DIFFUSION OF SORBING AND NON-SORBING RADIONUCLIDES

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Ines R. Triay, Kay H. Birdsell, Alan J. Mitchell, Martin A. Ott

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DIFFUSION OF SORBING AND NON-SORBING RADIONUCLIDES

Los Alamos National Laboratory
Los Alamos, New Mexico 87545
(505) 665-1755

ABSTRACT

Diffusion is considered one of the most important retardation mechanisms in fractured media. The diffusion experiments conducted involved solid tuff and groundwater from Yucca Mountain. The uptake of radionuclides by the tuff was studied utilizing containers made of tuff in the form of beakers. The solution containing the radionuclides of interest was placed in the tuff beaker cavity and the uptake of the radionuclides by the tuff was measured as a function of time. Our results indicate that the diffusion coefficient for nonsorbing radionuclides into saturated Yucca Mountain tuff is on the order of $10^4$ cm$^2$/s. Large anions, such as pertechnetate are excluded from tuff pores and their diffusion coefficients are on the order of $10^2$ cm$^2$/s. Comparison of the predictions for the uptake of sorbing radionuclides by the tuff with the actual data obtained indicates that conservative transport calculations will result from predicting diffusion using the batch sorption coefficient for the sorbing radionuclide and the diffusion coefficient obtained for tritiated water.

EXPERIMENTAL

The experimental technique involves fabricating containers in the form of a beaker (made of tuff). The rock beaker sits on a Plexiglas container (surrounded by a layer of groundwater on the sides). A stopper is utilized to prevent evaporation. The beaker has a cavity with a radius of approximately 1.4 cm and length of 2.5 cm. The rock beaker has a length of approximately 5 cm and a radius of 3.1 cm. Figure 1 shows a cross section of a rock beaker encapsulated in a Plexiglas container.

The radionuclides utilized for these experiments were H-3, Tc-99m, Np-237, Am-241, Sr-85, Cs-137, and Ba-133. A solution (prepared with groundwater from the well J-13) containing the radionuclides of interest was placed in the cavity of the rock beakers. Aliquots of the solution containing the radionuclides of interest were taken as a function of time and analyzed for radionuclide concentration.

Batch sorption experiments were performed utilizing the tuffs being studied. The batch sorption
procedure consisted of pretreating 1 g of tuff with 20 ml of a J-13 solution for two weeks, separating the phases, adding 20 ml of the J-13 solution containing the radionuclides of interest to the pretreated tuff, separating the phases by centrifugation, and determining the amount of radionuclide in both phases.

RESULTS AND DISCUSSION

Figure 2 shows the concentration left in the solution in the cavity of the rock beaker (made with tuff G4-737) divided by the initial concentration of the solution placed in the rock beaker as a function of time elapsed after the J-13 solution was placed in the cavity. The diffusion data were fitted to the diffusion equation, given by Equation (1), utilizing the transport code TRACRN. Figure 3 shows the fits obtained (solid lines) utilizing TRACRN for nonsorbing radionuclides (H-3 and Tc-95m) in a beaker made of tuff G4-737. The diffusion coefficients obtained for the nonsorbing radionuclides in all the tuffs studied are given in Table 1. Large anions such as pertechnetate are excluded from tuff pores because of their size and charge. The diffusion coefficients obtained for nonsorbing radionuclides agree well with previous results.

The relationship between F and C describes the sorption mechanism that is observed for the sorbing radionuclides. If sorption is linear, reversible, and instantaneous, then F/C is equal to a sorption coefficient (Kd). In order to test this assumption, batch sorption experiments were performed utilizing the tuffs under study. The sorption coefficients (Kd) determined by batch sorption techniques are given in Table 2.

The diffusion of the sorbing radionuclides could not be fitted assuming reversible, instantaneous, and linear sorption. Figure 4 shows the fit that would be obtained assuming the diffusion coefficient determined for tritiated water in tuff G4-737 and the sorption coefficient determined for each sorbing radionuclide in tuff G4-737. Comparison of the fits for the sorbing radionuclides with the actual data obtained (see example in Figure 5) indicates that conservative transport calculations will result from a batch sorption Kd and the diffusion coefficient obtained for tritiated water.

The results obtained from rock beaker experiments agree with previous results. Tuff wafer experiments were utilized to study the uptake of sorbing radionuclides by tuff. The rate constants for uptake of the sorbing cations on tuff were consistent with a model that is diffusion limited (where diffusion occurs in two stages). First, the cations diffuse into rock through water-filled pores and then the cations diffuse into narrower intracrystalline channels. This dif-
Figure 1: Sample Rock Beaker

Figure 2: Diffusion Data for Rock Beaker G4-737.
Figure 3: Fits (solid lines) to Diffusion Data for Rock Beaker G4-737.

Table 1: Diffusion Results.

<table>
<thead>
<tr>
<th>Tuff Sample</th>
<th>Major Minerals, %</th>
<th>Porosity</th>
<th>D (cm²/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>HTO</td>
</tr>
<tr>
<td>G4-737</td>
<td>alkali feldspar, 68 cristobalite, 28</td>
<td>0.07</td>
<td>2.2 x 10⁻⁶</td>
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<tr>
<td>GU3-304 #1</td>
<td>alkali feldspar, 75 cristobalite, 25</td>
<td>0.06</td>
<td>1.5 x 10⁻⁶</td>
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<tr>
<td>GU3-304 #2</td>
<td>alkali feldspar, 76 cristobalite, 15</td>
<td>0.10</td>
<td>1.6 x 10⁻⁶</td>
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<tr>
<td>GU3-433</td>
<td>alkali feldspar, 70 cristobalite, 15</td>
<td>0.10</td>
<td>3.5 x 10⁻⁶</td>
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<tr>
<td>GU3-1119</td>
<td>alkali feldspar, 70 quartz, 19</td>
<td>0.10</td>
<td>2.0 x 10⁻⁶</td>
</tr>
<tr>
<td>Topopah</td>
<td>alkali feldspar, 59 cristobalite, 23 quartz, 12</td>
<td>0.07</td>
<td>1.0 x 10⁻⁶</td>
</tr>
</tbody>
</table>
Table 2: Sorption Results.

<table>
<thead>
<tr>
<th>Tuff Sample</th>
<th>Major Mineral, %</th>
<th>( K_d ) (ml/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Np</td>
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<td>G4-737</td>
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<td>GU3-433</td>
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<tr>
<td>GU3-1119</td>
<td>alkali feldspar, 70 quartz, 19</td>
<td>8</td>
</tr>
<tr>
<td>Topopah Outcrop</td>
<td>alkali feldspar, 59 cristobalite, 23 quartz, 12</td>
<td>9</td>
</tr>
</tbody>
</table>

Figure 4: Calculated Diffusion Data for Rock Beaker G4-737.
Figure 5: Comparison of Fit Obtained (solid line) with Actual Cs Data (solid squares) for Rock Beaker G4-737.

ACKNOWLEDGEMENTS

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The data utilized to compile this document is referenced in Notebook TWS-INC11-9/88-6.

The code TRACRN used for data analysis has not been fully qualified per the Yucca Mountain Site Characterization Project Software Quality Assurance Plan.

REFERENCES


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