present at concentrations below detection levels in most materials. In this case, the P content was calculated by assuming that it is present in the concrete as a plasticiser at a concentration just below the detection limit reported for chemical analysis of the concrete.

Energetics

In initial studies⁴, a selected number of important energy producing reactions were identified and the potential biomass which could be produced from the microbial utilisation of such energy calculated. With increasing knowledge of the diversity of microbial metabolic sys-

tems, it is difficult to justify the selection of particular reactions without use of a more formal methodology. Following McCarty³, all reactions which could be utilised are listed as redox half reactions, which readily allows the most favourable energy producing couple to be identified (cf. also paper by Arter et al., this issue). This approach may result in consideration of reactions which, although energetically favourable, involve redox couples which are not utilised by microbes. As long as the most favourable reactions can be identified, however, this method is conservative – overpredicting the available energy. Of particular importance in the hyperalkaline cement pore waters is the correction of the half-reaction

Table 4. Possible microbially catalysed redox half-reductions. The free energy change in these reactions (in kJ mol⁻¹) is given for a pH = 12, normalised by the number of electrons involved (n) (from Grogan and McKinley¹)

	$\frac{\Delta G_r}{n}$ (pH = 12)
$H_2O_2 + 2e^- + 2H^+ \rightarrow 2H_2O$	– 101.7
$NO_2^- + 3e^- + 4H^+ \rightarrow \frac{1}{2}N_{2(g)} + 2H_2O$	– 56.1
$O_2 + 4e^- + 4H^+ \rightarrow 2H_2O$	– 50.1
$NO_3^- + 5e^- + 6H^+ \rightarrow \frac{1}{2}N_{2(g)} + 3H_2O$	– 37.9
$NO_3^- + 2e^- + 2H^+ \rightarrow NO_2^- + H_2O$	– 10.6
$NO_3^- + 8e^- + 9H^+ \rightarrow NH_3 + 3H_2O$	– 1.4
$NO_2^- + 6e^- + 7H^+ \rightarrow NH_3 + 2H_2O$	1.8
$\langle HCHO \rangle + 4e^- + 4H^+ \rightarrow CH_{4(g)} + H_2O$	29.1
$S_{(s)} + 2e^- + H^+ \rightarrow HS^-$	40.2
$SO_3^{2-} + 6e^- + 7H^+ \rightarrow HS^- + 3H_2O$	44.5
$\langle HCHO \rangle + 2e^- + 2H^+ \rightarrow \langle HCH_2OH \rangle$	46.1
$SO_4^{2-} + 8e^- + 9H^+ \rightarrow HS^- + 4H_2O$	53.0
$CO_3^{2-} + 8e^- + 10H^+ \rightarrow CH_{4(g)} + 3H_2O$	56.3
$Fe(OH)_3 + 2e^- + 3H^+ \rightarrow Fe_{(s)} + 3H_2O$	57.1
$SO_4^{2-} + 6e^- + 8H^+ \rightarrow S_{(s)} + 4H_2O$	57.3
$CO_3^{2-} + 6e^- + 8H^+ \rightarrow \langle CH_2 \rangle + 3H_2O$	59.3
$2SO_4^{2-} + 8e^- + 10H^+ \rightarrow S_2O_3^{2-} + 5H_2O$	59.4
$2CO_3^{2-} + 8e^- + 11H^+ \rightarrow CH_3COO^- + 4H_2O$	61.4
$CO_3^{2-} + 5e^- + 7H^+ \rightarrow \langle CH \rangle + 3H_2O$	61.6
$3CO_3^{2-} + 14e^- + 19H^+ \rightarrow CH_3CH_2COO^- + 7H_2O$	61.7
$CO_3^{2-} + CH_3COO^- + 6e^- + 8H^+ \rightarrow CH_3CH_2COO^- + 3H_2O$	62.1
$H^+ + 1e^- \rightarrow \frac{1}{2}H_{2(g)}$	68.5
$CO_3^{2-} + 6e^- + 8H^+ \rightarrow \langle HCH_2OH \rangle + 2H_2O$	71.0
$Fe_3O_{4(s)} + 8e^- + 8H^+ \rightarrow 3Fe_{(s)} + 4H_2O$	76.5
$CO_3^{2-} + 4e^- + 6H^+ \rightarrow \langle HCHO \rangle + 2H_2O$	83.5
$CO_3^{2-} + 2e^- + 4H^+ \rightarrow \langle HCOO \rangle + H_2O$	96.1
$\alpha FeO(OH)_{(s)} + 1e^- \rightarrow Fe(OH)_3^- + H_2O$	99.9
$\alpha Fe_3O_{4(s)} + 2e^- - H^+ \rightarrow 3Fe(OH)_3^- - 5H_2O$	134.7

energetics to appropriate pH values (as most such reactions involve proton production or consumption).

A list of redox half-reactions for the main reactive components are shown in table 4. From such tables, the maximum available energy or rate of energy production can

be assessed (kinetic constraints derived simply from water supply flow rates and empirical material degradation rates). Here, the minimum useable energy is taken to be 15 kJ/mole of electrons transferred (cf. discussion in Thauer and Morris⁸). This is probably reasonable for this case, but could be examined further.

The energy production and resultant biomass calculated is summarised in table 5. In table 5,1) the main active components of the materials in the near field are identified and the kinetics of reaction specified by a material lifetime (cf. more detailed discussion in Grogan and McKinley¹). In table 5,2) the energy production profile is summarised for the slowly evolving near field. When iron is present, it is the most favourable electron donor with, initially (< 270 years after emplacement), SO_3^{2-} from the ion-exchange resin being the main electron acceptor. During the period from 270 to 3000 years after closure, the main electron acceptors are SO_4^{2-} and CO_3^{2-} leached from the cement. For times greater than 3000 years, only very low energy supply rates result from oxidation of the aromatic backbone of the ion-exchange resin by groundwater $\text{NO}_3^-/\text{NO}_2^-$ (cf. table 5,3).

At times > 3000 years, the consequences of the previously mentioned energy utilisation cut-off (at 15 kJ/mol of electrons transferred) is illustrated. A lower cut-off would allow sulphate to be considered as an oxidant of the aromatic carbon, increasing the energy supply by a factor of between 3 and 5.

Table 6 converts the energy supply rates to equivalent biomass or biomass production rates. Also included here are the contributions due to oxidants in the original trapped air and resulting from radiolysis. At short times after waste emplacement, ≈ 4 g of biomass/standard cubic metre could be produced by trapped air/radiolytic products. Thereafter ≈ 2 g/year could be produced over the period during which the ion-exchange resins degrade (loss of sulphate and trimethylamine functional groups) which drops to ≈ 0.1 g/year until all Fe is consumed and ≈ 4 mg/year thereafter when the main oxidant supply to the system comes from NO_3^- in the groundwater and the hydrocarbon backbone of the ion-exchange resins is the main reductant.

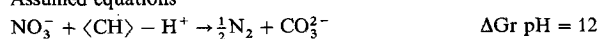
Table 5. Steady state energetics (from Grogan and McKinley¹)

1) Source (mass in unit volume)	Reactant	Inventory (mol)	Supply rate (mol/a)	Life- time (a)
Steel (121 kg)	$\text{Fe}_{(s)}$	2200	7.2×10^{-1}	3×10^3
Concrete (2138 kg)	SO_4^{2-}	87	8.7×10^{-3}	1×10^4
	CO_3^{2-}	232	2.3×10^{-2}	1×10^4
	Fe^{3+}	328	3.3×10^{-2}	1×10^4
Organics (73 kg)	$\langle \text{C}_6\text{H}_6 \rangle$	400	1.48	270
	$\langle \text{CHCH}_2 \rangle$	400	1.48	270
	$\langle \text{N}(\text{CH})_3 \rangle$	200	0.74	270
	$\langle \text{SO}_3 \rangle$	200	0.74	270
Groundwater	SO_4^{2-}		7.5×10^{-3}	
	NO_3^-		4.9×10^{-4}	
	NO_2^-		3.2×10^{-5}	
2) Energy production profile	Oxidant	Reductant	Energy (kJ/a)	
a) Time 0–270 years (groundwater NO_2^- ignored)	SO_4^{2-}	$\text{Fe}_{(s)}$	3.0	
	SO_3^{2-}	$\text{Fe}_{(s)}$	142.2	
	CO_3^{2-}	$\text{Fe}_{(s)}$	4.0	
	NO_3^-	$\text{Fe}_{(s)}$	<u>0.3</u>	149.5
b) 270– 3×10^3 years (groundwater NO_2^- ignored)	SO_4^{2-}	$\text{Fe}_{(s)}$	3.0	
	CO_3^{2-}	$\text{Fe}_{(s)}$	4.0	
	NO_3^-	$\text{Fe}_{(s)}$	<u>0.3</u>	7.3
c) 3×10^3 – 1×10^4 years	SO_4^{2-}	$\langle \text{CH} \rangle$	1.12*	
	NO_3^-	$\langle \text{CH} \rangle$	0.24	
	NO_2^-	$\langle \text{CH} \rangle$	<u>0.01</u>	0.25
d) Times > 1×10^4 years	SO_4^{2-}	$\langle \text{CH} \rangle$	0.52*	
	NO_3^-	$\langle \text{CH} \rangle$	0.24	
	NO_2^-	$\langle \text{CH} \rangle$	<u>0.01</u>	0.25

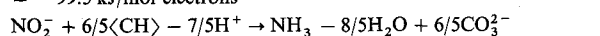
* n.b.: $\text{SO}_4^{2-} + \langle \text{CH} \rangle$ gives only 8.6 kJ/mol electrons and is therefore not included in the energy production calculations

3) Resin degradation at times > 3×10^3 years

a) Assumed equations



= – 99.5 kJ/mol electrons



= – 59.8 kJ/mol electrons

$\Delta \text{Gr pH} = 12$

(n.b. $\text{SO}_4^{2-} + \langle \text{CH} \rangle$ gives only 8.6 kJ/mol electrons)

b) Supply rates

NO_3^- ; 4.9×10^{-4} mol/a; $5e^-$ transferred/mol $\text{NO}_3^- \rightarrow 0.24$ kJ/a

NO_2^- ; 3.2×10^{-5} mol/a; $6e^-$ transferred/mol $\text{NO}_2^- \rightarrow 0.01$ kJ/a

Total = 0.25 kJ/a

Consequences of microbial activity

From the mass balance arguments in section 6, P is clearly the limiting element in this system. If the maximum

Table 6. Energy to biomass conversion (from Grogan and McKinley¹)

	Energy	Biomass (dry wt)
1 Trapped air	71.4 kJ	1.1 g
2 Radiolysis	189.6 kJ	3.0 g
3 Profile		
0–270 a	149.5 kJ/a	2.3 g/a
270– 3×10^3 a	7.3 kJ/a	0.1 g/a
3×10^3 – 1×10^4 a	0.25 kJ/a	3.9×10^{-3} g/a
> 10^4 a	0.25 kJ/a	3.9×10^{-3} g/a

possible biomass production from P in concrete is assumed to be constrained by the degradation of the concrete matrix (over 10^4 years), an average production rate of ≈ 0.7 g (dry weight)/year would result. This factor would limit biomass production over the first 270 years (cf. table 6) but, thereafter, available energy would be the main constraint on microbial activity. It is interesting to note, en passant, that the difference between the nutrient and energy constraints is never very large (factors of 2–5) and is probably within the range of uncertainty in such simple calculations.

Of the possible microbial perturbations, gas production would not be expected to be particularly significant for this waste type (but could be important for cellulosic waste – see paper by Colasanti et al., this issue). More important would be the production of organic byproducts which could act as radionuclide complexants.

In the simplest case we can assume a steady state biomass and an initial production of 0.7 g/a biomass balanced by an equivalent loss of organic complexant. In the water flux of 0.3 l/a assumed, this would give rise to ≈ 2.4 g/l of dissolved organic carbon – considerably higher than natural levels in groundwater (in the order of mg/l). To put this number in context, assuming 1 complexing group (e.g. $-\text{CO}_2^-$) per 100 daltons of 'organic' would give a concentration of 24 mmoles of complexant per litre. Such a concentration is high relative to radionuclide inventories in the waste and could be especially important for nuclides with low solubilities in such hyperalkaline conditions (e.g., Ni-59, Tc-99, Pd-107, Sn-126).

At times $> 3 \times 10^3$ years, biomass production will have dropped by approximately two orders of magnitude but, even then, organic byproducts could be produced at up to an order of magnitude higher rates than the supply from groundwater.

Future modelling developments

The basic approach described has only been applied to a single waste type, and its sensitivity to assumptions therein needs to be tested by application to other waste sorts. In the previous sections some limitations were mentioned which could be improved, i.e.:

- 1) Poor elemental inventories in repository materials;
 - 2) Lack of data on elemental availability;
 - 3) Very simplistic treatment of oligotroph energetics.
- More fundamentally, the modelling approach needs to be tested against experimental studies or, possibly, well defined natural systems. Improvement of the background experimental database will allow both improvement of the thermodynamic approach and also examination of alternative kinetic approaches which may be less overly conservative.

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