CONF-950216--40

LA-UR-95- 753

Title: TRACER STUDIES OF RADIONUCLIDE MIGRATION IN FRACTURED ROCK AT A SCALE OF 1 METER

Author(s): ROBERT S. RUNDBERG

BRIAN J. TRAVIS

Submitted to: WASTE MANAGEMENT '95
TUCSON, AZ
2/26/95 - 3/02/95

Los Alamos
NATIONAL LABORATORY

DISCLAIMER

DISCLAIMER

TRACER STUDIES OF RADIONUCLIDE MIGRATION IN FRACTURED ROCK AT A SCALE OF 1 METER

R.S. Rundberg, B. Travis,Los Alamos National Laboratory, Los Alamos, NM USA

T.T. Vandergraaf and D.J. Drew, Whiteshell Research, Pinawa, Manitoba, Canada

ABSTRACT

The Large Block Tracer Test is a series of experiments to study the migration of radionuclides through fractures in rock at the scale of one meter. The separate effects to be considered are sorption onto minerals within the rock matrix; diffusion of radionuclide species through the rock matrix, diffusion and hydrodynamic dispersion within the fracture; and the effect of heterogeneity in the fluid flow field (also known as macro dispersion or channeling). The rock fractures to be used have natural fractures or artificial fractures with engineered heterogeneity. These tracer experiments will provide data with well-defined geometry and conditions for use in code validation. The experiments also provide an experimental framework to test inverse methods. Results are presented for a series of migration experiments using conservative tracers in artificial fractures with near parallel plane and near wedge-shaped fractures. The results are compared with those predicted with transport code TRACR3D[1]. The fracture is treated as an equivalent porous medium with a "cubic law" permeability and a porosity that is proportional to the aperture. The results show good agreement, both between experimental results and those predicted by TRACR3D, but also between the distribution of the dye tracer in the fracture and the elution profiles. This suggests that the transport of a tracer through a fracture can be inferred from elution profiles.

INTRODUCTION

The modeling and prediction of radionuclide migration from an underground nuclear waste repository is a complex problem involving many physical and chemical processes. The coupling of these processes depend on the spatial distribution of hydrologic and chemical properties of rock. The exact details of these distributions will probably not be known. Approximations and some form of stochastic modeling will be used. The Large Block Tracer test will allow the testing of sorption/retardation models in complex water flow systems. The experiments also provide an experimental framework to test inverse methods.

The primary purpose of Large Block Tracer Test is to study the effects of diffusion and adsorption on the migration of radionuclides through fractures in rock at the scale of one meter. The one meter scale is an intermediate between smaller laboratory scale experiments and the field scale. One meter scale fractures are experimentally more readily characterized because the fluid volumes are on the order of 100 milliliters rather than 100 microliters. The greater fracture volumes allows the fluid velocity to be controlled over a broader dynamic range with standard pumps. The first set of experiments (those described in this work) involve artificial fractures with simplified geometry. This was done to reduce the uncertainty in the fluid flow field (i.e., macrodispersion) so that the effects of diffusion and adsorption could be isolated.

Three different engineered fractures are being considered to provide data for validation of transport models and to test inversion algorithms for inverse problems:

1. a fracture with a regular, rectangular cross section to study the effects of matrix diffusion and sorption without a need to consider channeling dispersion,
2. a fracture with grooves, 1 cm apart, parallel to the flow along the fracture, to study asymptotic Fickian dispersion at low transport solution velocities, and
3. a wedge shaped fracture with the wedge dipping across the flow field. This fracture geometry has a covariance function for the permeability field that resembles the geostatistical model used by Gelhar[2] to describe stratified aquifers. The wedged fracture approximates an aperture distribution where the correlation length is greater than or equal to the scale of the experiment. This situation has channeled flow and a large macrodispersivity. The results obtained with this type of fracture can therefore be used to model contaminant transport in natural fractures under field conditions.

The fractures described have been constructed in granite blocks and begun to provide experimental data. Tuff blocks have been excavated in the vicinity of Yucca Mountain and are being prepared for experimentation.

The first phase of experimentation, now completed, is intended to study only the fluid flow in the fracture. The fluid movement is studied visually using fluorescent dye. This phase tests our ability to construct idealized fracture geometries to an acceptable tolerance. Techniques were developed to determine deviations from design and compensate for the effects of those deviations.

EXPERIMENTAL

The Large Block Radionuclide Migration Facility (Drew et al. 1993)[3] is equipped to handle blocks of rock with a maximum dimension of I m and is designed as an IAEA Type B laboratory. Migration experiments and post-experiment radiochemical analyses, including two-dimensional gamma scanning, alpha scanning, and autoradiography of the fracture surface can be performed in this facility.

Slabs of pink granite, with dimensions of 48 x 48 x 4 inches were obtained from a commercial quarry on the Cold Spring quarry on the south eastern Rank of the Lac du Bonnet batholith near Lac du Bonnet Manitoba, Canada for use in the radionuclide migration experiments. To develop the techniques required to establish uniform flow through a fracture with a regular cross section, artificial fractures have been assembled from a 4 inch thick, 24 x 24 inch slab of polished pink granite and a 4 inch thick, 24 x 24 inch acrylic sheet, separated by 1/16 inch-thick strips of stainless steel. The surface of the granite slab was polished at the quarry with a mixture of iron oxides at the quarry and spray painted white to increase the visibility of colored dyes in the fracture. Deviations from flatness were measured both for the acrylic sheet and granite slab. The deviations were determined using a standard straight edge and feeler gauges.

To create uniform flow across the width of the fracture with the regular aperture, acrylic inlet and outlet plena were attached to two sides of the fracture; the other two sides were sealed with a silicone sealant (Canadian General Electric RTV-108). Porous polyethylene membranes with uniform pore size of 40 micrometers were inserted between the plena and the fracture to decrease the transmissivity between the inlet plenum and the fracture which would lead to an uneven introduction of the contaminant into the fracture. The outlet plenum was divided into eight equal sections to enable flow from each different areas of the fracture to be collected into individual fractions. The parallel plane fracture was used to calibrate the flow through the fracture. Since the transmissivity through a fracture with apertures on the order of 1/16 inch is very high. slight differences in transmissivity in the fracture result in large changes in flow through the fracture. To eliminate this problem, the individual chambers of the outlet plenum were equipped with lengths of very fine bore tubing with an outside diameter of 1/16 inch. By trimming the ends of this tubing, uniform flow could be achieved across the fracture.

Distilled deionized water was used as the transport solution and was pumped through the fracture at a flow rate of 30 ml/hr, giving a residence time in the fracture with the rectangular cross section of approximately 20 hours. A 2×10^{-4} mole/L uranine concentration was used as the conservative dye. These migration experiments were performed over periods of 48 hours. Images of the dye are obtained with a SONY Model AVC-P7 CCD video camera connected to a Data. Translation Model DT 3851 frame grabber installed in a 80486 personal computer. The camera is triggered by an external trigger using a DAS-16 I/O board installed in a portable 80386 personal computer. The images are stored as .IMG files on the hard drive of the 80486 computer and enhanced with Data Translation Global Image software to produce TIF files. Five milliliter samples of effluent from each of the 8 outlets were taken by automatic fraction collectors.

RESULTS

The results described were obtained from the first set of a series of migration experiments in artificial fractures with a regular, well defined aperture width.

1. TRACR3D calculations

Forward calculations of the transport of both sorbing and nonsorbing tracers have been made using the code TRACR3D. These calculations have been made to examine the effects of fracture geometry, adsorption, and sorption kinetics. The calculations have also been used to assist the design of the blocks. The current design has a porous material on the inlet and outlet of the fracture to make the flow field with the fracture less sensitive to small changes in pressure at the sampling locations. The code was used to predict the movement of the nonsorbing uranine dye. The deviation from flatness was incorporated into the fracture permeability distribution.

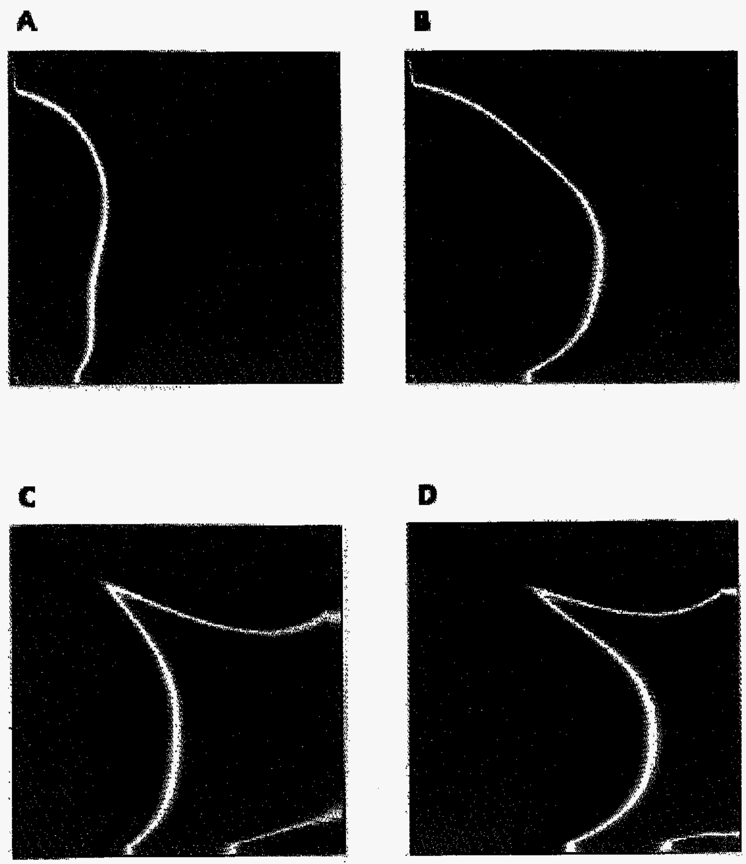

Figure 1 TRACR3D prediction of tracer profile in wedged fracture after 3(A), 6(B), 11(C), and 24(D) hours.

The uranine profiles for the parallel plate fracture and for the wedged fracture (shown in fig. 1) were calculated. The model had no sorption and no matrix diffusion. This was appropriate because the granite face had been coated with epoxy paint The agreement between the calculated profiies and observed fronts is good. Only minor artifacts are evident near the boundaries of the fracture.
The wedge fracture was modeled as an equivalent porous medium whose penneability is a function of the aperture ($k = w^2/12$). The porosity of each node was proportional to the average fracture aperture (porosity = local width /

maximum width). The permeability at each node is the average of the cubic law permeability for the fracture within the boundary of the node.

The target average flow velocity is 10^{-3} cm/s. The pressure gradient needed to provide this velocity is found by integrating the velocity across the breadth of the fracture of the fracture, i.e.,

$$\bar{u} = \frac{1}{L_x}\int_0^{L_x} u(x)dx = \frac{-\Delta p}{\mu L_x L_z}\int_0^{L_x} k(x)dx = \frac{-\Delta p}{\mu L_x L_z}\int_0^{L_x} \frac{\left[\frac{x w_o}{L_x}\right]^2}{12} dx = \frac{-w_o^2 \Delta p}{36 \mu L_z}$$

where u is the average velocity, L_x, is fracture width, L_z is the fracture length, w_o is the maximum aperture, Δp is pressure difference across the fracture, and μ is the viscosity. From this relationship the pressure gradient needed is 0.04 millibars. This calculation shows why it is experimentally difficult to acheive a uniform flow field given small variations in permeability across the components of the inlet and outlet of the block experiments.

The concentration of uranine in each of the 8 outlet ports in the wedged fracture was calculated using the same aperture distribution. These results are shown in figure 2. The earliest breakthrough is predicted in ports 3 and 4 in agreement with the concentration profiles.

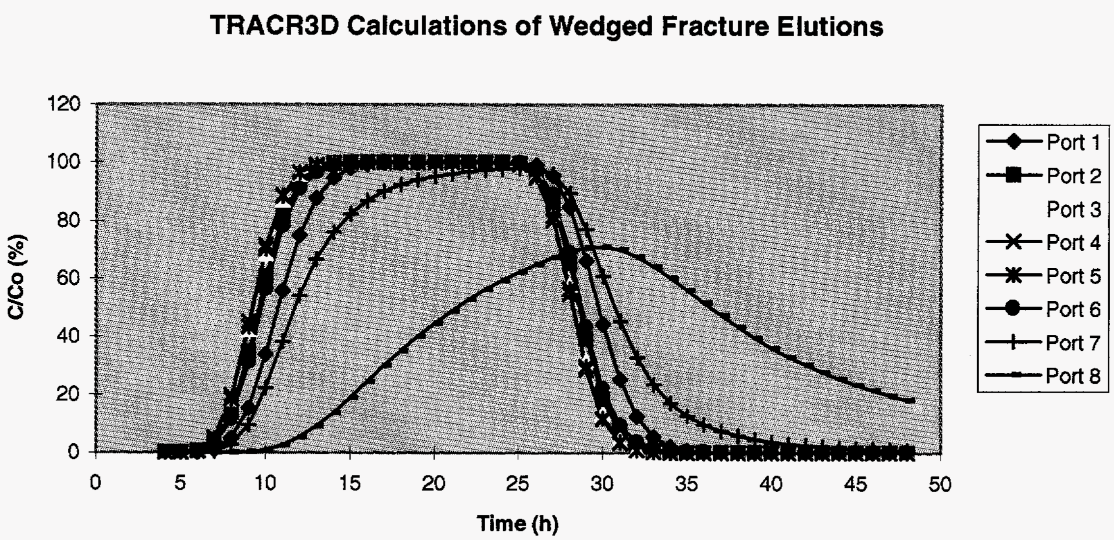

Figure 2. TRACR3D simulations of the effluent concentration at the outlet ports.

Calculations have been made for the grooved fracture. The calculations were made for nonsorbing tracers, sorbing tracers, and high molecular weight nonsorbing tracers. The results of these calculations showed that matrix diffusion tended to reduce the effect of channeling on the tracer concentration in the fracture fluid. Sorption retarded the migration of tracer. The most significant effect of the grooved fracture we seen for the high molecular weight tracer. The effect of channeling appeared to be enhanced for high molecular weight (low diffusivity) tracers.

2. Uranine profiles

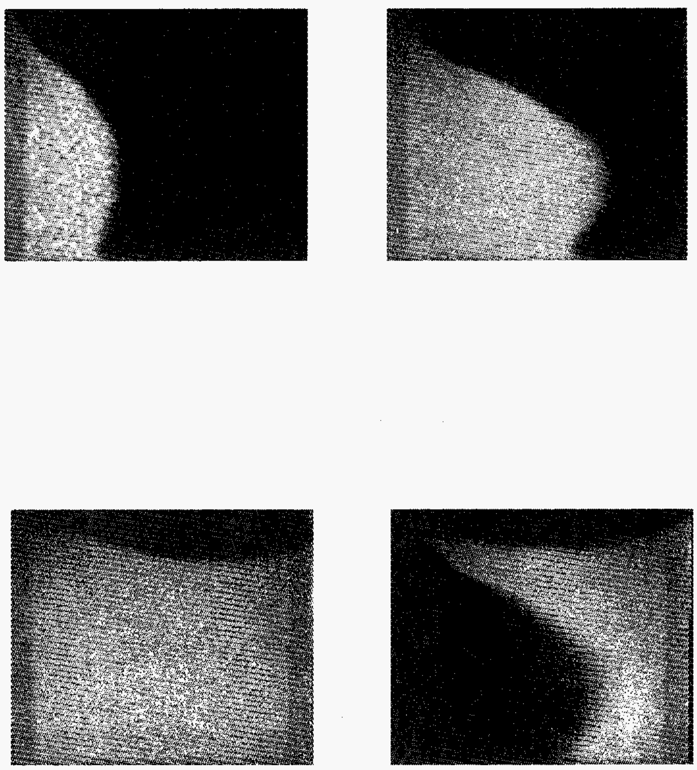

Figure 3. Image of actual profile of uranine tracer in wedged fracture after 3, 6, 24, and 30 hours.

Two different fracture aperture configurations have been used: a near parallel plane fracture and a wedge shaped fracture with a variable aperture. Inlet and outlet plena are fitted to the ends of the fracture and the transport solution is injected into and withdrawn from the complete width of the fracture. The outlet plenum is subdivided into eight compartments with the eight outlets leading to individual fraction collectors. Although uniform or near uniform flow could be established by trimming the fine bore tubing leading from the chambers in the outlet plenum to the fraction collectors, the shape of the front of the injected uranine indicated that the flow through the center of the fracture was slightly higher than at the edges. This is due to a slight concavity of the polished granite slab and the acrylic sheet. Since the linear flow velocity through the fracture varies with the square of the fracture aperture, a slight deviation from a uniform cross section is sufficient to create differences in the linear velocity through the fracture. In the case of the rectangular fracture, the aperture al the edges was 1/16 inch. six hours after the start of the uranine injection, the leading edge of the dye had moved approximately 6 inches along the edges of the fracture, while the center of the front had moved 12 inches. This corresponds to a deviation of 1/38 inch from a perfectly flat surface. After 24 hours the dye injection was discontinued and the fracture flushed with water. The spacer was removed from one side of the block and the spacer on the other side was replaced by a 3/16 inch spacer. This kept the same average permeability for the fracture. The dye injection was discontinued after 24 hours and the fracture flushed with water. The resulting profiles are shown in figure 2 for 3, 6, 24, and 30 hours. respectively. The wedged fracture geometry dominated the flow pattern but the effect of the cupped granite surface was still evident.

The effluent from each of the eight outlet plena was sampled using fraction collectors. The sample concentrations were analysed using spectrophotometry. The concentrations in the eluted fractions are shown in Fig. 4. The effect of concavity of the fracture is again evident with the earliest breakthrough occuring in outlet ports 3 and 4. The breakthroughs were delayed (as expected) in the narrower part of the fracture. The dispersion (as evidenced by the lower slope of the leading edge) increased as the fracture became narrower. This is due to the greater deviation (across the portion of the fracture sampled) from the average flow in the portion of the fracture sampled.

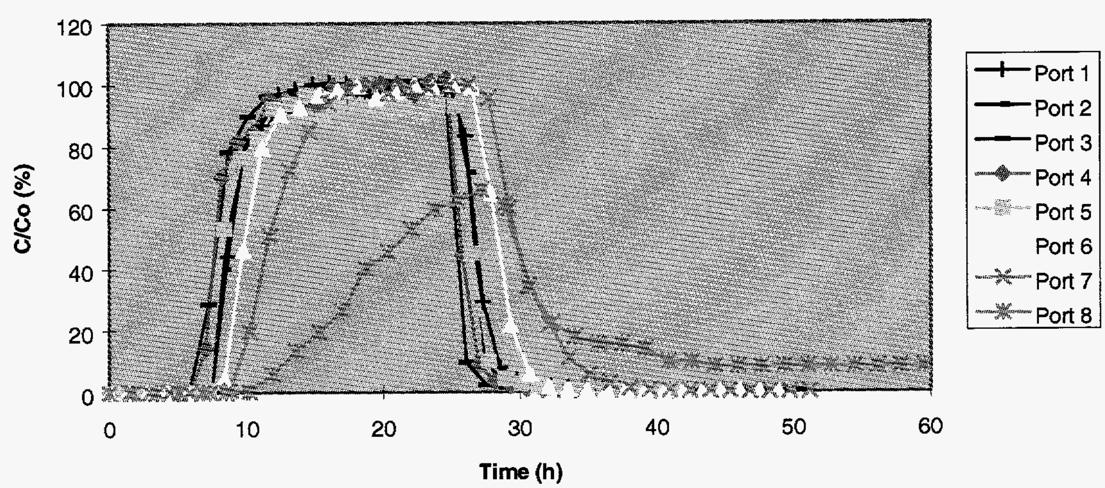

Figure 2 Concentration in outlet plenum as a function of time and position across fracture.

The results show good agreement, both between experimental results and those predicted by TRACR3D, but also between the distribution of the dye tracer in the fracture and the elution profiles. This suggests that the transport of a tracer through a fracture can be inferred from elution profiles.

An algorithm for inverting two dimensional integral equations has been developed. The method applied is that of Tikhonov[4] regularization. This code will be used to sharpen the autoradiographic images of tracers sorbed in the rock matrix. The code has been tested using regular images What have been distorted by a gaussian filter. The recovered images are greatly sharpened and in good agreement with the actual image. This algorithm has also been incorporated into the TRACRI[5] code. TRACRI will be used to infer permeability fields from tracer experiments.

Transport of a nonsorbing tracer in the wedged fracture has been calculated with TRACR3D and with one dimensional approximations. The stochastic model of Gelhar agrees very well with the TRACR3D calculation. The differences are small for case of nonsorbing tracers. NonFickian dispersion appears to cause the dispersivity inferred from nonsorbing tracers to be too small to adequately predict the dispersion of sorbing tracers[6].

CONCLUSION

Construction of I meter scale fractured blocks have been completed. Calculations have been completed that predict the effects of matrix diffusion and sorption on the transport of radionuclides through the fractures. Experiments are starting to provide data for validation of the TRACR3D code.

The experiments have shown shown that: 1. It is experimentally difficult to engineer an ideal fracture geometry but deviations can be accounted for in the modeling. 2. Simple fractures, i.e., a wedged fracture provides a realistic model of macrodispersion. Because the covariance function is similar to some geostatistical models. This is validated by the excellent agreement between calculated and measured concentration profiles. 3. The fluid dynamics in these simple fracture geometries can be predicted with sufficient accuracy that the effects of adsorption and diffusion can be studied unambiguously.

We are most interested in determining under what conditions can the spatial distribution of fracture aperture and connectedness (i.e., the permeability) be found from flux and transport measurements. This is an inverse problem, and in general, such problems cannot be solved uniquely to any arbitrary level of resolution. However, it is possible to resolve the unknown structure down to some spatial scale with an uncertainty in resolution inversely related to the scale. given sufficient observations of the system's response. In our present experimental design, with fluxes measured at 8 outlet ports, the flowpaths through the wedge fracture can only be coarsely estimated. This uniqueness difficulty can be alleviated by adding constraints. In this case, we have the curves of concentration vs. time at each outlet port. This information can be used to reduce the magnitude of uncertainty in the fracture geometry. If we add one further constraint, that the geometry has a self-affine structure, for example, further improvement in resolution is possible. Through a sequence of simulations with our inversion model TRACRI, we show how addition of each of these data sets and constraints improves the resolution of the fracture geometry.

The results of these preliminary studies look promising for the development of a better understanding of the consequences of channeling dispersion to radionuclide migration through rock.

ACKNOWLEDGEMENTS

This work was partially support by the Yucca Mountain Project Office of the U. S. Department of Energy Office of Civilian Radioctive Waste Management. The authors thank Dr. Robert Levich of the US DOE for his continued support.

REFERENCES

1. B.J. Travis and K. H. Birdsell, 'TRACR3D: A Model of Flow and Transport in Porous Media," Los Alamos National Laboratory roport LA-11798-M (April 1991).
2. L.W. Gelhar, A. L. Gutjahr, and R. L. Naff, "Stochastic Analysis of Macrodispersion in a Stratified Aquifer," Water Resources Research **15**(6), 1387 - 1397 (1982).
3. D.J. Drew, D.M. Grondin. and T.T. Vandergraaf, 'The Large Block Radionuclide Migration Facility' . Atomic Energy of Canada Limited Technical Record TR-519. 1990.
4. A. N. Tikhonov and V. Arsenin, 'Solution of Ill-posed Problems, ' V. H. Winston & Sons, Washington D. C. 1977
5. B.J. Travis, 'Recovery of Discontinuities using a Parametric Form of Regularized Inversion, " in prep.
6. R. S. Rundberg, I. R. Triay, M. A. Ott, and A. J. Mitchell, "Observation of Time Dependent Dispersion in Laboratory Scale Experiments with Intact Tuff," Radiochimica Acta **52/53**, 219-228 (1991).