Modelling groundwater contamination above a potential nuclear waste repository in the Columbia River Basalt, USA

Michael O. Schwartz¹

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Abstract

Potential groundwater contamination above the one-time candidate repository for high-level nuclear waste in the Columbia River Basalt (CRB), USA, is evaluated in a flow-transport model with 260,895 elements for a period of 1 million years. The hydrogeologic database originates from the Basalt Waste Isolation Project, which ended in 1987, giving way to the Yucca Mountain project. The CRB data have been scanned recently with the object of finding a storage site for natural gas and a sequestration site for CO₂. However, the impact on the nuclear issue in the USA or other countries with similar large flood-basalt provinces has never been evaluated since the suspension of the project. The peak radioactive dose predicted for a potential repository in the CRB is at least eight orders of magnitude lower than the US generic regulatory limit (0.15 mS/a) even for the most pessimistic scenario.

Keywords Nuclear-waste disposal · Basalt · Groundwater contamination · USA

Introduction

Flood basalt has extraordinary characteristics that make it a prime candidate for a radioactive-waste disposal site. Flood basalt is less common than granite or shale, which are the host rocks of repositories under construction. The best studied occurrence of flood basalt is the Columbia River Basalt (CRB), in Washington, Oregon and Idaho; USA (Fig. 1). A huge amount of geoscientific data have been collected (reviewed in Reidel et al. 2002). This occurred mainly in the period from 1968 to 1987 when the 1200 km² Hanford Site was investigated with the object of identifying a disposal site. The work was suspended, because Yucca Mountain, Nevada, was designated by the “Waste Policy Act as Amended” (DOE 2004) in 1987 to be the deep geological repository for burnt fuel and other types of high-level nuclear waste. On April 14, 2011, the US government decreed to stop funding the Yucca Mountain project. Since then, the US Department of Energy (DOE) has not conducted any site-specific investigations but only generic studies, most of them related to crystalline rocks.

In terms of exploratory drilling density, the CRB study area (1200 km²) and Yucca Mountain study area (1350 km²) are equivalent. However, the CRB project lacks underground workings, whereas the Yucca Mountain project has an exploratory adit.

Prior to the suspension of the CRB project, there have been some rudimentary approaches to 2D modelling of groundwater flow. The results are contradictory and not suitable for a performance assessment (Duguid and Kowall 1981; DOE 1988). With the development of 3D flow-transport parallel codes, the situation has changed.

This paper bridges the time gap between data collection and advent of suitable hard and software. The high hydraulic gradients remain a challenge in terms of numerical stability but some modern codes can cope with these. The study is the first of its kind. Although it is a case study, most of the findings should apply to potential repositories located in basalt in other parts of the world. A possible exception is the groundwater chemistry. The CRB groundwater has a high pH, averaging 9.4, which allows copper-shielded waste containers to be in thermodynamic equilibrium with groundwater, thus minimizing the risk of damage by corrosion.

✉ Michael O. Schwartz
mathgeol@yahoo.de

¹ MathGeol, Postfach 101204, 30833 Langenhagen, Germany

Fig. 1 **a** Location of the Columbia River Basalt (CRB), USA. **b** Pasco Basin with the location of the model area. **c** Map of the model area and traces of cross sections (Fig. 4)

Study area

Hydrogeological database

The 1226-km² model is located in the Pasco Basin in the central part of the CRB. An irregular cover of Quaternary through Upper Tertiary sedimentary deposits overlies the basaltic rocks. The major subdivisions of the CRB are the Saddle Mountains Basalt Formation (isotopic age 6–13 ma), Wanapum Basalt Formation (14.5–15.6 ma), Grande Ronde Basalt Formation (15.6–16.5 ma), and Imnaha Basalt Formation (17.5 ma). Stratigraphically, the model comprises the Quaternary and the Schwana Sequence of the Grande Ronde Basalt Formation (Table 1).

The CRB has been subjected to folding. The best documented tectonic feature is the high-amplitude fold of the Rattlesnake Hills. The topographic high of the model lies in the anticlinal area (Figs. 1, 2). This coincides more or less with the maximum water-level at 360 m a.s.l. (Whiteman et al. 1994). The topographic low is represented by the Columbia River valley (lowest water-level at 110 m a.s.l.).

Hydraulic properties

Reidel et al. (2002) summarised the results of field tests for the interior of basalt flows (22 measurements), flow tops (157) and interbeds (48). Most tests consisted of single borehole tests conducted in boreholes that were progressively drilled and tested. Some tests were conducted in existing boreholes in which test zones were isolated using straddle packers.

The hydraulic conductivity of the basalt interiors ranges from 10^{-16} to 10^{-10} m/s (Fig. 3). The maximum value, approximately corresponding to a permeability of 10^{-17} m², is used for the base-case calculations. The individual interbeds and underlying flow tops are combined to single individual units. These have a permeability of 10^{-15} m², corresponding to the median for the flow tops in the Grande Ronde Basalt Formation. The overburden only occupies a minimal portion of the model and hardly influences the contaminant transport calculations. For the sake of numerical stability, the overburden has the same hydraulic properties as those of the interbeds/flow tops.

The effective porosity, which refers to the interconnected void space, is set at 0.15 for the interbeds/flow tops in agreement with the modelling of gas storage capacity of the CRB (Reidel et al. 2002). The porosity of the flow interiors is set at 0.005.

The repository

According to the latest layout, the CRB repository (Hanford Reference Repository) should be located in the Sentinel Bluffs Sequence of the Grande Ronde Basalt Formation at a depth of 900 m (DOE 1988). An earlier version has an additional option with the Umtanum Flow of the Schwana Sequence at a depth of 1100 m (DOE 1982). The Nuclear Waste Policy Act of 1982 specifies that the first US repository cannot exceed the equivalent of 70,000 MTHM (metric tons of heavy metal, including uranium and other radioactive heavy metals) until a second repository becomes operational. The statutory limit is still valid today and corresponds to 11,629 waste containers (Table 2; DOE 2007a). The horizontal dimensions of the repository are 9000 m × 6000 m (DOE 1982).

Table 1 Hydrostratigraphic units and hydraulic properties of the Columbia River Basalt (CRB)

Formation	Member/sequence	Sediment/basalt stratigraphy	Rock type	Thickness (m)	Base-case permeability (m ²)	Porosity (–)	Repository horizon
Ringold		Upper Ringold	S	50	10 ^{–15}	0.15	
		Lower Ringold	S	17	10 ^{–15}	0.15	
Saddle Mountains Basalt	Elephant mountain	Pomona	B	50	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
			B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
	Esquatzel		B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
	Umatilla		B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
Wanapum Basalt	Priest rapids		B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
	Roza		B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
			B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
	Frenchman springs	Sentinel gap flow	B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
			B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
			B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
			B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
			B	45–55	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
Grande Ronde Basalt	Sentinel bluffs	Flow I	B	50	10 ^{–17}	0.005	Sensitivity case
			FT	17	10 ^{–15}	0.15	
		Rocky coulee flow	B	50	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
		Cohasset flow	B	50	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
		McCoy Canyon flow	B	50	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
	Schwana	Umtanum flow	B	50	10 ^{–17}	0.005	Base case
			FT	17	10 ^{–15}	0.15	
		Very high-Mg flow	B	50	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
		Flow I	B	50	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	
		Flow II	B	50	10 ^{–17}	0.005	
			FT	17	10 ^{–15}	0.15	

S sediment, *B* basalt interior, *FT* flow tops/interbeds

There are various disposal schemes for the CRB repository. The latest studies indicate that copper-shielded waste containers emplaced in a bentonite buffer are the preferred option (Lutton et al. 1986). Thus, the CRB concept would be the equivalent to the KBS-3 scheme of the Scandinavian repositories under construction (SKB 2006). Accordingly, the containers are emplaced in deposition holes driven from deposition tunnels, which are connected to a

nearly horizontal system of access and transport tunnels. The containers that host the waste have a 5-cm-thick corrosion-resistant copper shell around an inner deformation-resistant cast-iron canister; the containers are embedded in a 35-cm-thick bentonite buffer.

Fig. 2 Block diagram of the CRB model mesh showing nodal positions. Ten-fold vertical exaggeration (z direction)

Fig. 3 Histograms showing the hydraulic conductivity (m/s) of the Saddle Mountains Basalt Formation, Wanapum Basalt Formation and Grande Ronde Basalt Formation of the Columbia River Basalt (CRB)

Model setup

Geometry

The model mesh calculated with WINGRIDDER (Pan 2007) has a total of 260,895 elements and a maximum of 37 layers. The mesh is irregular vertically and regular horizontally, and measures $38,800 \times 31,600 \times 1510$ m (Figs. 2, 4). The nodal distance in the x and y directions is 400 m. The top layer has nodal z values representing the water-level (90–360 m a.s.l.; Fig. 5). The following layers have a thickness that roughly represents the thickness of the interior of the individual basalt flows (colonade/entablure), on the one hand, and flow tops (including interbeds), on the other hand. The bottom of the mesh is the -1150 -m level.

The top layer, which serves to maintain constant temperature, pressure and salinity, exclusively consists of infinite-volume boundary elements (10^{52} m³). The bottom layer cells have a volume of 10^{12} m³. This volume is large enough for maintaining nearly constant temperature and salinity throughout the simulation period of one million years but simultaneously flexible enough to account for pressure adjustments. The volume of the remaining cells is calculated according to their nodal positions.

The water-level remains fixed in the base case (110–360 m a.s.l. range). Transient conditions are calculated in a sensitivity case, which simulates sea-level lowering. In the simulation year 50,000, the water-level changes from the 110–360 m range to the 90–360 m range, whereby the values above 170 m a.s.l. remain unchanged, because they have relatively little influence on contaminant transport. The transient condition corresponds to an increase in the hydraulic gradient by a factor of 1.33 in the domain with water-level values < 170 m a.s.l..

Boundary conditions and initial conditions

The infinite-volume elements (10^{52} m³) of the top layer, which impose Dirichlet conditions, have a pressure of 10^5 Pa, a temperature of 14 °C and zero salinity. The large volume elements of the bottom layer (10^{12} m³) impose nearly Dirichlet conditions with respect to temperature (64 °C) and salinity (0.0017 mass fraction brine or 0.00042 mass fraction salt), but allow flexible pressure adjustments. The remaining layers have initial values along the gradients defined by the top and bottom layers, which are close to the present-day values. These boundary conditions and an initialisation pre-run with a high diffusivity of brine (10^{-7} m²/s) allow suitable vertical gradients throughout the actual simulation with standard brine diffusivity (10^{-9} m²/s), unless disturbed by radioactive heat sources.

The definition of the outer horizontal boundaries is straightforward on three sides of the model. They are treated as closed boundaries, because they are more or less aligned along natural boundaries: the Columbia River acts as discharge area NNE and ESE; and the Rattlesnake Hills act as drainage divide SSW (Figs. 1, 5).

Only a small section of the WNW boundary is well defined. This is the Cold Creek Fault, which acts as aquiclude. The potentiometric surface on the WNW side of the fault (i.e., outside the model area) is tens of meters above the potentiometric surface on the ESE side (i.e., inside the model area; Reidel et al. 2002, Fig. 3.4). Thus, the hydraulic gradient in the direction at right angle to the fault (WNW–ESE direction) is near zero for a length of 15 km. This is the area occupied by the potential repository (Fig. 5).

Table 2 Setup of the Columbia River Basalt (CRB) model

General properties	
Simulation period (a)	1,000,000
Rock grain density (kg/m ³)	2800
Rock grain specific heat (J/kg °C)	950
Formation heat conductivity (W/m °C)	2.3
Number of simultaneous contaminant sources (–)	1, 10
Repository	
Capacity (MTHM) ^a	70,000
Number of waste containers (–)	11,629
Depth (m)	900 [Sentinel bluffs sequence] 1100 [Umtanum flow]
In-situ temperature (°)	50 [Sentinel bluffs sequence] 57 [Umtanum flow]
Initial inventory	
Am-241 (Bq)	8.65×10^{18}
Cs-135 (Bq)	1.56×10^{15}
I-129 (Bq)	9.55×10^{13}
Np-237 (Bq)	1×10^{15}
Pa-231 (Bq)	1.01×10^{13}
Pu-240 (Bq)	1.44×10^{18}
Pu-242 (Bq)	6.40×10^{15}
Se-79 (Bq)	2.15×10^{14}
Tc-99 (Bq)	4.02×10^{16}
Th-230 (Bq)	1.01×10^{12}
U-233 (Bq)	1.10×10^{14}
U-234 (Bq)	3.41×10^{15}
U-236 (Bq)	7.67×10^{14}
U-238 (Bq)	9.04×10^{14}
Half-life	
Am-241 (a) ^b	4.33×10^2
Cs-135 (a)	2.3×10^6
I-129 (a)	1.57×10^7
Np-237 (a) ^b	2.14×10^6
Pa-231 (a)	3.28×10^4
Pu-240 (a) ^c	6.56×10^3
Pu-242 (a) ^d	3.75×10^5
Se-79 (a)	2.9×10^5
Tc-99 (a)	2.13×10^5
Th-230 (a) ^c	7.54×10^4
U-233 (a) ^b	1.59×10^5
U-234 (a) ^e	2.46×10^5
U-236 (a) ^c	2.34×10^7
U-238 (a) ^d	4.47×10^9
Distribution coefficient	
Cs-135 (mL/g)	–
I-129 (mL/g)	–
Np-237 (mL/g)	200
Pa-231 (mL/g)	1000
Pu-240 (mL/g)	20
Pu-242 (mL/g)	20
Se-79 (mL/g)	8
Tc-99 (mL/g)	29

Table 2 (continued)

Th-230 (mL/g)	5000
U-233 (mL/g)	40
U-234 (mL/g)	40
U-236 (mL/g)	40
U-238 (mL/g)	40
Diffusivity	
Cs-135 (m ² /s)	9×10^{-13}
I-129 (m ² /s)	8×10^{-15}
Np-237 (m ² /s)	4×10^{-14}
Pa-231 (m ² /s)	4×10^{-14}
Pu-240 (m ² /s)	4×10^{-14}
Pu-242 (m ² /s)	4×10^{-14}
Se-79 (m ² /s)	4×10^{-14}
Tc-99 (m ² /s)	4×10^{-14}
Th-230 (m ² /s) ^c	4×10^{-14}
U-233 (m ² /s)	4×10^{-14}
U-234 (m ² /s)	4×10^{-14}
U-236 (m ² /s)	4×10^{-14}
U-238 (m ² /s)	4×10^{-14}
Solubility	
Cs-135 (M/L)	Unlimited
I-129 (M/L)	Unlimited
Np-237 (M/L)	10^{-15}
Pa-231 (M/L)	3×10^{-7}
Pu-240 (M/L)	10^{-9}
Pu-242 (M/L)	10^{-9}
Se-79 (M/L)	10^{-15}
Tc-99 (M/L)	10^{-12}
Th-230 (M/L)	10^{-9}
U-233 (M/L)	10^{-8}
U-234 (M/L)	10^{-8}
U-236 (M/L)	10^{-8}
U-238 (M/L)	10^{-8}
Release fraction	
Cs-135 (–)	0.13
I-129 (–)	0.13
Np-237 (–)	0.0001
Pa-231 (–)	0.001
Pu-240 (–)	0.001
Pu-242 (–)	0.001
Se-79 (–)	0.0001
Tc-99 (–)	0.0001
Th-230 (–)	0.001
U-233 (–)	0.001
U-234 (–)	0.001
U-236 (–)	0.001
U-238 (–)	0.001
Biodose conversion factor	
Cs-135 (Sv/a:Bq/m ³)	1.45×10^{-8}
I-129 (Sv/a:Bq/m ³)	1.29×10^{-7}
Np-237 (Sv/a:Bq/m ³)	2.47×10^{-7}
Pa-231 (Sv/a:Bq/m ³)	2.44×10^{-6}

Table 2 (continued)

Pu-240 (Sv/a:Bq/m ³)	9.51×10^{-7}
Pu-242 (Sv/a:Bq/m ³)	9.07×10^{-7}
Se-79 (Sv/a:Bq/m ³)	2.42×10^{-8}
Tc-99 (Sv/a:Bq/m ³)	1.12×10^{-9}
Th-230 (Sv/a:Bq/m ³)	1.08×10^{-6}
U-233 (Sv/a:Bq/m ³)	8.97×10^{-8}
U-234 (Sv/a:Bq/m ³)	8.19×10^{-8}
U-236 (Sv/a:Bq/m ³)	7.67×10^{-8}
U-238 (Sv/a:Bq/m ³)	7.87×10^{-8}

^aMetric tons heavy metal (uranium and other radioactive heavy metal)^bDecay sequence: Am-241, Np-237, U-233^cDecay sequence: Pu-240, U-236^dDecay sequence: Pu-242, U-238^eDecay sequence: U-234, Th-230 (U-234 diffusivity used for Th-230) **Fig. 4** Vertical cross sections of the CRB model

There are not enough data that allow to define the remaining part of the WNW boundary either as closed or open. As a consequence, the whole of the WNW boundary is treated as closed to avoid unnecessary bias. This has little practical

consequences. However, it must be kept in mind that this boundary definition is an idealization and the actual shape of the contaminant plume would have more irregularities on

Fig. 5 **a** Present-day water level of the base case. **b** Water level in the year 50,000 (sensitivity case only)

Fig. 6 Repository-wide thermal load (kW) (DOE 2008a)

its WNW side than shown in the figures of “Radionuclide transport”.

Time-dependent Neumann boundary conditions that allow for heat production of the nuclear waste are imposed on all cells in the 9000×6000 m repository area of the Sentinel Bluffs Sequence or Umtanum Flow of the Schwana Sequence. The heat production of the radioactive waste (Fig. 6; DOE 2008a) is divided by 55, which is the number of canisters divided by the number of repository cells, and is then used as heat-generation data for the individual cell.

Furthermore, time-dependent Neumann boundary conditions that allow for contaminant injection (Fig. 7) are imposed

on one cell in the central repository area (base case) and additional nine cells on the periphery of the repository (sensitivity case). The injection rate “ R_i ” for the radionuclide “ i ” (Bq/a) in the simulation year “ n ” is calculated with the simplification that the fraction of the inventory released per year remains constant during the simulation period:

$$R_i = \frac{I_i}{N} \times \left(1 - \frac{F_i}{10^6 - s}\right)^{n-s} \times \left(F_i \times \frac{1}{10^6 - s}\right) \quad (1)$$

with “ I_i ” being the inventory of the radionuclide “ i ”, “ F_i ” being the fraction of the radionuclide “ i ” released until the end of the simulation period (1 million years), “ s ” being the year in which the radionuclide release starts (simulation year 200), and “ N ” being the number of canisters (11,629). A conservative approach is used for defining the release fraction, based on data of DOE (2008b):

- $F_i = 0.13$ for radionuclides without solubility limits.
- $F_i = 0.001$ for radionuclides with high solubility limits.
- $F_i = 0.0001$ for radionuclides with low solubility limits.

Solubility limits for 10 radionuclides out of a total of 13 are provided by Salter and Jacobs (1982) together with distribution coefficients for CRB groundwater. The missing data (I-129, Pa-231 and Th-230) are taken from a compilation for low-salinity granite groundwater, together with diffusivities for all radionuclides (Table 2; Lindgren and Lindström 1999).

Computer code

TOUGHREACT version 3 is a numerical simulation programme for chemically reactive non-isothermal flows of multi-phase fluids in porous and fractured media (Xu et al. 2014). The programme was developed by introducing reactive chemistry into the multi-phase flow code TOUGH2 (Pruess et al. 1999). The governing equations are discretised using integral finite difference for space and fully implicit first-order finite difference in time. All simulations of this study are performed with the EOS7 module, which treats the aqueous phase as a mixture of water and brine. The ECO2N module, which treats the aqueous phase as a mixture of water and salt, fails at initialisation but would be suitable for the actual simulation.

Theoretically, it is possible to use the EOS7R module of the other members of the TOUGH2 family of codes (including iTOUG2; Finsterle 2000, and TOUGH2-MP; Zhang et al. 2008) for the non-reactive transport simulations. However, the convergence behaviour of the EOS7R module in all versions of the TOUGH2 code is by far inferior to that of the EOS7 module of TOUGHREACT version

Fig. 7 Annual near-field release of radionuclides (Bq/a) from one defective waste container from year 200 to year 1,000,000

3. The FEHM code (Zyvoloski et al. 2011) has excellent convergence behaviour, but is too slow for long simulation periods and large models.

Radionuclide transport

The near-field base case assumes that one canister randomly chosen in the repository layout has a production defect. The variability of the possible contaminant plumes is depicted by selecting one source (base case) and nine additional sources (sensitivity case) at the centre and the periphery of the repository, respectively (Fig. 1c). The plumes emanating from the remainder of the canister locations would be intermediate between these geometric extremes (not calculated).

The radioactive dose “ D ” (Sv/a) uses the biodose conversion factor (BDCF), representing the all-pathway dose (DOE 2007b). The future radiation dose to a hypothetical “reasonably maximally exposed individual” (RMEI) considers the most important exposure pathways:

- water intake;
- radiation exposure to contaminated soil;
- inhalation of radioactive particles and gases;
- consumption of contaminated plants;
- consumption of contaminated animal product foodstuff.

The dose is calculated according to the equation:

$$D = \sum X_i B_i, \quad (2)$$

Fig. 8 Maps of the second topmost layer of the CRB model with fixed water-level showing the radioactive dose (Sv/a) in the simulation year 200,000 and 1,000,000. **a** Base case with 1 defective canister. **b** Sensitivity case 1 with base-case permeabilities multiplied by 10 as well as base-case repository depth and 1 defective canister. **c** Sensitivity case 2 with base-case repository depth decreased by 200 m as well as base-case permeabilities and 1 defective canister. **d** Sensitivity case 3 with 10 defective canisters as well as base-case permeabilities and base-case repository depth

where “ X_i ” is the concentration (Bq/L) of the radionuclide “ i ” in the second topmost cell and “ B_i ” is the BDCF (Sv/a:Bq/L) of the radionuclide “ i ”.

Fig. 9 Peak radioactive dose (Sv/a) of the second topmost layer of the CRB model. The base case refers to fixed water-level and one defective canister. The sensitivity case 1 refers to base-case permeabilities multiplied by 10 as well as fixed water-level, base-case repository depth and 1 defective canister. The sensitivity case 2 refers to base-case repository depth decreased by 200 m as well as base-case permeabilities, fixed water-level and 1 defective canister. The sensitivity case 3 refers to 10 defective canisters as well as fixed water-level, base-case repository depth and base-case permeabilities. The sensitivity case 4 refers to transient water-level (increase of the hydraulic gradient by a factor of 1.33 in the simulation year 50,000) as well as base-case permeabilities, base-case repository depth and 1 defective canister

The peak doses are the relevant parameters for the various release paths (Figs. 8, 9). At the end of the simulation period, the peak doses are far below the US generic regulatory limit (0.15 mSv/a; GAO 2000) or the limit for Yucca Mountain, USA (0.15 mSv/a < 10,000 years and 1 mSv/a > 10,000 years; NRC 2014).

Note that these low doses are calculated for an error scenario, e.g., production defects in the waste container or emplacement errors. In the standard scenario, a copper-shielded waste container emplaced in a bentonite buffer with admixtures of hematite (Fe_2O_3) should not release any radionuclides to the far-field environment. Native copper in average CRB groundwater in the presence of hematite is thermodynamically stable (Fig. 10) or, at least, close to thermodynamic equilibrium. Even if the copper shell is corroded by aqueous sulphide diffusing through the bentonite barrier, the corrosion rates will be extremely low (Schwartz 2018). Thus, the lifetime of a copper-shielded waste container is far beyond the 1 million years envisaged for the CRB repository.

Fig. 10 Eh–pH diagram for the system Cu–(Fe)–S–Cl–O–H at 25 °C and 0.1 MPa. The thermodynamic data are calculated with the SUPCRT92 program (Johnson et al. 1992) except for CuCl^0 (Xiao et al. 1998) and $\text{FeSO}_4(\text{c})$ (Hemingway et al. 2002). Two types of Cu^0 – Cu_2S boundaries are shown: the black lines refer to the Fe-free system with ΣS of 10^{-2} and 10^{-5} ; the red interrupted line shows the limit of the Cu^0 stability field for $\Sigma\text{S}=10^{-5}$ in the presence of hematite and $\text{FeSO}_4(\text{c})$

Sensitivity tests

The sensitivity tests address the greatest uncertainties in a performance assessment:

- permeability in regions with undetected vertical fractures;
- location of failed waste containers;
- number of failed waste containers;
- climate change and associated sea-level lowering.

The interiors of the basalt flows (colonnade/entablature) have an extremely low permeability, the median being 10^{-20} m^2 . The base case uses the maximum recorded value of 10^{-17} m^2 . It is possible that this assumption is not conservative enough. To capture the effect of undocumented vertical fractures, the first sensitivity case has all permeabilities increased by one order of magnitude. The second sensitivity test considers the decrease of the repository depth from 1100 to 900 m. The third sensitivity test considers the increase of the number of defective canisters from one (base case) to ten.

All the above sensitivity cases have in common that the ratio between base-case values, on the one hand, and sensitivity-case values, on the other hand, decrease with time. In other words, the peak dose of the pessimistic sensitivity cases tends to converge toward that of the base case (Figs. 8, 9). Of course, the areal extent of the contaminant plume for the sensitivity case with ten defective canisters is much

larger than that for the cases with only one defective canister (Fig. 8).

Introducing transient water-level conditions has a small effect. An increase in the hydraulic gradient by a factor of 1.33 in the year 50,000, simulating sea-level lowering, results in a maximum increase in the peak radioactive dose by a factor of approximately 3.

Except for the speculative high-permeability case, the contaminant travel times are extremely long. Hardly any contaminant would come near the potentiometric surface before 100,000 years when field-test supported permeabilities are used for the calculations. This is in perfect agreement with groundwater age modelling. The deep CRB groundwater is very old, at least 100,000 years according to helium data (Reidel et al. 2002).

Conclusions and discussion

Most of the countries that produce high-level nuclear waste have the option to choose between granite and clay when looking for a disposal site. Some have the additional alternative of a basalt repository such as the USA.

In terms of heat conductivity, a quartz-poor rock such as continental flood basalt or clay has a disadvantage relative to a quartz-rich rock such as granite. The bentonite buffer around a waste container emplaced in basalt or clay is heated to higher temperatures than the buffer in a granitic environment. Since most high-level waste producing countries postpone the waste disposal into the distant future, the heat issue will become less important. For example, the heat output of 50-year-old waste would decrease to half of its original value within 40 years.

In terms of groundwater salinity, continental flood basalt has an advantage over clastic sedimentary rock such as clay. The same is true with respect to granite when fractures allow the intrusion of basinal brines. Salinity is critical, because high salt concentrations reduce the corrosion resistance of the copper shell around the waste container (Schwartz 1996, 2008).

In terms of vertical permeability, continental flood basalt has an advantage over granite with near-vertical fractures. The possibilities to offset the disadvantage for granite by increasing the repository depth are limited by the salinity gradient of granite groundwater.

For groundwater pH, a general statement is not possible. The only deep basalt groundwater with sufficient data is the CRB groundwater. The high pH, which averages 9.4 in the Grande Ronde Basalt Formation, would allow the copper shell around the waste container to be at or near thermodynamic equilibrium. The prerequisite is a sulphide-free bentonite buffer with admixtures of hematite (Fe_2O_3). Even though the reason for the high pH is not fully understood,

the CRB data justify to investigate deep basalt groundwater in other parts of the world.

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