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A CRITICAL LOOK AT STUDIES OF RADIONUCLIDE MIGRATION IN FRACTURED GRANITE CORES

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AND
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TO BE PRESENTED AT THE NEA WORKSHOP
ON EXPERIMENTAL METHODOLOGIES IN RADIONUCLIDE SORPTION
AT OECD HEADQUARTERS IN PARIS, JUNE 6-7, 1983

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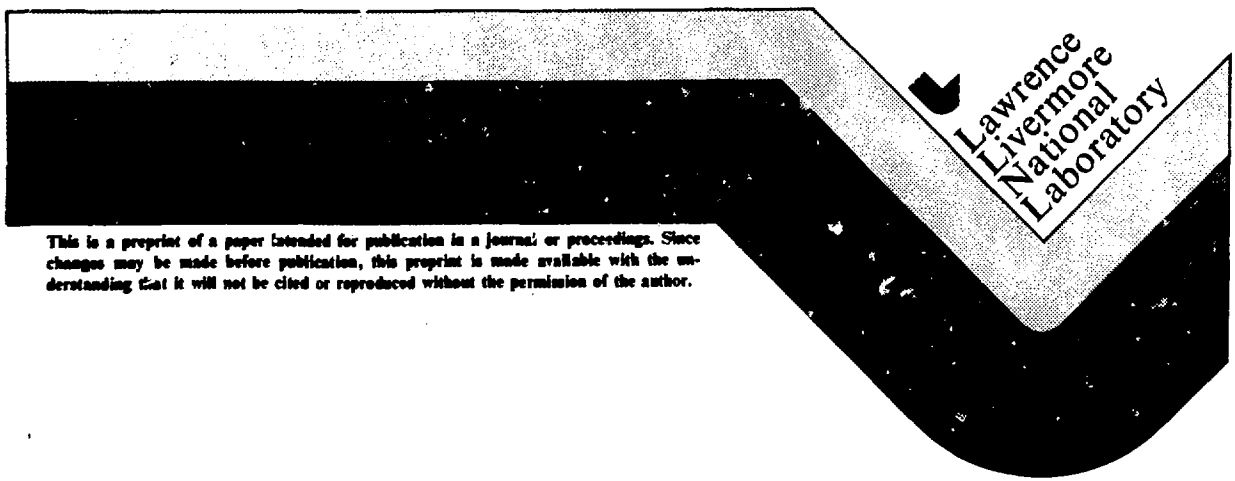

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DISCLAIMER

Introduction

A series of laboratory experiments studying radionuclide migration were conducted on eleven fractured granite cores from the Climax Stock at the Nevada Test Site. Failor et al. (1982) discuss the equipment used, the preparation of the core, the experimental procedure, the data reduction, and the experimental results. They give estimates of the average fracture apertures, retardation values of ^{85}Sr, ^{95m}Tc, and ^{137}Cs relative to ^{3}H, and the percentage of each radionuclide retained in the core after each run. To determine the effect of fracture fill material and solution composition on radionuclide migration, they studied both natural and artificial fractures using either natural Climax ground water or distilled water. The results are summarized below along with a discussion of the problems inherent in the experiments and suggestions to minimize these problems.

Core sorption studies are needed to determine the scaling factor from the laboratory to the field and to test whether laboratory studies can accurately reflect in situ conditions. If so, laboratory studies with cores can be used to supplement data from the field studies which, because of time and cost disadvantages, will be limited. It is our belief that the use of static sorption tests or column experiments using crushed rock are not acceptable substitutes for core experiments when studying migration in fractured media. Improved core studies represent a better use of laboratory time and resources.

Experimental Results

The following results are summarized from Failor et al. (1982). The eleven cores were all 2.5 x 2.5 cm granite cylinders each containing a fracture parallel to the core axis. A confining pressure of 9 to 15 MPa was applied to simulate lithostatic pressure. Flow rates were held relatively constant ($\pm$ 10%) during each experiment and ranged from .002 to .04 ml/min. This resulted in residence times for an unretarded radionuclide of 1.3 to 38 minutes. After a run, the core was sliced normal to its axis and the slices γ-counted to determine the amount of radioactivity remaining in the core.

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For one core containing a natural fracture, autoradiography of the slices identified the location of the radioactivity. In all cases, it was found only in the fracture except on the core's end pieces where the solution came in direct contact with the granite.

The elution curves for ^{85}Sr showed little or no retardation (R) relative to tritiated water (R = 1.0 $\pm$ 0.2 to 2.5 $\pm$ 1.2) irrespective of the type of fracture or transport solution. The transport solution did affect the amount of ^{85}Sr retained in the core. With natural ground water, less than 2.5% of the ^{85}Sr was retained. With distilled water, 7.7 to 37% of the ^{85}Sr was retained. A nonlinear isotherm combined with a competition for sorption sites between calcium and strontium may be the cause.

The elution curves for ^{95m}Tc also showed little or no retardation relative to tritiated water (R = 1.0 $\pm$ 0.2 to 1.8 $\pm$ 0.4). The amount of ^{95m}Tc retained in the cores (0.1 to 46%) did not correlate with type of transport solution or fracture.

Cesium-137 showed a varied retardation from core to core. The amount of retardation (R = 1 $\pm$ 10 to 120 $\pm$ 120) did not correlate with type of transport solution or fracture in any clear way. With natural ground water, less than 73% of the ^{137}Cs was retained in the core. With distilled water, 88 to 99% of the injected ^{137}Cs was retained in the core. With the natural fractures containing calcite 68 to 72% of the injected ^{137}Cs was retained in the core. With the artificial fractures and a natural fracture containing no calcite, 31 to 37% of the ^{137}Cs was retained, yet the autoradiographs showed less radioactivity in the regions where calcite was present.

A Critical Look

The large uncertainties in the retardation values result from the ratio of the apparatus dead volume to the fracture volume. The core sorption apparatus was designed to apply confining pressures up to 35 MPa. To do this, end pieces are attached to the core which is then placed in a large pressure vessel. The volume of these end pieces combined with the volume of the lines to the spike loop, the transducers and fraction collector add up to a dead volume which is approximately two orders of magnitude larger than the fracture volume. The retardation value is the ratio of the elution volumes for the radionuclide and tritium. The elution volumes are determined by subtracting the dead volume from the accumulated volume that passes through the core from

the time of injection to the time of peak activity for a particular radionuclide. Even small uncertainties in the dead volume will thus result in large uncertainties in the elution volume.

Reducing the dead volume by redesigning the apparatus will help minimize this source of uncertainty; however, it is unlikely given the small core size presently used that sufficient improvement can be accomplished in this way. That is, unless the core size (hence the fracture volume) is greatly increased. Problems created by this latter approach depend on the site where the samples are taken. At sites where the fractures are strongly cemented, it may be possible to drill out large cores with intact fractures running parallel to the axis. However, at sites such as Climax where the fractures are poorly cemented and easily broken open, drilling will partially destroy the fracture orientation and loss of the fracture fill material is likely.

A second source of uncertainty in the retardation values lies in the data points. The "noise" in the analytical measurements makes a precise determination of the peak location difficult. For radionuclides with retardation values only slightly larger than one (e.g., 2 to 10), the uncertainties in the peaks overlap the uncertainties in the tritium peak.

A third source of uncertainty occurs with radionuclides that have significantly larger retardation values. These peaks occur later in the run and are usually smeared out. They appear more as a "hump" on the curve rather than a well defined peak. We can only estimate the peak location somewhere in the middle of the "hump".

A fourth problem with interpreting the data is the large amount of tailing found in these curves. It implies that the residence time of the radionuclides within the fracture is insufficient for equilibrium to be reached. If sorption is incomplete, this will lead to a premature elution of the radionuclide followed by continued desorbing of the remaining radionuclide. This could explain the apparent lack of retardation with ^{85}Sr.

One more factor to be considered is the volume of the spike injected. In the experiments described here, the injection volume was 0.5 ml which is large compared to the fracture volume (0.01 to 0.04 ml). Such a relatively large injection cannot be considered as an instantaneous or continuous injection. It partially explains the asymmetry of the elution curves and could be responsible for overloading of the sorption sites.

In this critique, we wish to acknowledge, but will not discuss, (1) the role of diffusion into the rock matrix--the small residence time minimizes its importance; (2) the need for simulating the overburden pressure--it would greatly simplify the experimental procedure if it were eliminated; (3) the role kinetics (the absolute and relative rate of sorption and desorption reactions) plays in the shape of the elution curves; (4) or the role of heterogeneity in geologic samples which influences reproducibility.

The experiments described in Failor et al. (1982) are admittedly incomplete; however, the information they provide emphasizes the importance of solution composition and fracture fill material. We have preferred to emphasize problems encountered in our experiments as a basis for discussions with others who may have similar problems.

REFERENCE

Failor, R., D. Isherwood, E. Raber, and T. Vandergraaf, 1982, Laboratory Studies of Radionuclide Transport in Fractured Climax Granite, Lawrence Livermore National Laboratory, UCRL-53308.

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