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LA-10532-MS

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JAN 12 1988

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***Two-Dimensional Numerical Simulation of
Geochemical Transport in Yucca Mountain***

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Prepared by Nevada Nuclear Waste Storage Investigations (NNWSI) Project participants as part of the Civilian Radioactive Waste Management Program. The NNWSI Project is managed by the Waste Management Project Office of the US Department of Energy, Nevada Operations Office. NNWSI Project work is sponsored by the Office of Geologic Repositories of the DOE Office of Civilian Radioactive Waste Management.

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Two-Dimensional Numerical Simulation of Geochemical Transport in Yucca Mountain

B. J. Travis
H. E. Nuttall*

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TWO-DIMENSIONAL NUMERICAL SIMULATION OF GEOCHEMICAL TRANSPORT IN YUCCA MOUNTAIN

by

B. J. Travis and H. E. Nuttall

ABSTRACT

Several physical and chemical processes can affect transport of radionuclides in Yucca Mountain. Geometric spreading and lateral flow will reduce the concentration of contaminated water reaching the accessible environment. Travel times to the water table are calculated to be at least several tens of thousands of years. Colloid transport can also enhance radionuclide transport, but will be important only if fracture flow is. Finally, the heat load from repository waste can alter the distribution of some naturally occurring minerals. In particular, dissolution and precipitation of SiO_2 should lead to a region of reduced permeability around waste canisters.

Introduction

The Nevada Nuclear Waste Storage Investigations (NNWSI) Project of the US Department of Energy is studying the suitability of an area in southern Nevada as a potential repository for high-level radioactive waste. The investigations include site and regional studies of the Nevada Test Site (NTS) area to determine whether the site is technically acceptable. Los Alamos researchers are contributing to these investigations through studies of the geochemistry, mineralogy, and petrology of Yucca Mountain and its environs and through engineering design for the exploratory test shaft. Our goal is to ascertain whether the site will retard the transport of radionuclides to the accessible environment over the lifetime of the waste package.

Figure 1 presents a cross section of Yucca Mountain. The potential site lies in the Topopah Spring Member, some 300 m below the surface and about 200 m above the water table. The stratigraphy is detailed in a recent report.¹ At the repository location Yucca Mountain has five stratigraphic units: (1) Surficial units are alluvium and densely to moderately welded, fractured Tiva Canyon tuff. (2) Below this lies the upper clastic unit, which has bedded

and nonwelded tuffs of the Yucca Mountain and Pah Canyon Paintbrush tuffs. This unit is highly porous and relatively (matrix) permeable. (3) The next unit down is densely welded with a thin upper and lower vitrophyre and a thick central zone of densely welded ashflow. The matrix permeability of this central zone of the Topopah Spring member is very low, but the rock is highly fractured. (4) Below this lies the lower clastic unit, which contains nonwelded ashflow, ashfall, and reworked tuffs (the lowest part of the Topopah Spring tuff and the Calico Hills tuff). This unit is highly porous and relatively permeable. (5) The bottom unit consists of several nonwelded to moderately welded tuffs with some thinner layers of densely welded ashflow and bedded ashfall tuffs. These rocks appear to be moderately fractured. Much of this unit is below the water table.

In a previous report,² we estimated the effects of lithology and fractures on water flow and radionuclide transport in Yucca Mountain. We presented a sensitivity analysis of transport along a one-dimensional vertical pathway to the water table. The effect of waste heat on near-field water saturation was also estimated.

In this report, we address several questions not treated in that last report.

- 1) How will geometric spreading and lateral flow affect transport?
- 2) Will waste-heat-induced hydrologic transients significantly alter distribution of salts and minerals in pore water near waste canisters?
- 3) Under what conditions will colloidal transport be important?

Geometric Effects

In a previous study,² we estimated water flow and radionuclide transport through fractured Yucca Mountain tuffs. Travel times to the water table without fracture flow are very large because of the low recharge rate and because of sorption retardation. With fracture flow, travel times are less, but are still on the order of 10 000 yr for nonsorbing ions such as technetium-99 because of diffusion from fractures into the porous tuff matrix. In those studies, we assumed a source of infinite horizontal extent. A more realistic calculation of travel time would account for the finite extent of the source--namely, the placing and spacing of individual canisters. In addition, likely concentrations of nuclides at the canisters were not known, so only relative concentrations could be calculated.

Two calculations, based on the TRACR3D code,³ are described here that compute radionuclide transport from discrete sources for two flow scenarios. Processes included in these calculations are steady advection, molecular diffusion, hydrodynamic dispersion, radioactive decay, and equilibrium adsorption. In the first situation, flow is assumed vertical from the repository to the water table and then horizontal in the saturated zone. The canisters are assumed to be in long regularly spaced rows as shown in Fig. 2. Because of symmetry, we need calculate transport only between a vertical line through a canister and a vertical line midway between that canister and the next row. An average vertical recharge of 4.5 mm/yr⁴ is assumed. The radionuclides included in the calculation are listed in Table A-1 with some of their transport properties. In the highly fractured Topopah Spring tuff, transport is assumed to be primarily in fractures. For 100- μ m fractures with a 5-cm spacing,¹ fracture flow rate is approximately 2.3 m/yr, derived as follows:

$$V_c \cdot A_c = V \cdot A \rightarrow V_c = \frac{VA}{A_c} = 4.5 \text{ mm/yr} \times \frac{5 \text{ cm}}{10^{-2} \text{ cm}} = 225 \text{ cm/yr} .$$

In the Topopah Spring layer, we use equivalent fracture water flow rates V_c , for conservatism. Because of diffusion into the matrix, the effective flow rate for radionuclides must be modified. Sudicky and Frind⁵ have worked out an expression for this. The effective fracture transport velocity V_E in a medium with regular fracture spacing is

$$V_E = V_c / \left(\frac{\gamma}{2} \left\{ -1 + \left[1 + \frac{4}{\gamma} (1 + \beta) \right]^{1/2} \right\} \right)$$

where

$$\gamma = \frac{V_c^2}{\lambda DR} , \quad \beta = \frac{\theta \sqrt{R'D'}}{bR/\lambda} \tanh (\sigma/\lambda), \quad \lambda = \frac{\ln 2}{t_{1/2}} ,$$

$$D = \alpha_L V_c + D^*, \quad D' = \tau D^*, \quad \sigma = \sqrt{\frac{R'}{D'}} (B-b),$$

TABLE A-1
PROPERTIES OF TRANSPORTED RADIONUCLIDES

<u>Radionuclide*</u>	<u>Half-Life (yr)</u>	<u>Diffusivity (10^{-2} m²/yr)</u>
¹⁴¹ Ce	90	3.15
¹³⁵ Cs	3×10^6	6.30
⁹⁰ Sr	28	2.44
⁸⁹ Tc	2×10^5	3.15
²³⁸ U	4.5×10^9	3.15
²³⁷ Np	2×10^6	3.15
²³⁹ Pu	2.4×10^4	3.15
²⁴³ Am	8×10^3	3.15
¹⁵² Eu	90	3.15
¹³³ Ba	1.6×10^3	3.15

* ¹⁵²Eu and ¹³³Ba are analogues for ¹⁵¹Sm and ²²⁶Ra.

$t_{1/2}$ = half-life, θ = matrix porosity, b = fracture half-width, R' = matrix retardation coefficient, R = crack face retardation coefficient, V_c = steady fracture water velocity, D^* = molecular diffusivity, α_L = longitudinal dispersivity, τ = matrix tortuosity, B = half of the fracture spacing. For this study, we used $R=1$, $\theta = 0.11$ for Topopah Spring, $\theta = 0.20$ for Prow Pass, $b = 5 \times 10^{-3}$ cm, $B = 2.5$ cm, $\tau = 0.1$, $\alpha_L = 10$ cm, $V_c = 225$ cm/yr. This modification is like a retardation factor due to matrix diffusion from fractures, and is reflected in the retardation values given in Table A-2.

In the bedded tuff layer at the bottom of the Topopah Spring layer and in the Calico Hills formation, steady porous flow is assumed, with a flux of 4.5 mm/yr.

For our calculations, we include the bottom 50 m of the Topopah Spring unit, a 15-m-thick bedded tuff layer, 135 m of Calico Hills tuff, and the top 50 m of the Prow Pass unit.

Concentration of radionuclides in the water⁶ passing through canisters has the time histories shown in Table A-3. Linear interpolation is used for times between table entries. Concentrations are determined from a saturation-limited dissolution model,⁶ assuming high-level waste storage. Radioactive decay of the source is reflected in Table A-3. This gives the largest release for several possible waste conditions.

The results of the numerical simulation are shown in Figs. 3 through 22. Contour plots of concentration for several radionuclides are shown at selected times.

Transport times to the water table are considerably greater than in the previous study,² in part due to the more reasonable fracture flow rate used here for the Topopah Spring unit. The fastest moving species ⁹⁹Tc takes 15 000 yr to reach the water table. However, the concentration at the water table is six orders of magnitude smaller than in the source. By 20 000 yr, concentration of ⁹⁹Tc at the bottom of the Calico Hills is about 50 times lower than in the source. In addition, the chromatographic effect of dispersion in the tuff is evident. Because of the varying retardation factors, the migrating radionuclides are segregated from the initial mixture at the canister. In addition, we can estimate the absolute concentrations reaching the water table. These values are considerably less than at the canisters,

TABLE A-2

RETARDATION FACTORS FOR RADIONUCLIDES

Element	<u>Stratigraphic Unit</u>			
	<u>Topopah Spring*</u>	<u>Bedded Tuff</u>	<u>Calico Hills</u>	<u>Prow Pass**</u>
^{141}Ce	2000/ 1.11×10^4	3×10^4	7000	5000/ 3.03×10^4
^{135}Cs	6000/ 3.29×10^5	7×10^4	4×10^4	1200/ 1.2×10^5
^{90}Sr	1100/3865	8×10^4	2×10^4	140/ 3.7×10^3
^{99}Tc	7/389	12	2	2/201
^{238}U	40/2176	40	33	16/1530
^{237}Np	150/8288	100	50	40/4001
^{239}Pu	1300/ 6.50×10^4	540	630	480/ 4.632×10^4
^{243}Am	2.5×10^4 / 5.13×10^6	820	2×10^4	3000/ 2.12×10^5
^{152}Eu	10^4 / 1.78×10^4	3×10^4	2×10^4	10^4 / 3.67×10^4
^{133}Ba	2×10^4 / 1.61×10^5	2×10^5	7×10^5	4000/ 1.51×10^5

*For Topopah Spring and Prow Pass, the first number is matrix retardation coefficient; the second is the effective retardation coefficient for fracture flow with diffusion from fractures.

**In the fault, retardation values for fractured Prow Pass are used.

TABLE A-3

SOURCE CONDITIONS: CALCULATED CONCENTRATIONS IN
WATER PASSING THROUGH CANISTER (moles/l)

Element	Solubility Limit	Time After Emplacement (yr)			
		10^2	10^3	10^4	10^5
^{243}Am	10^{-8}	6.16×10^{-9}	8.59×10^{-9}	9.96×10^{-9}	3.22×10^{-12}
^{14}C	1.	3.14×10^{-9}	2.82×10^{-9}	9.52×10^{-10}	1.77×10^{-14}
^{135}Cs	1.	2.44×10^{-7}	2.44×10^{-7}	2.43×10^{-7}	2.37×10^{-7}
^{237}Np	3×10^{-3}	2.09×10^{-7}	2.28×10^{-7}	2.32×10^{-7}	2.26×10^{-7}
^{239}Pu	1.8×10^{-6}	1.2×10^{-8}	1.48×10^{-8}	2.88×10^{-8}	3.78×10^{-9}
^{226}Ra	10^{-7}	7.2×10^{-16}	3.65×10^{-14}	1.54×10^{-12}	1.20×10^{-11}
^{90}Sr	9.4×10^{-4}	6.06×10^{-8}	3.02×10^{-17}	0	0
^{99}Tc	1.	8.56×10^{-7}	8.54×10^{-7}	8.28×10^{-7}	6.18×10^{-7}
^{126}Sn	10^{-9}	2.99×10^{-10}	2.98×10^{-10}	2.85×10^{-10}	1.77×10^{-10}
^{238}U	2.1×10^{-4}	2.18×10^{-6}	2.18×10^{-6}	2.18×10^{-6}	2.18×10^{-6}

due to retardation and decay and lateral spreading. Not all the radionuclides were calculated. Of those that were, ^{99}Tc is the fastest traveling, then ^{238}U and ^{237}Np , then ^{239}Pu ; ^{243}Am migrates very little because of the very high retardation onto Topopah Spring tuff. Although the remaining nuclides were not calculated, they would lie between the results for ^{239}Pu and ^{243}Am . Other than ^{99}Tc , none of the radionuclides will reach the water table in less than 100 000 yr.

Transport through the Topopah Spring unit is seen to follow a narrow path because of the fracture flow assumption. In the remaining units (below 50 m depth), porous flow is assumed. Lateral spreading is much stronger because of the diffusive darcy flow.

In the second calculation, lateral flow is included. The horizontal scale is considerably greater, encompassing one end of the repository and some distance beyond, as shown in Fig. 23. In the calculation, a vertical section 250 m thick is considered: the bottom 50 m of Topopah Spring tuff, a 15-m-thick bedded tuff layer, a 135-m-thick Calico Hills unit, and the top 50 m of the Prow Pass. The horizontal extent is 250 m. We assume 32-m spacing between waste-canister rows. The last five rows of a hypothetical repository are included. Then a fault has been included 100 m horizontal distance from the last row of canisters. An additional 20 m horizontal distance beyond the fault brings us to the edge of the computed region. The conceptual hydrologic model discussed in Ref. 4 includes the possibility of lateral flow at the interface between the Calico Hills and the Topopah Spring units, and within the Calico Hills, and vertical flow down structural pathways (faults). The spacing used here between the end of the repository and a fault is arbitrary. The ratio of vertical to lateral flow rates and the rate of flow down faults are difficult to determine at this time. We are forced to use what we hope are reasonable estimates and expect the transport behavior that results to be qualitatively correct. Table A-4 lists vertical and horizontal flow rates used for each unit. Flow structure was not calculated. In this simulation, average vertical and horizontal velocities are assigned to each material region. In the fault region, we used retardation values calculated in Table A-2 for fractured Prow Pass tuff. Results of the second calculation are included in Figs. 24 to 37. Arrival times at the water table are essentially the same as in the previous calculation. Concentrations show some increase downstream in the lateral flow region between the Topopah Spring and Calico Hills units. The

lateral flow causes an overall shift of the concentration patterns but not to a large extent. In addition, in this calculation, lateral flow is not strong enough to transport radionuclides 100 m downstream to the fault. Retardation, diffusion, and vertical flow move species down into the Calico Hills

TABLE A-4
FLOW RATES USED IN SIMULATION (mm/yr)

<u>Unit</u>	<u>Vertical</u>	<u>Horizontal</u>
Topopah Spring*	2250	0
Bedded Tuff	3.5	3.0
Calico Hills	3.5	1.0
Fault*	3000	0
Prow Pass*	0	5000

*Fracture flow assumed.

before they can travel laterally to the fault through the bedded tuff layer. Eventually, however, species will get into the fault due to lateral movement in the Calico Hills unit. For the parameters of this calculation, the fault actually has little impact on transport to the water table. Absolute concentrations at the water table are quite small, due to lateral spreading and to retardation. Travel time to the water table for the fastest moving species, ^{99}Tc , is still about 15 000 yr. Travel time here means first arrival of solute molecules at a concentration of 10^{-6} of the source value. There can be no single, unique travel time because of dispersion/diffusion. Arrival of some particular concentration level or some kind of average time must be used.

It is important to remember that we are assuming a recharge rate of 4.5 mm/yr, which may be high. The results shown should scale roughly linearly with the recharge rate. Only two nuclides (^{99}Tc and ^{238}U) are considered. None of the others will even reach Calico Hills layer within 100 000 yr; therefore it would be meaningless to calculate them.

Effect of Thermal Pulse on Silicate Distribution

In our previous study,² we examined numerically the effects of waste heat on near-field hydrology. We found that a dry region developed around individual canisters and that this dry region was surrounded by a shell of elevated saturation. This condition persisted for several decades. There were major uncertainties, however, in material and hydrologic properties at that time. This heat load effect is being re-examined because we now have a more accurate picture of the hydrologic properties of Yucca Mountain tuffs and of the waste decay curve.

We are participating in a computer code benchmarking exercise coordinated by Sandia National Laboratories in which the heat-load effect is the target problem. We present some results from that benchmarking exercise here because we are considering an additional process--the variation in mineral concentration and deposition patterns due to heat-induced hydrologic changes.

Water from well J-13 has the composition listed in Table B-1. One of the main constituents is SiO_2 . SiO_2 can exist in several forms--quartz and cristobalite are two examples. In a sample of Topopah Spring tuff used in a heat alteration study,⁷ the mineral content was found to be 80-90% glass, 5-10% alkali feldspar, and 5-10% SiO_2 (cristobalite). The solubility of SiO_2 is strongly temperature dependent⁸ (about 10^{-3} M at 23°C and about 10^{-2} M at 150°C). In the heat alteration study described in Ref. 7, a great amount of SiO_2 had dissolved at elevated temperature (152°C) and then precipitated upon cooling. In addition, a great amount of clay was deposited. Placement of waste canisters will lead to the development of a temperature gradient in the surrounding Topopah Spring rock. This temperature gradient will then induce a solubility gradient. If water also flows, SiO_2 will be redistributed. Depending on the magnitude and duration of the heat-induced flow, SiO_2 may accumulate in some region. Deposition will result in reduced pore size, which will lower permeability. At late times, a region of reduced permeability around

a canister will isolate the canister from infiltrating water, a beneficial effect.

TABLE B-1
COMPOSITION OF WELL J-13 WATER⁷

<u>Species</u>	<u>Concentration (mg/l)</u>
Na	45.2
K	5.47
Ca	11.5
Mg	1.73
Al	0.026
SiO ₂	64.2
Sr	0.04
Ba	0.0023
Mn	0.0011
Fe	0.044
V	0.0425
Cl ⁻	6.4
SO ₄ ²⁻	18.1
F ⁻	2.1
NO ₃ ⁻	10.1
HPO ₄ ²⁻	0.10
HCO ₃ ⁻	143.

Whether this will happen in Yucca Mountain, however, is not clear. We have made a numerical study of this scenario using the WAFE code.⁹ Figure 38 shows the problem geometry.

First, a simplified analysis is made using a numerical solution for 1-D radial flow from a heat source (canister). Material properties for the calculation are given in Tables B-2 and B-3. We assume a uniform initial saturation of 87%, temperature of 23°C, and air pressure of 1.0 bar. We also assume a uniform concentration of SiO₂ in the pore water of 10⁻³ M. The heat source has a diameter of 63.6 cm and is centered at radius = 0. A reflecting boundary is imposed at radius = 16 m, the assumed midpoint between waste canisters. In addition to SiO₂, a relatively simple ion, Cl⁻, has been included. It will not adsorb and is not, to any significant extent, present in the tuff minerals. Its source is atmospheric fallout at the surface.

We are making a number of assumptions here, and the assumption of no interactions between the various water components is perhaps the most important. Solubility S is assumed to vary according to

$$\ln \frac{S}{S_0} = \frac{A}{T_0} \left(1 - \frac{T_0}{T} \right) \text{ where } T_0 = 23^\circ\text{C} (296^\circ\text{K}) \text{ and } A = \frac{\ln 10}{\left(\frac{1}{296} - \frac{1}{423} \right)}, \text{ and}$$

where S₀ is the solubility at the reference temperature T₀. Figures 39-44 show the calculated transient pressure, temperature, and saturation and concentration distributions. The total SiO₂ (dissolved and precipitated) is also shown.

Pressure and temperature go through an initial buildup. Saturation decreases at the wall of the canister hole. Beyond this, a broad, slightly elevated saturation region develops. Water boils and the water vapor moves outward, primarily through fractures. A strong capillary suction develops on both sides of the elevated saturation region, drawing water into the hot, low-saturation region as well as outward. After several decades, the pressure and temperature fields decay and the saturation returns to ambient. SiO₂ dissolves in a region near the canister and moves outward due to capillary suction where

it precipitates as it saturates. There is some return of SiO_2 to the depleted region, but a net redistribution of SiO_2 has occurred.

TABLE B-2
MATERIAL PROPERTIES

<u>Property</u>	<u>Value</u>
tuff permeability, sat. (cm^2)	1.925×10^{-14}
fracture permeability, sat. (cm^2)	1.013×10^{-7}
tuff porosity	0.11
fracture porosity	1.8×10^{-3}
tuff grain density (g/cm^3)	2.58
tuff specific heat ($\text{erg}/\text{g} \cdot ^\circ\text{C}$)	0.84×10^7
tuff thermal conductivity ($\text{erg}/\text{cm} \cdot ^\circ\text{C} \cdot \text{s}$)	$(1.75 + 0.60 \times \text{Sat.}) \times 10^5$
canister density (g/cm^3)	2.535
canister specific heat ($\text{erg}/\text{g} \cdot ^\circ\text{C}$)	0.325×10^7
canister heat production history ($\text{erg}/\text{s}/\text{cm}$) ⁹	$10^7 \times (31.7e^{-2.82 \times 10^{-5} t}$ $+ 48.6 e^{-1.29 \times 10^{-4} t}$ $+ 443.3e^{-1.76 \times 10^{-3} t}$ $+ 2123 e^{-0.02 t}$ $+ 431.5e^{-0.0635 t}$ $+ 321.9e^{-0.44 t})/505.,$ $t \text{ in years.}$

TABLE B-3
HYDROLOGIC PROPERTIES

<u>Property</u>	<u>Expression</u>
Water Rel. Permeability, K_{rw}	$\sqrt{S_e} [1. - (1. - S_e^{1/\lambda})^\lambda]^2$
Air Rel. Permeability, K_{rg}	$(1. - K_{rw})$
Matric Potential	$P_c = P_b [S_e^{-1/\lambda} - 1]^{(1-\lambda)}$ $P_b = 17.24 \text{ bars}$ $S_e = (S - S_o)/(1 - S_o),$ $S_o = 0.0801$ $\lambda = 0.4438$

Chlorine concentration increases initially in the hottest region because of decreasing water saturation due to boiling. Concentration in the elevated saturation region decreases, however, due to the addition of distilled water as the steam condenses. At late time, as the saturation returns to ambient, so too does the chlorine concentration profile.

Significant changes in the amount of SiO_2 dissolved or precipitated can lead to large permeability changes. If we assume that permeability is related to the pore diameters and that SiO_2 will precipitate onto pore walls uniformly, we can write an expression for permeability change as a function of porosity change. One possible relation is

$$\frac{K_2}{K_0} = (r_2/r_0)^{11} \left[(1 - \epsilon_0)/(1 - \epsilon_0(r_2/r_0)^3) \right]^2 \text{ (Kozeny-Carman)}$$

where K_0 is ambient saturated permeability, K_2 is permeability after precipitation or dissolution, r_0 is initial pore radius, and r_2 is pore radius after precipitation or dissolution. The value of r_2 is determined from $\Omega(\text{SiO}_2) = \epsilon_0 \frac{\Delta\Omega}{r_0^3} (r_0^3 - r_2^3)$, where $\Delta\Omega$ is a volume of rock, ϵ_0 is the ambient porosity, and $\Omega(\text{SiO}_2)$ is the volume of SiO_2 dissolved or precipitated in that volume of rock.

Figure 45 shows the permeability change we would expect based on our WAFE calculation by the end of the heat decay period. The SiO_2 precipitate distribution leads to an elevated permeability region in close to the canister with a reduced permeability region immediately outside the high permeability region. The elevated permeability is about an order of magnitude greater than ambient, while the reduced permeability is about 40 times smaller than ambient. The presence of these regions can have a significant effect on late time near-field hydrology. A reduced permeability shell around the canisters will tend to deflect downward-flowing water around instead of through the canister region. The exact quantitative magnitude of this effect depends, of course, on the details of the hydrologic behavior of Topopah Spring, in particular on the presence or absence of fractures. This effect of permeability reduction should occur wherever a capillary pressure gradient arises. In particular, it should occur at fracture faces, leading to sealing of fractures.

Our numerical simulation cannot treat the full complexity of the water composition and reactions. Instead, we greatly simplify by considering only SiO_2 dissolution, transport, and precipitation and neglecting chemical reactions. In addition, the permeability changes induced by silicate redistribution were not factored into the hydrologic flow. Finally, the rates of dissolution and precipitation were not varied with temperature. Other details of the benchmarking exercise referred to can be found in Ref. 10.

Colloids

Radionuclides released from waste canisters can travel in a variety of forms--as simple ions, as complexes either with other elements or as polymers (colloids), or embedded in or adsorbed on inert particles (pseudocolloids). Ionic transport has been studied much more than the other forms. In ionic form, most of the radionuclides of interest strongly adsorb to the tuffs of Yucca Mountain. In colloid form or attached to particulates, sorptive properties may be considerably different.

Colloids are a class of extremely small particles that were first studied early in the nineteenth century by Michael Faraday and other scientists.¹¹ They are characterized as particles containing several hundred to several thousand atoms, ions, or molecules and having diameters ranging from 10-1000 Å.

Matter in the colloidal state has a large surface area ($300 \text{ m}^2/\text{g}$); thus it is not surprising that the most important properties of colloids are those that are dependent on surface interactions such as adsorption. Drever¹² discusses the nature and geochemistry of colloids with emphasis on the charge surrounding colloids and its effect on suspension stability.

Olofsson et al.¹³⁻¹⁵ classify radiocolloids as true colloids or pseudocolloids, depending on their formation process. True colloids are formed by condensation of molecules or ions as a result of hydrolytic or precipitation processes. They consist mostly of hydroxides or polymers formed by hydrolysis. They have a very rapid formation rate and are favored at high pH values. Pseudocolloids, on the other hand, are formed as a result of adsorption on impurities in the solution and tend to be much larger than true colloids (up to $5000 \text{ \AA}$). Pseudocolloids can be of two types, reversible and irreversible. The formation rate of pseudocolloids is basically determined by the sorption rate on colloidal impurities.¹³

Radiocolloids are colloids containing radionuclides. They possess all the properties of normal colloids but in addition are radioactive. They may migrate in the ground matrix by virtue of groundwater and may also be captured by existing colloids. Radiocolloids are believed to be a significant vector for the transport of radionuclides from failed waste repositories and might actually accelerate this transport.^{16,17} Radiocolloids may arise from a variety of sources. Leaching of the waste form with groundwater may lead to the isotope itself producing colloids. Champ et al.¹⁸ have demonstrated experimentally the existence and rapid transport of plutonium colloids using core samples and groundwater.

Olofsson et al.^{13,15} showed the formation and rapid nonadsorbing migration of americium colloids over a wide range of pH. In their 1975 laboratory studies, Freid et al.¹⁹ observed a fast-migration form of plutonium in column experiments on Bandelier tuff. Retardation of this polymeric form of Pu was only 1/10 that of the ionic form.

Corrosion of canister material can lead to the formation of adsorbant colloids. Degradation of engineered backfills at the end of a repository's active life may lead to colloid formation. Also, naturally occurring colloids from clays (smectites, vermiculites, illites, kaolinite, and chlorite) present in groundwater may adsorb radionuclides.

Colloid transport depends on at least three factors: filtration, water velocity, and diffusion.

Filtration will be an effective barrier to transport in the tuff matrix. Pore size is an important element in filtration. Pseudocolloids will obviously not flow through pores that are smaller than the pseudocolloid carrier-particle size. In very small pores, true colloids will have a high probability of entering boundary layers around matrix particles where they can be captured. Pore size distributions have been measured²⁰ for several Yucca Mountain tuff samples. Mercury porosimetry data, measured by Quantachrome Co., on a sample of Topopah Spring tuff (Fig. 46) show that most of the pores are less than 1.0 μm . There is a small percentage larger than 1.0 μm , but they should be dry because the Topopah Spring saturation is less than 90%. More than 50% of the pore volume is made up of pores 0.1 μm in diameter or smaller. In samples from the Calico Hills (Fig. 47), 50% of the pores are smaller than 0.06 μm , but there are somewhat more of the largest pores (4 μm). These data indicate that in the tuff matrix, colloids must be on the order of 0.1 μm diameter or less to pass through.

Pore water velocities, assuming matrix flow, are estimated to be less than 1.0 cm/yr. Such slow flow is close to or less than molecular diffusion transfer rates. It is not clear whether such small velocities can provide sufficient momentum to allow bulk colloid movement. If fracture flow is significant, then colloid transport may be likely. Fracture apertures have been measured hydraulically and range between 50 and 250 μm . If fracture flow were dominant in the Topopah Spring tuff, average fracture flow rate would be roughly 2 m/yr, which is still close to molecular diffusion transfer rates. If fracture flow were episodic, corresponding to a few intense rainfalls per year, flow rates in fractures might be much larger (tens of meters per year) but would not last long.

In addition, colloids will be subject to gravitational settling¹⁷ for sizes greater than about 0.1 μm . The transporting ability of very small colloids will be diminished by diffusion of radiocolloids out of the carrier particles. Diffusivities in solid phase are small, but not zero. Solid-phase diffusivities for some radionuclides on several Yucca Mountain minerals have been measured²¹ at about 10^{-16} to 5×10^{-13} cm^2/s . A characteristic diffusion time from 0.1- μm -diameter particles is

$$D \approx R^2/T = \frac{L^2}{4T} \rightarrow T \approx \frac{L^2}{4D} \approx \frac{(10^{-5})^2}{4 \times 10^{-16}} = 2.5 \times 10^5 \approx 3 \text{ days}.$$

Small pseudocolloids, in fluids of low ionic concentrations, should not be able to retain adsorbed radionuclides for more than a few days. Very small true colloids may be able to migrate rapidly if fracture flow is occurring. Stability of true colloids depends on several factors. More experimental work is needed to determine the importance of true colloid transport. We mention one additional colloid-like mechanism that might allow accelerated transport of some radionuclides: microbes. This biological mechanism is outside the scope of this report, but the reader can pursue it through Ref. 22.

Conclusion

Two-dimensional simulations of transport in Yucca Mountain predict that travel time to the water table (here defined as first arrival of concentration level equal to 10^{-6} of source value) will be at least 15 000 yr, and much longer than this for most radionuclides.

Dissolution, transport, and precipitation of SiO_2 due to waste heat will result in a thin region of increased SiO_2 deposition around a canister, which will reduce permeability and provide a partial barrier against late-time water infiltration.

Small pore sizes, filtration, gravitational settling, and solid-phase diffusion will greatly diminish the transport capacity of colloids and pseudocolloids.

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Figure 1.

Cross section of Yucca Mountain.

Figure 2.

Assumed spacing of waste canisters in repository region. Region between dashed lines represents domain for calculations.

2-d yucca mountain, tc-99 5050.0 years

Figure 3.

Calculated concentration of ^{99}Tc in pore fluid (moles/l) at 5 000 years.

2-d yucca mountain, tc-99 10050.0 years

Figure 4.
Calculated concentration of ^{99}Tc in pore fluid (moles/l) at 10 000 years.

2-d yucca mountain, tc-99 15050.0 years

Figure 5.

Calculated concentration of ^{99}Tc in pore fluid (moles/l) at 15 000 years.

2-d yucca mountain, tc-99 20050.0 years

Figure 6.

Calculated concentration of ^{99}Tc in pore fluid (moles/l) at 20 000 years.

2-d yucca mountain, tc-99 10050.0 years

Figure 7.

Calculated concentration of ^{99}Tc absorbed onto rock matrix (moles/kg) at 10 000 years.

2-d yucca mountain, tc-99 20050.0 years

Figure 8.

Calculated concentration of ^{99}Tc absorbed onto rock matrix (moles/kg) at 20 000 years.

2-d yucca mountain, u-238 10150.0 years

Figure 9.

Calculated concentration of ^{238}U in pore fluid (moles/l) at 10 000 years.

2-d yucca mountain, u-238 50050.0 years

Figure 10.

Calculated concentration of ^{238}U in pore fluid (moles/l) at 50 000 years.

2-d yucca mountain, u-238 98175.0 years

Figure 11.
Calculated concentration of ^{238}U in pore fluid (moles/l) at 100 000 years.

2-d yucca mountain, u-238 10150.0 years

Figure 12.

Calculated concentration of ^{238}U absorbed onto rock matrix (moles/kg) at 10 000 years.

2-d yucca mountain, u-238 50050.0 years

Figure 13.

Calculated concentration of ^{238}U absorbed onto rock matrix (moles/kg) at 50 000 years.

2-d yucca mountain, u-238 98175.0 years

Figure 14.

Calculated concentration of ^{238}U absorbed onto rock matrix (moles/kg) at 100 000 years.

2-d yucca mountain, np-237 20250.0 years

Figure 15.

Calculated concentration of ^{237}Np in pore fluid (moles/l) at 20 000 years.

2-d yucca mountain, np-237 50250.0 years

Figure 16.

Calculated concentration of ^{237}Np in pore fluid (moles/l) at 50 000 years.

2-d yucca mountain, np-237 70250.0 years

Figure 17.

Calculated concentration of ^{237}Np in pore fluid (moles/l) at 70 000 years.

2-d yucca mountain, np-237 100250.0 years

Figure 18.

Calculated concentration of ^{237}Np in pore fluid (moles/l) at 100 000 years.

2-d yucca mountain, np-237 100250.0 years

Figure 19.

Calculated concentration of ^{237}Np absorbed onto rock matrix (moles/kg) at 100 000 years.

2-d yucca mountain, am-243 100500.0 years

Figure 20.

Calculated concentration of ^{243}Am in pore fluid (moles/l) at 100 000 years.

2-d yucca mountain, pu-239 102500.0 years

Figure 21.

Calculated concentration of ^{239}Pu in pore fluid (moles/l) at 100 000 years.

2-d yucca mountain, pu-239 102500.0 years

Figure 22.

Calculated concentration of ^{239}Pu absorbed onto rock matrix (moles/kg) at 100 000 years.

Figure 23.

Cross section of Yucca Mountain showing conceptual flow model. Dashed line encompasses domain of calculation.

1 yucca mountain, vertical and lateral flow, tc-99 5100.0 ;

Figure 24.

Calculated concentration of ^{99}Tc in pore water (moles/l) at 5 000 years.

I yucca mountain, vertical and lateral flow, tc-99 10100.0

Figure 25.
Calculated concentration of ^{99}Tc in pore water (moles/l) at 10 000 years.

1 yucca mountain, vertical and lateral flow, tc-99 15100.0

Figure 26.

Calculated concentration of ^{99}Tc in pore water (moles/l) at 15 000 years.

yucca mountain, vertical and lateral flow, tc-99 20100.0

Figure 27.

Calculated concentration of ^{99}Tc in pore water (moles/l) at 20 000 years.

1 yucca mountain, vertical and lateral flow, tc-99 5100.0 ,

Figure 28.

Calculated concentration of ^{99}Tc absorbed onto rock matrix (moles/kg) at 5 000 years.

I yucca mountain, vertical and lateral flow, tc-99 10100.0

Tracer 1 Solid Concentration (Y-level = 0.000)

Figure 29.

Calculated concentration of ^{99}Tc absorbed onto rock matrix (moles/kg) at 10 000 years.

I yucca mountain, vertical and lateral flow, tc-99 15100.0

Tracer 1 Solid Concentration (Y-level = 0.000)

Figure 30.

Calculated concentration of ^{99}Tc absorbed onto rock matrix (moles/kg) at 15 000 years.

yucca mountain, vertical and lateral flow, tc-99 20100.0

Tracer 1 Solid Concentration (Y-level = 0.000)

Figure 31.

Calculated concentration of ^{99}Tc absorbed onto rock matrix (moles/kg) at 20 000 years.

yucca mountain, vertical and lateral flow, u-238 10323.0

Figure 32.
Calculated concentration of ^{238}U in pore water (moles/l) at 10 000 years.

yucca mountain, vertical and lateral flow, u-238 50283.1

Figure 33.
Calculated concentration of ^{238}U in pore water (moles/l) at 50 000 years.

yucca mountain, vertical and lateral flow, u-238 100233.2

Tracer 1 Fluid Concentration (Y-level = 0.000)

Figure 34.

Calculated concentration of ^{238}U in pore water (moles/l) at 100 000 years.

yucca mountain, vertical and lateral flow, u-238 10323.0

Figure 35.
Calculated concentration of ^{238}U absorbed onto rock matrix (moles/kg) at 10 000 years.

yucca mountain, vertical and lateral flow, u-238 50283.1

Figure 36.

Calculated concentration of ^{238}U absorbed onto rock matrix (moles/kg) at 50 000 years.

yucca mountain, vertical and lateral flow, u-238 100233.2

Tracer 1 Solid Concentration (Y-level = 0.000)

Figure 37.

Calculated concentration of ^{238}U absorbed onto rock matrix (moles/kg) at 100 000 years.

Figure 38.

Hypothetical waste canister emplacement geometry. The qualitative effect of waste heat on near-field water saturation is shown.

Figure 39.
Calculated pressure
distribution at
selected times.

10.0 years

Figure 40.
Calculated temperature distribution
at selected times.

100.1 years

Figure 41.
Calculated saturation distribution
at selected times.

Figure 42.
Calculated chlorine distribution
in pore water
at selected times.

Figure 43.
Calculated SiO_2
distribution in
pore water at
selected times.

Figure 44.
Calculated total
SiO₂ (dissolved
and precipitated)
at selected times.

Figure 45.

Calculated permeability assuming relationship between pore volume and permeability.

TOPOPAH SPRING

Figure 46.

Distribution of pore sizes by % volume for a sample of Topopah Spring tuff.

CALICO HILLS

Figure 47.

Distribution of pore sizes by % volume for a sample of Calico Hills tuff.

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