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Journal of Nuclear Materials 298 (2001) 125–135

journal of nuclear materials

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Performance assessment of the disposal of vitrified high-level waste in a clay layer

Dirk Mallants *, Jan Marivoet, Xavier Sillen

Belgian Nuclear Research Centre, Section Performance Assessment, SCK·CEN, Boeretang 200, 2400 Mol, Belgium

Abstract

Deep disposal is considered a safe solution to the management of high-level radioactive waste. The safety is usually demonstrated by means of a performance assessment. This paper discusses the methodological aspects and some of the results obtained for the performance assessment of the disposal of vitrified high-level waste in a clay layer in Belgium. The calculations consider radionuclide migration through the following multi-barrier components, all of which contribute to the overall safety: (1) engineered barriers and the host clay layer, (2) overlying aquifer, and (3) biosphere. The interfaces between aquifers and biosphere are limited to the well and river pathway. Results of the performance assessment calculations are given in terms of the time evolution of the dose rates of the most important fission and activation products and actinides. The role of the glass matrix in the overall performance of the repository is also discussed.

1. Introduction

Approximately 60% of the total electricity production in Belgium originates from nuclear power plants. The Belgian nuclear energy programme has been scheduled to last for 50 years. The first two nuclear reactors became operational in 1975; whereas, in 1985 the last two reactors were connected to the electricity net. With an estimated reactor lifetime of 40 years, the first reactors will theoretically be decommissioned in the year 2015 and the last two in 2025. The total installed capacity of the seven nuclear reactors is 5.5 GW(e).

About 5000 tonnes of uranium heavy metal are required for the scheduled nuclear programme. At present contracts for the reprocessing of only 630 tU have been concluded with the French COGEMA and the resulting waste will be disposed of in Belgium. No decision has been taken yet about the future handling of the remaining 4370 tU, i.e. reprocessing or direct disposal of spent fuel. In the event that only 630 tU will be reprocessed, 420 stainless steel canisters of 0.15 m^3 vitrified high-level waste each will require disposal. Under the condition that all the spent fuel will be reprocessed, a total of 3915 canisters will require disposal.

The spent fuel contains considerable amounts of volatile elements such as ^{129}I and ^{36}Cl. During reprocessing, part of these volatile elements are released as iodine or chlorine gas. However, at present there is uncertainty about the amount of ^{129}I remaining in the reprocessed waste. To cope with these uncertainties, a relatively high and arbitrary value of 1% was given to the ^{129}I remaining in the vitrified high-level waste.

At present the Belgian reference site for disposal studies of high-level waste (HLW) is the Boom Clay layer at the Mol site (province of Antwerp). The underground repository currently considered is assumed to be located underneath the present SCK·CEN site at Mol at a depth of approximately 230 m below surface. This is in the middle of the Boom Clay layer (Fig. 1). At the Mol site, the Boom Clay layer has a thickness of 100 m and is overlain by the 180-m thick Neogene aquifer. The Boom Clay is a marine sediment of tertiary, Rupelian age. It is a plastic, organic-rich clay and has an extremely low hydraulic conductivity K (i.e., $K \cong 2.5 \times 10^{-12}$ m/s [1]). Furthermore, the actual driving force for water flow through the Boom Clay, i.e. the hydraulic gradient, is negligible (i.e., at the Mol site the downwards oriented gradient is 0.02 m/m [2]).

* Corresponding author. Tel.: +32-14 333 219; fax: +32-14 323 553.

E-mail address: dmallant@sckcen.be (D. Mallants).

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PII: S0022-3115(01)00577-3

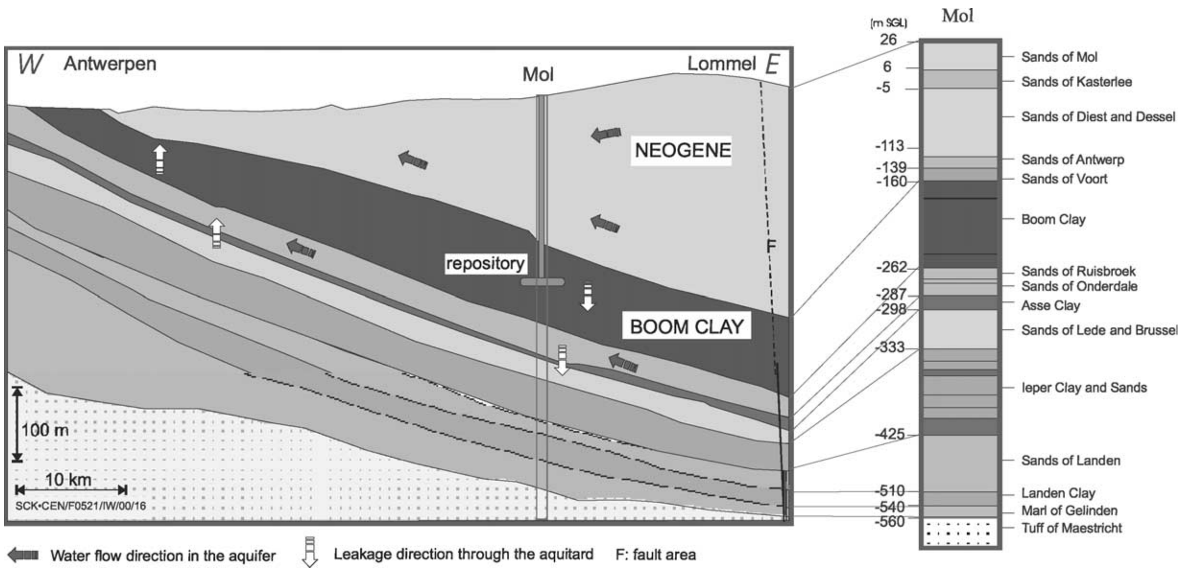

Fig. 1. Schematic view of Boom Clay layer and location of proposed repository.

A schematic view of the considered repository is given in Fig. 2. Access to the underground facility is secured by means of two 6-m diameter vertical shafts that are connected by means of a 400-m long transport gallery. Two disposal areas can be distinguished, i.e., one for HLW and one for medium-level waste (MLW). The two disposal areas are connected by means of two 3.5-m inner diameter transport galleries. The HLW will be disposed in eight 800-m long horizontal galleries which are placed orthogonal to the transport galleries and which are partitioned into 4 segments of 200 m. The HLW disposal galleries will have an inner diameter of approximately 2 m. The spacing between the galleries should be sufficiently large so as to avoid (i) too high temperatures in the zone surrounding the disposal galleries and (ii) a significant warming up of the overlying aquifer.

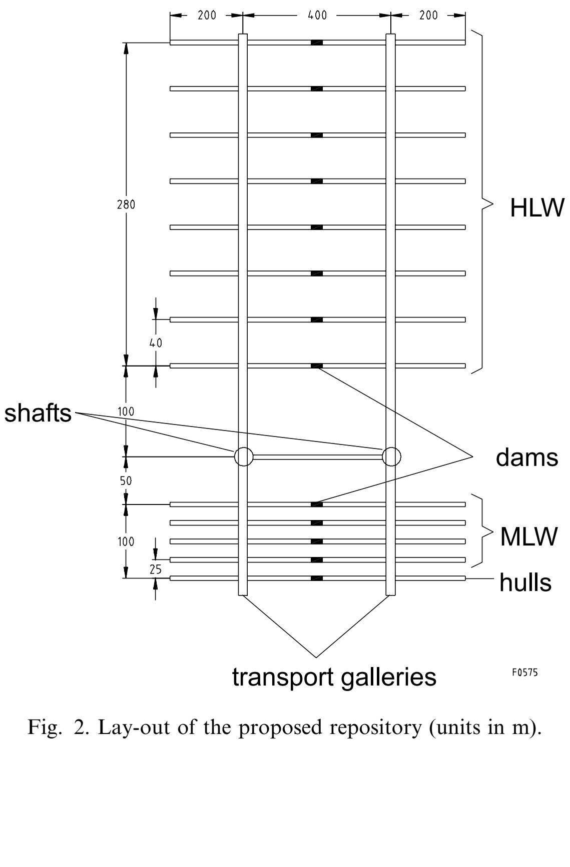

Fig. 2. Lay-out of the proposed repository (units in m).

Details on the engineered barriers surrounding the HLW canisters can be found in Fig. 3. Each of the 0.43-m outer diameter canisters is placed in a 0.02–0.03-m thick metallic overpack which will be pushed inside a disposal tube in the disposal gallery. The purpose of the overpack is to avoid the contact of water with the HLW canisters and the vitrified waste during the thermal phase of the repository. The 0.03-m thick overpack has a

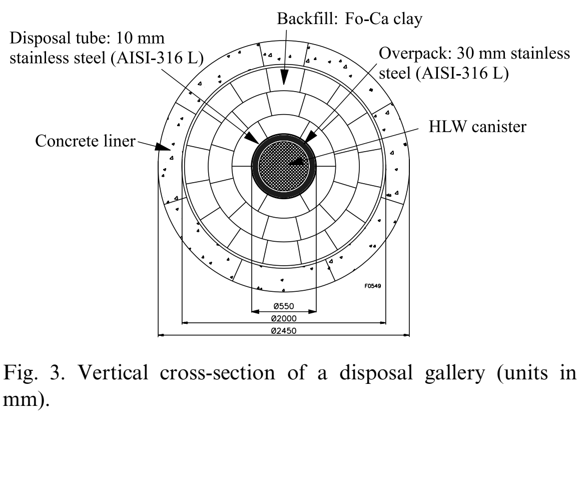

Fig. 3. Vertical cross-section of a disposal gallery (units in mm).

design lifetime of approximately 1000 years during which it should remain intact. Currently the considered material is AISI-316L hMo (L = low carbon, hMo = high molybdenum) stainless steel. The overpack will be placed in a 0.01-m thick disposal tube. The latter allows for placement of the 1-m thick backfill material prior to placement of the radioactive waste and is not a long-term barrier against the release of radionuclides. A mixture of Fo–Ca clay (60%), sand (35%), and graphite (5%) will be used as backfill material. The Fo–Ca clay is a natural clay occurring in France in which Ca-bentonite is one of the main components.

As a result of the disposal of the HLW, the waste will be isolated from man and environment for a sufficiently long time such that an acceptable level of protection is ensured. This isolation from the biosphere is obtained by means of the multi-barrier concept (see e.g., [3]). The latter refers to a number of barriers that are independent (i.e., barrier performance is independent from that of other barriers), and diverse (i.e., performance of different barriers is based on different processes leading to a series of complementary barriers). We note that the term independent refers to the functioning of the barrier; e.g., failure of the backfill material will not lead to a failure of the glass matrix. It does not mean, however, that its contribution to the overall performance of a repository should be taken to be independent from that of other barriers, including natural barriers. The following components are considered in the repository system: conditioned HLW/canister/overpack/backfill/clay layer/aquifer/biosphere. Note that the conditioned (i.e. the vitrified) waste is only one component of a whole series of barriers. In performance assessment studies, the long-term behaviour of the disposal system as a whole (i.e., all barriers together) is investigated. In the United States the term total system approach is used [4]. Usually these components are represented by different submodels, whose levels of phenomenological detail is not necessarily the same.

The objective of this paper is to illustrate by means of a sensitivity analysis what the relative importance is of a particular submodel in the performance of the total system. The submodel considered describes the corrosion of the glass matrix under the prevailing geochemical conditions in Boom Clay. In this way the required level of phenomenological detail for a given submodel can be identified.

2. Methodology

The methodology applied in this performance assessment is consistent with the methodology applied in previous calculations for disposal of HLW in Boom Clay, including UPDATING 1990 [5], and PAGIS [6]. The methodology is also consistent with performance assessments carried out elsewhere in Europe and the United States [4]. The two main steps are: (1) scenario identification and selection, and (2) consequence analysis. In the first step a systematic analysis of the features, events and processes (FEPs) that might influence the behaviour of the repository system is completed. In the second step the doses are calculated that result from the selected set of the relevant scenarios. This step requires the use of several models, including source term models, groundwater flow and contaminant transport codes, pathways-to-man models, and sensitivity techniques.

Once a list of relevant FEPs has been established, the FEPs are split into two categories: those belonging to the normal evolution scenario (NES) and those belonging to the altered evolution scenario (AES). The NES considers all phenomena that are almost certain to occur. As a result, a large number of FEPs are treated together in the NES. Table 1 gives an overview of all FEPs considered in the NES of the near field, clay, and aquifer (based on [7]).

Altered evolution scenarios relevant for the performance of the glass matrix are the early failure of the overpack, poor sealing scenario, and scenarios related to human activities [2]. Poor sealing refers to an improper backfilling of the disposal galleries and sealing of the access shafts. A typical human intrusion scenario is the one dealing with unsealed boreholes. As it cannot be ruled out that in the future a number of investigation boreholes will be drilled at the selected repository site, one or more of these boreholes could be poorly sealed. Discussion of the potential impact of such alternative scenarios is beyond the scope of this paper.

The evaluation of the long-term behaviour of the repository under the NES will be based on the so-called robust performance assessment concept. This means that the very complex repository concept is reduced to a much simpler one that can be modelled with a high degree of confidence. The simpler concept only accounts for a limited number of known physico-chemical processes. The latter are described by a limited number of parameters that are generally determined with a high degree of confidence. Furthermore, for processes that are insufficiently known and/or characterized, the principle of conservatism is used. This means that, out of a realistic range of parameter values, the one that contributes least to the safety is selected. The robust approach increases the defensibility of the evaluation.

Applying the robust concept to the radioactive waste disposal in Boom Clay means that the following components and the associated parameters will be distinguished [8]:

The near field:

the overpack lifetime;

the corrosion rate of the waste form;

the solubility limits of the radionuclides in the near field;

Table 1
List of features, events, and processes (FEPs) considered in the normal evolution scenario (NES)

Type	FEP-number	FEP	Influence
Transport and geochemical	1.2.05	Diagenesis	C
	1.2.06	Uplift and subsidence	A
	1.2.11	Rock heterogeneity	C, A
	1.3.01	Precipitation, temperature and soil water balance	C, A
	1.3.04	Sea-level change	A
	1.3.05	Periglacial effects and glaciation	A
	1.4.02	Denudation	A
	1.4.03	River, stream, channel erosion	A
	1.5.01	River flow and lake level changes	A
	1.5.03	Recharge to ground water	A
	1.5.04	Ground water discharge	A
	1.5.05	Ground water flow	C, A
	1.5.06	Ground water conditions	A
	1.6.01	Advection and dispersion	C, A
	1.6.02	Diffusion	N, C, A
	1.6.06	Solubility limit	N, C
	1.6.07	Sorption	N, C, A
	1.6.08	Dissolution, precipitation and cristallization	N, C
	1.6.09	Colloid formation, dissolution and transport	N, C, A
	1.6.10	Complexing agents	N, C
	1.6.13	Mass, isotopic and species dilution	N, C, A
Design and construction	2.1.11	Chemical effects: oxidation of the host rock	N
	2.1.12	Excavation effects	N, C
	2.2.05	Heterogeneity of waste forms (chemical, physical)	N
	2.3.11	Ground water abstraction	A
	2.4.10	Quarries, near surface extraction	A
Thermal	3.1.01	Differential elastic response	N
	3.1.02	Non-elastic response	N, C
	3.1.04	Induced hydrological changes	N, C
	3.1.05	Induced chemical changes	N, C
Chemical	3.2.01	Metallic corrosion	N
	3.2.02	Interactions of host materials and ground water with repository materials	N
	3.2.03	Interactions of waste and repository materials with host materials	N
	3.2.04	Non-radioactive solute plume in geosphere	C
	3.2.06	Introduced complexing agents and cellulosis	N
Mechanical	3.3.02	Changes in in-situ stress field	N
	3.3.03	Embrittlement and cracking	N
Radiological	3.4.01	Radiolysis	N
	3.4.02	Material property changes	N, C
	3.4.04	Radioactive decay ingrowth	N, C, A

(components influenced by FEP: N = near field; C = clay; A = aquifer).

The clay layer:
- effective thickness of the clay layer;
- transport parameters of the radionuclides: pore diffusion coefficient, accessible porosity, and retardation factor;

The aquifers:
- well pathway: Darcy velocity, dispersivity, and aquifer thickness;
- river and soil pathways: detailed aquifer modelling required;

The biosphere:
- various biosphere components.

In addition to the robust concept, the three-dimensional real world geometry of the disposal system is simplified into a two-dimensional model of the Boom Clay. This is illustrated in Fig. 4. Such a reduction in the complexity

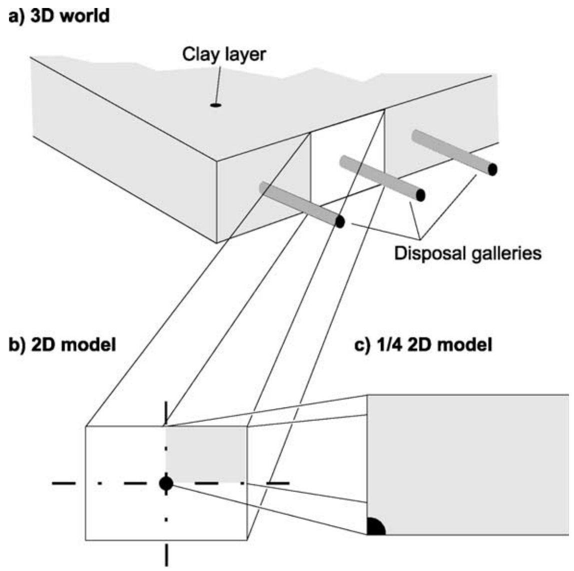

Fig. 4. Simplification of the 3D real world geometry into a 2D model of the near field and far field.

is done by first considering equally spaced disposal galleries with the waste homogeneously distributed over the galleries. Furthermore, the Boom Clay is also considered homogeneous. Therefore, the release of radionuclides into the Boom Clay will be the same for each gallery. Transport parallel with the disposal galleries is considered zero because of the absence of any driving force (i.e., no hydraulic gradient, no concentration gradient). As a result, transport will be restricted to a two-dimensional rectangular area whose vertical and horizontal dimensions are, respectively, the thickness of the Boom Clay and the distance between the galleries. The two-dimensional model can be further simplified by the presence of two symmetry axes, both passing through the disposal gallery. The final domain used in the consequence analysis is thus only 1/4 of the whole two-dimensional domain.

2.1. Safety functions of the robust multi-barrier system

The different barriers of a repository system all contribute to one or more of the following basic safety functions: physical confinement, retardation/spread release (i.e., dilution over time), dispersion/dilution, and limited accessibility [3]. An effective retardation results in a maximum radioactive decay and significantly reduces the amounts of long-lived radionuclides that appear in the biosphere per unit of time (spread release). The first three safety functions are related to the normal evolution of the system, i.e., the exposure pathway 'radionuclides in waste $\Rightarrow$ geosphere $\Rightarrow$ biosphere $\Rightarrow$ man'. The fourth safety function is related to the altered evolution scenario, in particular to the intrusion by man.

Physical confinement refers to the perfect isolation of the waste in a watertight barrier during the first phase of a repository's lifetime. As long as this safety function is effective, no contact between groundwater and the radionuclides in the waste can occur. As a result, there will be no release of radionuclides and other components. This is of particular importance for the heat generating wastes, including vitrified HLW and spent fuel. During the thermal phase of the repository, which usually lasts for several hundreds of years, the contact between Boom Clay water and the glass matrix should be avoided because higher temperatures are expected to result in higher glass corrosion rates. Also, several coupled processes may occur that could alter the Boom Clay properties and hence the behaviour close to the waste galleries. Therefore, contact between the groundwater and waste should be avoided within this period. For HLW this function is performed by the overpack of the conditioned waste form. An overpack lifetime of 1000 years has been considered in the present study.

The second safety function deals with retardation and spread release into the biosphere. Once the performance of the first safety function becomes reduced, the second safety function takes over. Water interacts with the waste matrix and leaching of the different components begins. The radionuclides present in the HLW will not be released immediately, because several physico-chemical processes (sorption, diffusion, precipitation, complexation, etc.) will retard and spread the radionuclide release into the biosphere over time. For deep disposal in clay, the geological barrier (in this study it is the Boom Clay layer) is the barrier that contributes most to this safety function. For radionuclides that are not retarded by the Boom Clay (e.g., ^{129}I), the glass matrix also contributes to the spread release, although it is less important as compared with the contribution from the Boom Clay. Retardation and spread release are the most important safety functions of the Boom Clay layer, because the long-lived radionuclides such as ^{129}I (half-life $= 15.6 \times 10^6$ years), ^{235}U (half-life $= 7.0 \times 10^8$ years) and ^{238}U (half-life $= 4.5 \times 10^9$ years) will survive any physical confinement. Only a sufficient retardation and spread release may lead to a safe disposal.

A third safety function is obtained by dispersion and dilution in the aquifers. Once the components leave the clay layer, they are released into the surrounding aquifers. As a result of the natural processes of dispersion and dilution, which take place in the aquifers, rivers, and lakes, the radionuclide concentrations that appear in the biosphere are further reduced. Although this dispersion/dilution process is of secondary importance as compared with the two previous safety functions, it does contribute to the overall safety level of the disposal system, and is therefore accounted for in the performance assessment.

Limited accessibility of a repository is the fourth safety function and is related to possible human intrusion in the repository (i.e., pathway 'man ⇒ waste'). A typical example is the analysis of waste-containing cores by a geologist (exploratory drilling). For deep disposal, such as discussed here, the Boom Clay guarantees limited accessibility.

2.2. Source term models

Since one of the objectives of this paper is to investigate the role of the glass matrix in the overall repository performance, a sensitivity analysis was carried out with different source term models. These models represent different assumptions about the glass dissolution processes, and their use in a sensitivity study can be helpful in identifying processes that are relevant to safety.

The HLW canisters are filled by pouring molten borosilicate glass into cylindrical stainless steel containers. Because of different thermal expansion coefficients of the glass and the steel cylinder, a concentric fracture pattern will be formed during cooling. As a result, the total surface of the fractured glass will be easily 5 to 27 times larger than the surface of an intact glass block [1,9,10]. Furthermore, the dissolution of the glass matrix and the concomitant release of radionuclides is a time-dependent process. Because the evolution of the glass corrosion over long time scales cannot be accurately modelled, we here apply a constant glass corrosion rate that is estimated from the available experimental results [1]. For the purpose of investigating the sensitivity of the overall repository performance with respect to differences in glass corrosion, four theoretical source terms models will be used in the calculations.

By introducing the robust concept, the glass matrix is assumed to be the only waste package component that significantly contributes to the spread release of the radionuclides (the overpack is not part of the waste package but is considered to be an engineered barrier). The corrosion of the waste matrix (i.e. the borosilicate glass) is described by only two parameters, i.e. the corrosion rate of the glass and the glass fracturation ratio [1].

Also in the robust concept, the HLW canisters are not considered to act as a barrier to radionuclide release. Their lifetimes are conservatively taken as zero. Unlike the canisters, the lifetime of the overpack is effectively used in the modelling of the release of radionuclides.

2.2.1. The reference source term model

In the calculation of the radionuclide release from the glass canister, we assumed that the glass matrix corrodes at a constant rate of 0.3 μm/a. This value represents the best estimate of experimental corrosion tests on SON68 glass in Boom Clay at a temperature of 16°C [1]. We further assumed that the best estimate value for the glass fracturation ratio FR ($FR =$ total surface area/surface area of the unfractured glass block) is 10 [1]. The glass corrosion rate together with the glass fracturation ratio determine the glass dissolution rate, i.e. glass dissolution = glass corrosion rate $\times FR = 3$ μm/a. The initial radius of the cylindrical glass matrix equals 0.215 m. Given a constant glass dissolution rate of 3 μm/a, the glass matrix will be completely dissolved after 0.215 m/3 μm/a $\cong$ 72 000 years. In the calculation we will use a value of 70 000 years. In the source term model adopted here we will assume that the radionuclides present in the glass matrix will dissolve at a constant rate during 70 000 years. This is the reference source term model, and it can be considered as a realistic-conservative estimate.

2.2.2. Source term model with maximum corrosion rate and glass fracturation ratio

The second source term model is a variant of the reference model. It uses the maximum glass corrosion rates and maximum glass fracturation ratios obtained from the same experiments [1]. These maximum values are, respectively, 0.4 μm/a and 27. The calculated glass dissolution rate then becomes 11 μm/a. Complete dissolution of the glass matrix occurs after approximately 20 000 years. The second source term model, referred to hereafter as Variant 1, considers a constant dissolution of radionuclides during 20 000 years. This value can be considered as a conservative case.

2.2.3. Source term model with minimum corrosion rate and glass fracturation ratio

The third source term model is also a variant of the reference model. It considers the minimum glass corrosion rates and minimum glass fracturation ratios obtained from the same experiments mentioned for the reference model [1]. These minimum values are, respectively, 0.002 μm/a and 5. The calculated glass dissolution is equal to 0.01 μm/a. Complete dissolution of the glass matrix occurs after approximately 21 000 000 years. In the calculations of the third source term model, referred to hereafter as Variant 2, we will use a smaller value of 1 000 000 years. It is a non-conservative value.

2.2.4. Source term model with instant release

In a fourth source term model, glass dissolution is assumed to occur immediately after perforation of the overpack. All radionuclides present in the glass matrix will thus be released instantaneously. Although it is impossible that such a situation will ever happen, this hypothetical behaviour is included in the calculations because it represents the most nonconservative situation with regard to the source term. The instant release model is Variant 3. These results can be used in a comparison with the other source terms to estimate the contribution of the glass waste matrix to the spread release of the radionuclides.

3. Mathematical models

In the consequence analysis we calculate the annual dose that man receives as a result of uptake of radionuclide containing food stuff, drinking water, etc. Three consecutive steps were distinguished in this analysis: (1) calculation of the radionuclide release from the Boom Clay layer into the Neogene aquifer, (2) calculation of transport of radionuclides in the aquifer towards wells, rivers, and soil, and (3) calculation of annual dose received by man for the most important exposure pathways. For each step different mathematical models are used. Only a brief description of the various models is given here. For a detailed description the reader is referred to [15].

Radionuclide release from the near field and transport through the Boom Clay is calculated by means of the PORFLOW code [11]. Owing to the very low hydraulic conductivity and hydraulic gradient in the Boom Clay, transport of radionuclides is dominated by molecular diffusion. Therefore, the three most important transport parameters in the Boom Clay are the pore diffusion coefficient, $D_p(\mathrm{m^2/s})$, the diffusion accessible porosity, η (–), and the retardation factor, R (–). Furthermore, the dissolution of the majority of the radionuclides in the interstitial pore water of backfill materials such as clay is chemically limited. This is accounted for in the model by applying solubility limits. The PORFLOW code is then used to numerically solve the two-dimensional diffusion equation for the computational domain defined in Fig. 4. Best estimate values for the transport parameters were taken from [1].

The radionuclide fluxes out of the Boom Clay calculated in the previous step can be used as input fluxes for a transport model of the overlying aquifer. Such a model was built and described by [12]. Using this model, we calculated the distribution of the radionuclide concentrations in the Neogene aquifer and the activity fluxes into the rivers as function of time and activity sources, i.e. the amount of radionuclides that enter the model through its base in the area located above the repository.

In the last step the dose for each radionuclide for a water well pathway is obtained by multiplying the maximum radionuclide concentrations calculated in the aquifer with a so-called biosphere conversion factor, BCF. The well pathway assumes that a self-sustaining farmer community uses water from a well drilled at that point in the aquifer where the highest radionuclide concentration occurs. The BCF is calculated by the biosphere model and accounts for all major exposure pathways, including ingestion of contaminated food or water, direct irradiation by contaminated soil or sediment, etc. [13,14]. The calculation of doses for the river pathway is performed similarly: the flux out of the Boom Clay for each nuclide is multiplied by the fraction of this flux that effectively enters the river and by the dose conversion factor calculated by the biosphere model.

4. Results and discussion

Uncertainty about the glass dissolution process is investigated by considering four different source term models. The effect of different source term models on the release of radionuclides is different for different radionuclides. This is illustrated by considering the following radionuclides: ^{94}Nb ($T_{1/2} = 2.0 \times 10^4$ years), ^{129}I, ^{135}Cs ($T_{1/2} = 2.3 \times 10^6$ years), and ^{107}Pd ($T_{1/2} = 6.5 \times 10^6$ years). Under the geochemical conditions of Boom Clay (pH = 8.5, Eh = −250 mV), the first three radionuclides are not solubility limited.

Owing to the relatively short half-life of ^{94}Nb (i.e., 20 300 years), the source activity for the reference case, Variant 1, and 2 was calculated accounting for radioactive decay. The resulting activities for the source term are shown in Fig. 5. These were used as input for the

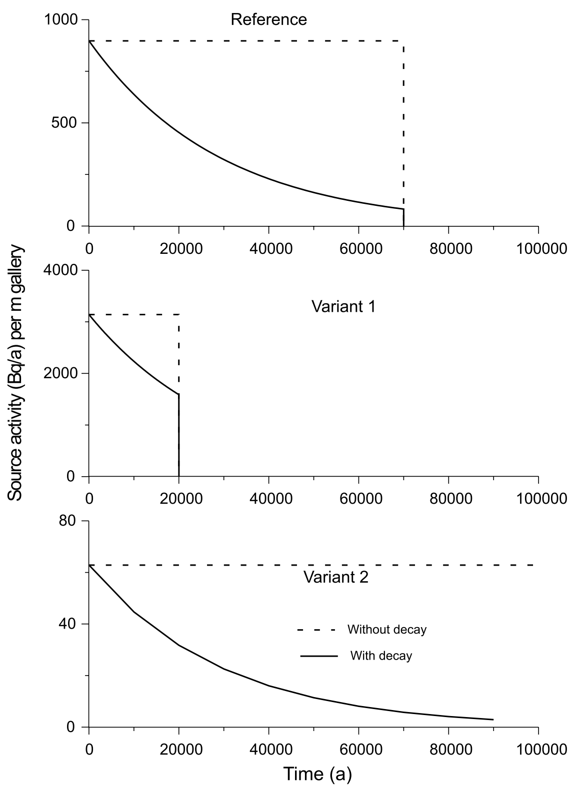

Fig. 5. Different source term models used for ^{94}Nb.

transport calculations with PORFLOW. The effect of these different source term models on the calculated fluxes at the clay–aquifer interface is shown in Fig. 6. As could be expected, the maximum flux is obtained for Variant 3 (instantaneous release owing to immediate and total dissolution); whereas, Variant 2 gives the lowest flux. Intermediate fluxes are obtained for Variant 1 and the reference model. The maximum fluxes for these three models all occur between 200 000 and 300 000 years after closure of the repository. These results indicate that the variation of the ^{94}Nb flux considering the four different source term models is limited to a factor of approximately 37. The contribution of the glass matrix to the spread release of ^{94}Nb can be calculated by comparing the flux evolution for Variant 3 (instantaneous release) with that of the reference model (or alternatively with the pessimistic behaviour of Variant 1). On the basis of such comparison we observe that the glass matrix with a lifetime of 70 000 years reduces the maximum flux approximately 3 times compared to the hypothetical situation where the glass would dissolve instantaneously. In the pessimistic case that the lifetime of the glass would be only 20 000 years, the maximum flux is only 1.4 times lower than the one obtained in the absence of a glass matrix. Only when the lifetime of the glass is sufficiently long, such as for Variant 2 (i.e., one million years), is there a significant contribution from the glass matrix to the overall safety of the integrated repository. More specifically, the slow dissolution of the glass matrix results in a further reduction of the maximum flux by a factor of 37 in addition to the spread release owing to slow diffusion and sorption in the Boom Clay.

The second radionuclide investigated is the non-sorbing ^{129}I. Owing to the very long half-life of ^{129}I ($T_{1/2} = 15.6 \times 10^6$ years), all source models except the instant release model exhibit a nearly uniform flux with time. Compared to the ^{94}Nb results, a slightly different behaviour is observed for ^{129}I, with smaller differences in maximum flux between the reference model and the Variant 1 and 3 model. The maximum flux is nearly identical for these three cases. Fig. 7 shows that a slow release during one million years results in a relatively flat curve with its maximum almost 7 times smaller than the reference model. This maximum is reached after 400 000 years and continues to exist until 1 000 000 years. For the reference source term model the maximum flux occurs 94 000 years after closure of the repository. For a higher glass dissolution rate, such as in the Variant 1 model, the maximum flux will appear earlier (i.e. around 50 000 years after closure). If the source term model considers an instantaneous release of all ^{129}I (Variant 3), then the peak flux appears 44 000 years after closure. For ^{129}I the contribution of the glass matrix to the overall performance is limited, as the differences in maximum flux between the instant release, realistic, and pessimistic case are less than 20%.

The third radionuclide whose behaviour for different source terms is investigated is the strongly retarded ^{135}Cs. The flux behaviour of ^{135}Cs is almost indifferent for the four source term models (Fig. 8). The reference model, together with Variant 1 and Variant 3, all have the same maximum flux which occurs around 17 000 000 years. Only Variant 2 has a slightly lower maximum flux owing to some decay in the source term. The nearly identical behaviour of the four models is explained by

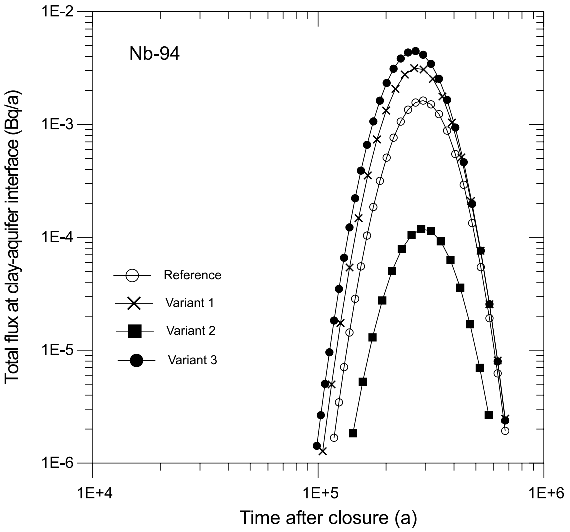

Fig. 6. Simulated ^{94}Nb fluxes at the clay–aquifer interface considering four different source term models.

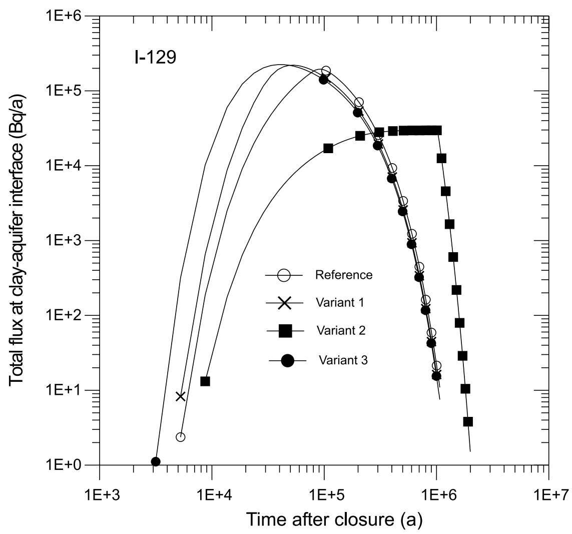

Fig. 7. Simulated ^{129}I fluxes at the clay–aquifer interface considering four different source term models.

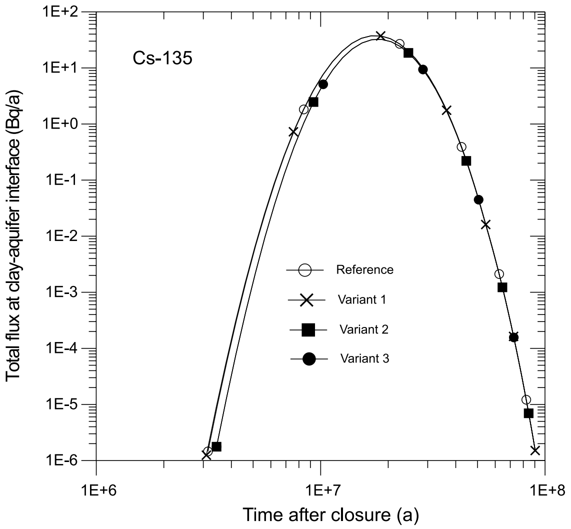

Fig. 8. Simulated ^{135}Cs fluxes at the clay–aquifer interface considering four different source term models.

the high sorption of ^{135}Cs onto the Boom Clay (i.e. $R = 3600$). In other words, the differences in release time (e.g., 70 000 for the reference model versus 1 000 000 for Variant 2) are small compared to the extremely long travel time in the Boom Clay (17 million years).

A last example considers the solubility limited ^{107}Pd. For all source models, the solubility limits are reached. As a result, all curves show the same behaviour (Fig. 9). The glass matrix does not contribute to the flattening of the curve, and has thus no positive effect on the released fraction. Note that we assume that the dissolution of the glass matrix does not lead to significant increase in solution pH and hence does not affect the solubility of ^{107}Pd. Up to now no experimental evidence exists that shows a significant pH variation in Boom Clay as a result of glass dissolution. Furthermore, such effects are not expected in Boom Clay because it has a sufficient buffering capacity owing to the high amounts of bicarbonate. Finally, the solubility of 10^{-7} mole/l applied here is a conservative value valid for a pH range between 8.5 and 9.5, and it is five times higher than the experimental value [1].

For other strongly sorbed radionuclides, including activation and fission products as well as actinides, differences between different source term models were small or negligible. In other words, for radionuclides with long travel times in the Boom Clay the differences in flux observed at the source will have disappeared at the clay–aquifer interface owing to a maximum radioactive decay and an enhanced dilution over time. For non-adsorbing radionuclides such as ^{129}I a lifetime of the glass longer than 1 million years is required to obtain a significant effect on the released fraction out of the Boom Clay.

Another parameter that affects the influence of the glass dissolution on the spread release is the solubility limit. In the case for which radionuclides are solubility limited, the spread release is governed by the solubility limit and not so much by the glass dissolution rate. In such a case, slow dissolution of the glass will not much improve the performance of the waste repository.

The calculated dose rates for fission and activation products (Fig. 10) and actinides (Fig. 11) are shown considering the well pathway. For the fission and

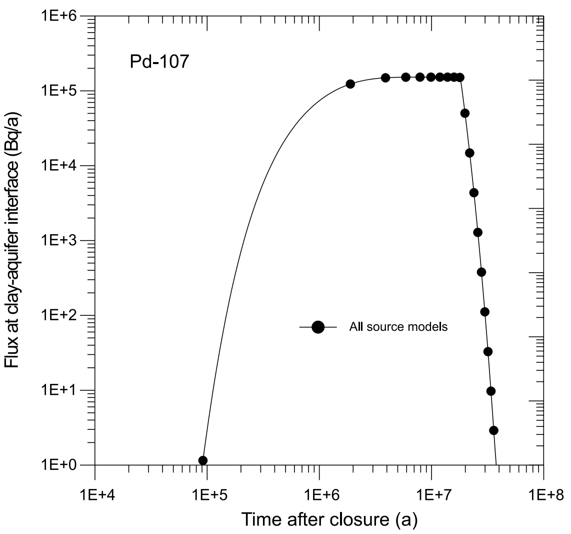

Fig. 9. Simulated ^{107}Pd fluxes at the clay–aquifer interface considering four different source term models.

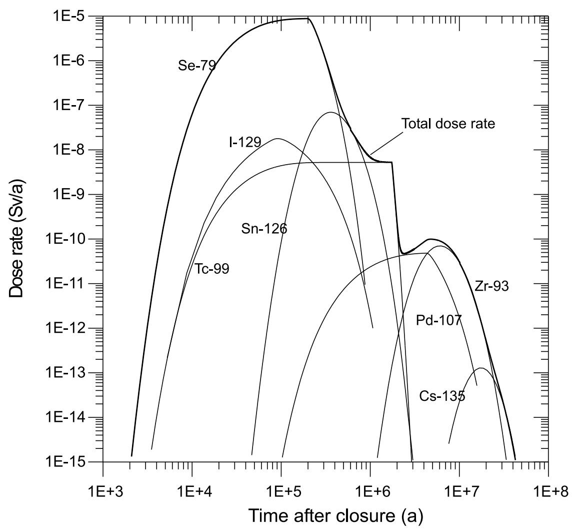

Fig. 10. Calculated total dose rate and dose rates due to activation and fission products for the well pathway (reference source term).

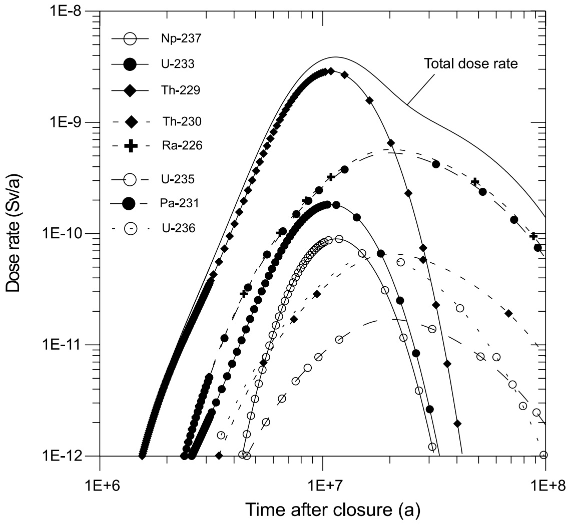

Fig. 11. Calculated total dose rate and dose rates due to actinides for the well pathway (reference source term).

activation products, the three highest dose rates are due to ^{79}Se ($T_{1/2} = 1.1 \times 10^6$ years), ^{129}I, and ^{126}Sn $T_{1/2} = 1.0 \times 10^5$ years). All other radionuclides have dose rates that are significantly smaller. The highest dose rate of 9×10^{-6} Sv/a is still 100 times smaller than the internationally recommended dose limit of 10^{-3} Sv/a. The maximum dose rate for the river pathway was 7×10^{-8} Sv/a.

The concentrations in the well water and the associated dose rates for actinides were also calculated. The dose rates are shown in Fig. 11. The total maximum dose rate considering all actinides amounts to 4×10^{-9} Sv/a. Even lower dose rates were calculated in case of the river pathway (results not shown).

Up to now only results for the NES have been presented. An important part of the consequence analysis deals with the altered evolution scenarios (AES). Future studies will deal with these AES. Among the possible scenarios in which the glass matrix might play a more significant role we mention early failure of the overpack and human intrusion. Such analysis will be made in 2001 within the BENIPA EC project which is in preparation within the EC's 5th Framework Programme.

Ewing et al. [4] illustrated the worth of sensitivity analysis as a means to establish the relative importance of specific submodels by comparing radionuclide releases from human intrusion scenarios with releases from groundwater pathways (the normal evolution scenario) considering the WIPP repository. The groundwater pathway model became increasingly more complex over time and used increasingly more detailed submodels of flow and corrosion chemistry within the repository. These model improvements resulted in a significant decrease of the released radionuclide fluxes. The human intrusion scenario considered direct releases to the surface owing to exploratory drilling. Because the releases from the human intrusion were much more important than those from the groundwater pathways, improvements in the model of contaminant transport proved to be unnecessary [4].

5. Conclusions

The long-term safety of deep disposal of vitrified high-level waste in the Boom Clay layer was assessed on the basis of numerical simulations using the most recent information on the repository design, barrier performance, and transport parameters for the relevant radionuclides. The capability of the Boom Clay layer to confine the radionuclides present in the waste was evaluated on the basis of the calculated radionuclide fluxes and dose rates received by man. The latter was done for the well and river pathway. The results show that the Boom Clay layer is a very efficient barrier in confining most of the radionuclides until their activity has become negligible. The radionuclides that are not or only poorly retained (i.e ., ^{79}Se, ^{129}I, and ^{99}Tc) will reach the aquifer before they have completely decayed, but the magnitude of their fluxes released into the surrounding aquifers are strongly limited by the Boom Clay. Maximum dose rates the well pathway for ^{79}Se, ^{129}I, and ^{99}Tc are, respectively, 9×10^{-6}, 2×10^{-8}, and 5×10^{-9} Sv/a. Concerning the river pathway, maximum dose rates are 7×10^{-8}, 3×10^{-10}, and 5×10^{-11} Sv/a, respectively. Such rates are still low and hundreds times lower than the internationally recommended dose limit.

In the performance assessment study, a number of issues can still be further improved. For instance, considerable uncertainty exists about the amounts of volatile elements present in the waste inventory. This is especially true for ^{129}I and ^{36}Cl, which are not retarded by the Boom Clay. Other uncertainties are related to the dissolution of the glass matrix. However, under the conditions of the normal evolution scenario, the sensitivity analysis shows that the glass matrix has a limited contribution to the overall performance of the disposal system because of the presence of a very effective clay barrier. The contribution from the glass matrix would be more significant if it can be demonstrated that its lifetime exceeds one million years. Finally, the influence of the glass matrix also depends on the solubility limits of the radionuclides. Radionuclides having a low solubility limit under disposal conditions will already show a spread release to which the glass matrix may contribute little. The glass matrix is undoubtedly an integral part of the multi-barrier, and thus adds to the overall repository safety. Furthermore, its functioning is considered independent from the functioning of other barriers. Its precise contribution, however, depends on a number of factors, including the type of scenario considered, the radionuclide properties, etc.

The relative importance of the glass matrix in the overall repository performance should be further explored for other scenarios, including altered evolution scenarios such as the ones discussed here. Only then can a definite statement on its role in the safety of a repository be made.

Acknowledgements

This work has been financially supported by the Belgian National Agency for Radioactive Waste and Nuclear Fuels (NIRAS/ONDRAF).

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