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Radionuclide Migration Laboratory Studies for Validation of Batch Sorption Data

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RADIONUCLIDE MIGRATION LABORATORY STUDIES FOR VALIDATION OF BATCH SORPTION DATA

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Abstract

Advective and diffusive migration experiments (within the Dynamic Transport Column Experiments and Diffusion Studies of the Yucca Mountain Site Characterization Project) involve utilizing crushed material, intact, and fractured tuff in order to test and improve (if necessary) transport models by experimentally observing the migration of sorbing and non-sorbing radionuclides on a laboratory scale. Performing a validation of the sorption data obtained with batch techniques (within the Batch Sorption Study) is an integral part of the mission of the Dynamic Transport Column Experiments and Diffusion Studies. In this paper the work scope of the radionuclide migration laboratory experiments (as they apply to validation of batch sorption data) is reviewed.

Previous results of the radionuclide migration laboratory experiments are summarized. Column experiments performed with crushed tuff and pure mineral separates indicate that the sorption coefficients obtained with batch techniques agree with those obtained under flowing conditions for alkaline and alkaline earth elements. The transport data for the actinides through crushed tuff and pure mineral columns can not be fitted with batch sorption coefficients; a small fraction of the actinides is eluted in the column experiments with a sorption coefficient smaller than the one determined in batch experiments. Preliminary results indicate that the reason for the discrepancy is slow kinetics of speciation in natural groundwaters for the actinides. If these results are confirmed with further experimentation, we can conclude that the batch sorption coefficients provide conservative estimates for transport calculations.

Anions including pertechnetate have been studied using crushed tuff column experiments. These experiments indicate that anion exclusion is observed in Yucca Mountain tuffs. Anion exclusion causes anions to migrate faster than non-reactive tracers (such as tritiated water). This effect is due to steric hindrance caused by the large size of anions compared to the apertures of porous tuffaceous minerals (such as zeolites and clays) and repulsion by the negatively charged tuffaceous mineral surfaces in contact with natural groundwater.

Tuff columns made with solid tuff, under saturated conditions, indicate that batch sorption coefficients, obtained utilizing batch techniques, can be used to fit elution data of alkaline and alkaline earth elements provided that a detailed characterization of the hydrologic properties of the tuff is obtained. The radionuclide sorption mechanism observed under saturated conditions is expected to be the same as that observed under unsaturated conditions. The hydrologic properties dominating the transport of radionuclides under unsaturated conditions are expected to be quite different. The experimental methods to test this hypothesis (which will allow the utilization of sorption parameters obtained under saturated conditions to be applied to unsaturated transport) are currently under design.

Results from column experiments with fractured tuff indicate good agreement between fracture transport models and the observed elution of nonsorbing radionuclides. The sorbing radionuclides exhibited a small fraction which eluted unretarded through the fracture. Whether or not this observation is due to complicated hydrologic properties (such as channeling, dispersion) in the fracture is unknown.

Diffusion is considered one of the most important retardation mechanisms in fractured media. The transport laboratory experiments under diffusive conditions performed indicate that the diffusion coefficient for nonsorbing radionuclides into saturated Yucca Mountain tuff is on the order of 10^{-6} cm^2/s. Large anions, such as pertechnetate are excluded from tuff pores and their diffusion coefficients are on the order of $10^{-7} cm^2/s$. The diffusion of the sorbing radionuclides into saturated tuff could not be fitted assuming reversible, instantaneous, and linear sorption. Comparison of the fits for the sorbing radionuclides with the actual data obtained indicates that conservative transport calculations will result from a batch sorption coefficient and the diffusion coefficient obtained for tritiated water.

The strategy to validate batch sorption data utilizing laboratory transport experiments is presented. This strategy consists of performing column experiments utilizing crushed material and comparing the results to the results obtained from batch sorption studies in order to study the effects of kinetics of speciation, mass transfer kinetics, or non-linear, non-reversible, or non-instantaneous sorption. When differences between batch sorption experiments and crushed rock columns are resolved, the next level of complexity will be tackled. Solid tuff columns will be utilized to assess the effects of surface properties (caused by crushing) on batch sorption coefficients. Diffusion experiments utilizing solid tuff will also be utilized for validation of batch sorption coefficients. Crushed rock columns and solid tuff advective and diffusive experiments will be conducted under varying degrees of saturation. This will allow assessment of the validity of utilizing batch sorption coefficients (obtained under saturated conditions) to calculate transport under unsaturated conditions. The final level of complexity will involve utilizing fractured tuff columns to validate the concept of utilization of batch sorption coefficients in fractured media.

Introduction

The transport of radionuclides in porous media is governed[1] by advection, diffusion, or kinematic dispersion. Dissolved species are carried along by the movement of fluid in an advective migration mechanism. Species are transfered from zones of high concentration to zones of low concentration by diffusion. Kinematic dispersion is a mixing phenomenon linked to the heterogeneity of the microscopic velocities inside the porous medium. The transport equation[1] describing the migration of a solute in a saturated porous medium is given by equation 1.

$$\nabla\bullet(D\nabla C - CU) = \varepsilon\, \partial C/\partial t + Q, \qquad (1)$$

where

D = dispersion tensor,
C = concentration of solute in solution phase,
U = Darcy's velocity,
ε = porosity of medium,
t = time,
$Q = 0$ for a non-reactive solute,
$Q = (1-\varepsilon)\rho_S\, \partial F/\partial t$ for a sorbing solute,
ρ_S = density of the solid particles, and
F = mass of solute sorbed per unit mass of solid.

Dispersion has three components: D_L, the longitudinal dispersion coefficient in the direction of the flow and D_T, the transverse dispersion coefficient in the two directions at right angles to the velocity of the flow. The longitudinal and transverse dispersion coefficients are given by equations 2 and 3, respectively.

$$D_L = \varepsilon d + \alpha_L |U|, \qquad (2)$$

$$D_T = \varepsilon d + \alpha_T |U|, \qquad (3)$$

where

d = effective diffusion coefficient in medium, and
α = dispersivity.

In the absence of advection ($U=0$), equation 1 becomes equation 4 which describes the migration of solutes by diffusion in a saturated medium.

$$\nabla\bullet(\varepsilon d\nabla C) = \varepsilon\, \partial C/\partial t + Q \qquad (4)$$

The mechanism of sorption determines the relationship between F and C. When sorption is linear, reversible, and instantaneous, the relationship between F and C is given by equation 5, where K_d is the sorption coefficient.

$$K_d = F/C \tag{5}$$

Substitution of equation 5 into equation 1 yields equation 6. The expression in brackets in equation 6 is the retardation factor, R_f. Equation 7 provides a means of comparing results for sorption coefficients obtained under advective and diffusive conditions with the sorption coefficients obtained utilizing batch sorption experiments. The expression for R_f (given in equation 7) is only valid if sorption is linear, reversible and instantaneous. The Langmuir and the Freundlich isotherms are examples of non-linear relationships between F and C.

$$\nabla \bullet (D\, \nabla C - CU) = \varepsilon\, [1 + (1 - \varepsilon)\, \rho_s\, K_d/\varepsilon\,]\, \partial C/\partial t \tag{6}$$

$$R_f = 1 + (1 - \varepsilon)\, \rho_s\, K_d/\varepsilon \tag{7}$$

When migration occurs under unsaturated conditions, dispersion is a function of moisture content and equation 1 becomes equation 8.

$$\nabla \bullet (D(\theta) \nabla C - CU) = \varepsilon C \partial \theta / \partial t + Q, \tag{8}$$

where

θ = moisture content.

Inspection of equations 1, 4, and 8 indicates that advection and diffusion experiments can be utilized to validate the utilization of batch sorption coefficients for transport calculations.

Experimental Work Scope

The work scope of the Dynamic Transport Column Experiments Study[2] of the Yucca Mountain Site Characterization Project consists of performing crushed material, solid, and fractured tuff column experiments to validate the sorption coefficients obtained via batch experiments. Column experiments with crushed tuff and pure mineral separates are utilized to test the results of batch sorption experiments under flowing conditions without altering the surface properties of the tuff. The crushed material column studies are most sensitive to multiple solution species formation and colloid formation. The kinetics of sorption are studied by varying the water velocity in column experiments.

Solid tuff columns are used to address the question of whether crushing affects sorption measurements in the batch experiments. The column experiments will be performed as a function of saturation to address whether sorption coefficients obtained via batch sorption techniques (under saturated conditions) are applicable to unsaturated transport calculations.

Utilization of gravity as a driving force to obtain homogeneous distributions of water for the study of radionuclide transport in an unsaturated system requires very long times. Consequently, we will utilize vacuum[3] or centrifugation[4] to conduct transport experiments in unsaturated media.

The Soil Measurement Systems apparatus (designed by Wierenga) for unsaturated transport studies (see Figure 1) provides accurate determination of radionuclide migration through unconsolidated media.[3] In this system, water containing the radionuclide of interest is introduced into a tuff-filled column. The typical dimensions of the column are given in the Figure 1. A precision syringe pump is utilized to maintain constant fluid flow. The regulator-controlled vacuum chamber provides a nearly constant degree of saturation throughout the column and houses the fraction collector. Tensiometers allow accurate measurement of soil-water pressure via a transducer. When tensiometer values are nearly equal, the water distribution throughout the column is considered to be homogeneous. After elution through the column, small fractions of the solution are collected in the fraction collector. These fractions are analyzed for radionuclide concentration to determine the rate of radionuclide migration.

Centrifugation techniques can also be utilized[4] to study unsaturated transport. Conca designed the Unsaturated Flow Apparatus (see Figure 2) consisting of a modified ultracentrifuge with an ultra constant rate flow pump that provides water (containing the radionuclide of interest) to the surface of the solid material through a rotating seal. Rotation speeds up to 20,000 rpm are attainable at temperatures up to 150°C and flow rates down to 0.1 ml/h, making possible investigations with unconsolidated and consolidated materials. Elutions are collected in the collection vessel and analyzed for radionuclide concentration. The Unsaturated Flow Apparatus is capable of running two columns at the same time; however, only one column is shown to be operational in Figure 2.

The systems in Figures 1 and 2 (designed by Wierenga and Conca, respectively) will be adapted to study the transport of actinides (under unsaturated conditions). This will involve modifications to avoid contact of the solutions containing the actinides of interest with metallic non-geologic surfaces which tend to sorb the actinides.

Fracture tuff columns are utilized to investigate the transport of radionuclides with

FIGURE 1: SOIL MEASUREMENT SYSTEMS APPARATUS FOR UNSATURATED TRANSPORT STUDIES (from reference 3)

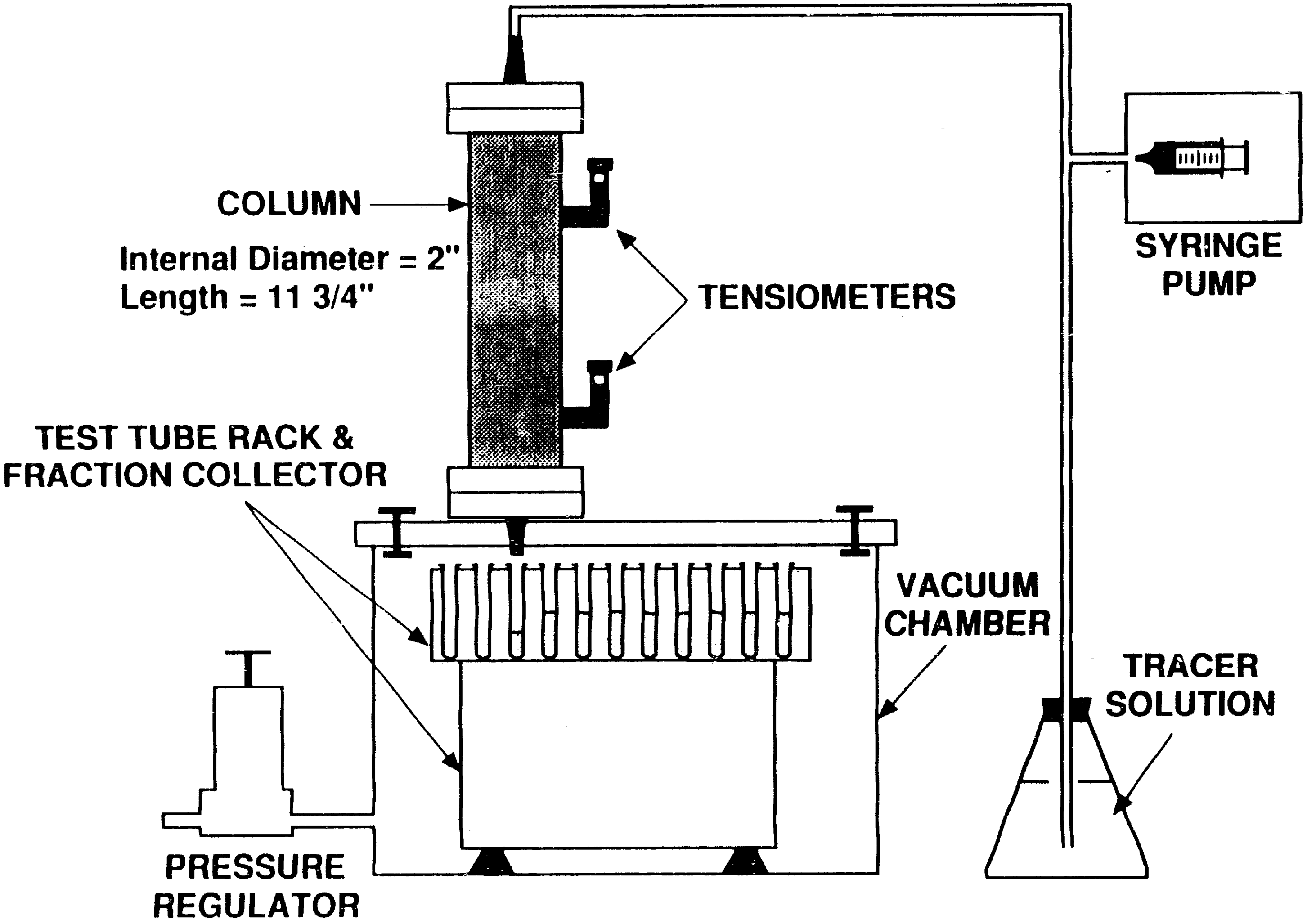
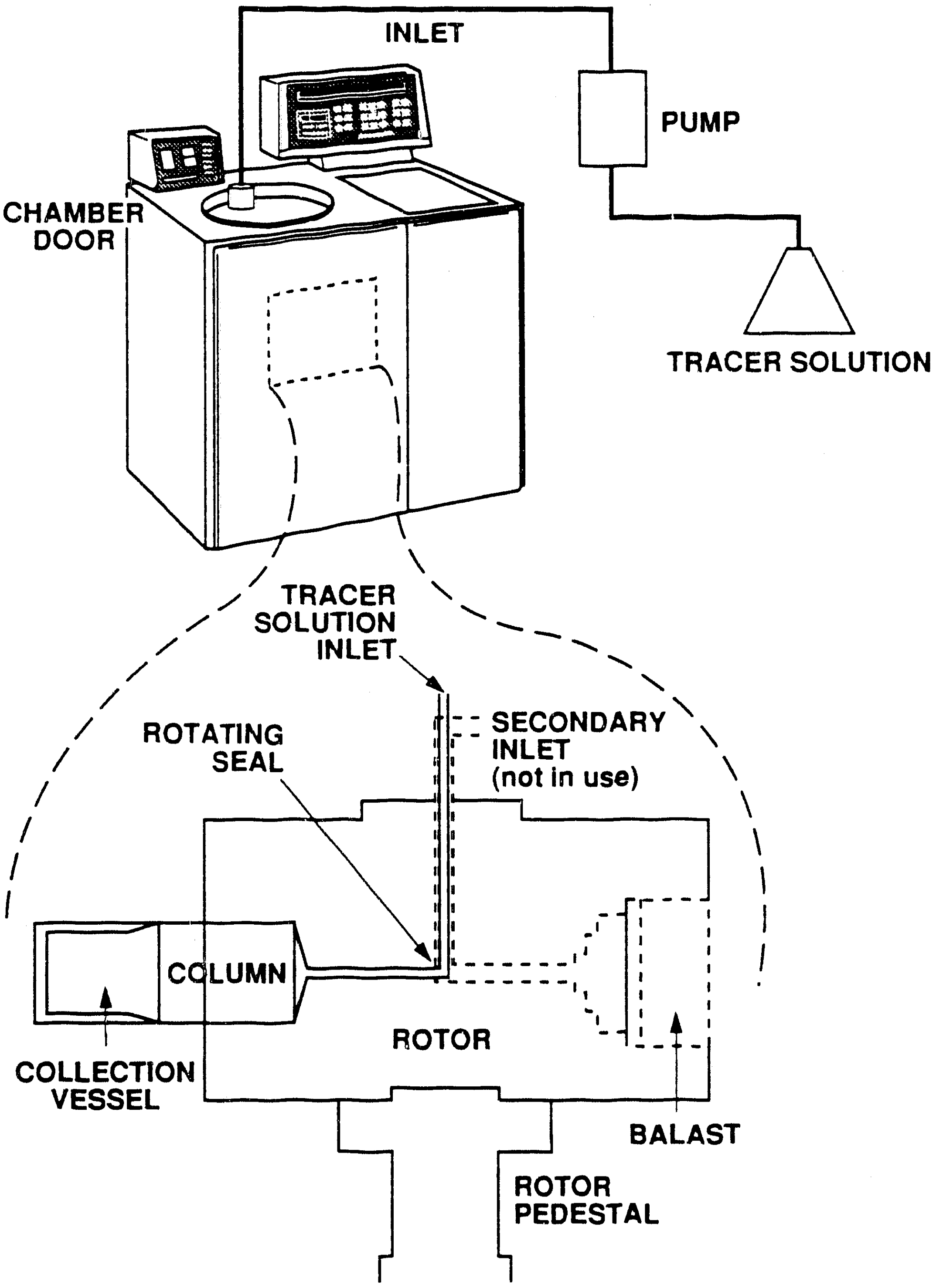


FIGURE 2: UNSATURATED FLOW APPARATUS
(from reference 4)



transverse matrix diffusion and channeled flow in fractures. These columns experiments will allow the assessment of the applicability of batch sorption coefficients to transport calculations in fractured media.

The work scope of the Diffusion Study[5] of the Yucca Mountain Site Characterization Project consists of performing transport experiments with solid tuff (in the absence of advection) under various degrees of saturation. The first type of diffusion experiment planned consists of a cell with two chambers containing groundwater. The chambers are separated by a slab of tuff. Radionuclide(s) of interest is placed in one of the chambers and the untraced chamber is monitored as a function of time for the arrival of the radionuclide(s). The retardation factor is determined for sorbing radionuclides and compared to the sorption coefficients (determined via batch techniques).

The second type of diffusion experiment consists of studying the kinetics of sorption under diffusive conditions by immersing a wafer made of tuff in a solution containing the radionuclide(s) of interest. The uptake of radionuclides(s) by the thin wafers is determined. The actinides exhibit sorption onto non-geologic media (such as the walls of containers). The kinetics of sorption under diffusive conditions is studied utilizing containers made of tuff in the form of beakers. The solution containing the radionuclide(s) of interest is placed in the tuff beaker cavity and the uptake of the radionuclide(s) by the tuff is measured as a function of time.

Unsaturated diffusion experiments are also planned. A solution containing the radionuclides(s) of interest is injected into an unsaturated block of tuff. The distribution of radionuclide(s) in the tuff as a result of diffusion will be determined by sectioning the block of tuff and measuring the radionuclide(s) concentration throughout the tuff block.

Summary of Transport Results

The results of column transport experiments have been summarized[6]. Batch sorption coefficients have been compared with the sorption coefficients obtained from crushed tuff columns utilizing equation 7. The results are in good agreement for the cations cesium, strontium, and barium[7]. Elution of actinide solutions through columns made of crushed pure mineral separates[8] and crushed tuff[9] shows a fraction of the actinide eluting with a sorption coefficient corresponding to a smaller value than the one obtained with batch sorption techniques. For instance, Figure 3 shows a fraction of the Pu in a solution prepared with J-13 water and Pu(IV) polymer eluting before the tritiated water (which was utilized as a non-reactive tracer). Figure 4 shows the elution of Pu in a solution prepared with J-13 water and Pu(VI).

A preliminary conclusion of the column experiments reported by Triay et al.[8] and Thompson[9] is that the equilibration of actinide species in solution is slow. Slow kinetics

Figure 3: Elution of Tritium (●) and Pu(IV) Polymer (❑)

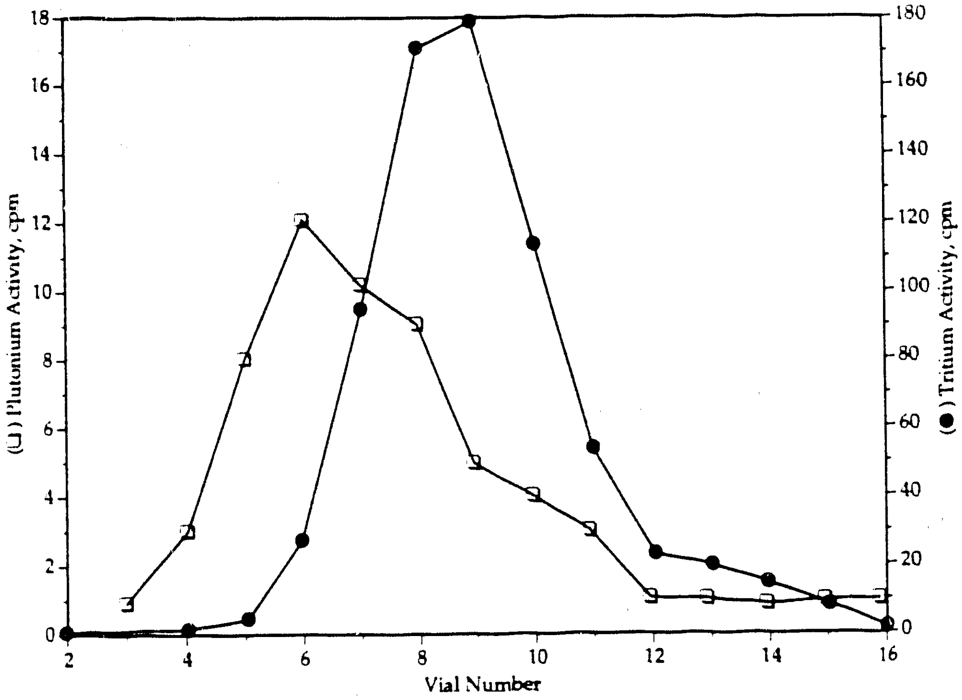
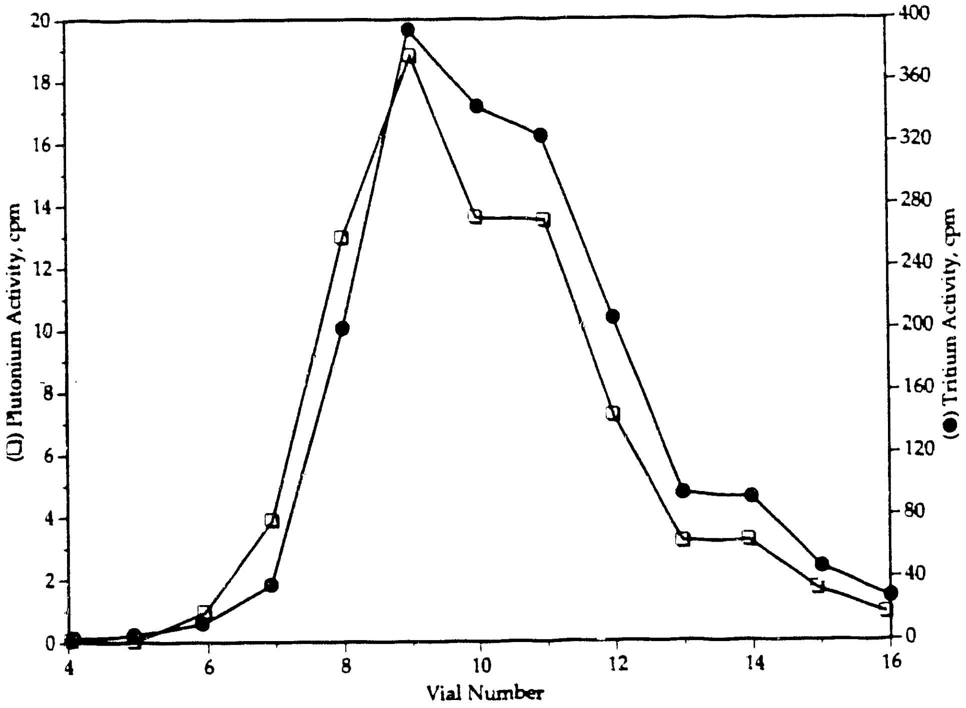


Figure 4: Elution of Tritium (●) and Pu(VI)(❑)



of speciation in solution will result in conservative batch sorption coefficients values. The reason for this conclusion is that if multiple actinide species exist in solution that vary in their ability to sorb, and interconversion between the lowly sorbing and highly sorbing species is slow, in large periods of time all the lowly sorbing species will be converted to highly sorbing species and will be retarded with a sorption parameter of at least the magnitude of that determined in the batch experiments.

Anions[6] including pertechnetate have been studied using crushed tuff column experiments. These experiments indicate that anion exclusion is observed in Yucca Mountain tuffs. Anion exclusion causes anions to appear to migrate faster than tritiated water. This effect is due to steric hindrance caused by the large size of anions compared to the apertures of porous tuffaceous minerals (such as zeolites and clays) and repulsion by the negatively charged tuffaceous mineral surfaces in contact with natural groundwater. The anion exclusion volumes determined (in the range of 0.03 to 0.1 ml/g) were found to agree with the intracrystalline pore volumes calculated from the zeolitic and clay content of the tuffs utilized.

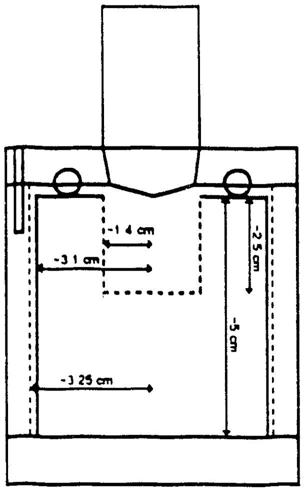
The migration of radionuclides through intact solid rock columns was investigated.[10] The elution of sorbing cations (Cs, Sr, and Ba) through devitrified tuff exhibited greater tailing in the elution curves that could be fitted by adjusting the dispersion coefficient in equation 1. The elution of the sorbing cations through solid tuff columns could be fitted with the measured batch sorption coefficient provided time-dependent dispersion was invoked. Whether the anomalous dispersion behavior is the result of the finite size of the solid columns is currently under study. Bacri et al. [11] have reported concentration profiles (in unsaturated porous media) that exhibit long tails as a result of the sample length being too small to achieve a statistical random walk.

A transport model[12] was utilized to fit the elution of nonsorbing radionuclides through fractures[13]. The model consists of vertical flow through fractures with horizontal diffusion into the matrix perpendicular to the fracture.[12] Elution of the sorbing radionuclides through the fracture exhibited a small fraction of the radionuclide eluting early. This elution behavior could be due to dispersion or channeling phenomena.

Rock beaker experiments were utilized to study the diffusion of radionuclides through saturated Yucca Mountain tuffs.[14] The experimental technique involved fabricating containers in the form of a beaker (made of tuff). Figure 5 shows a cross section of a rock beaker encapsulated in a plexiglass container. The rock beaker sits on the plexiglass container (surrounded by a layer of J-13 water on the sides). A plexiglass stopper is utilized to prevent evaporation. The beaker has an orifice with a radius of 1.4 cm and length of 2.5 cm. The rock beaker has a length of 5 cm and a radius of 3.1 cm.

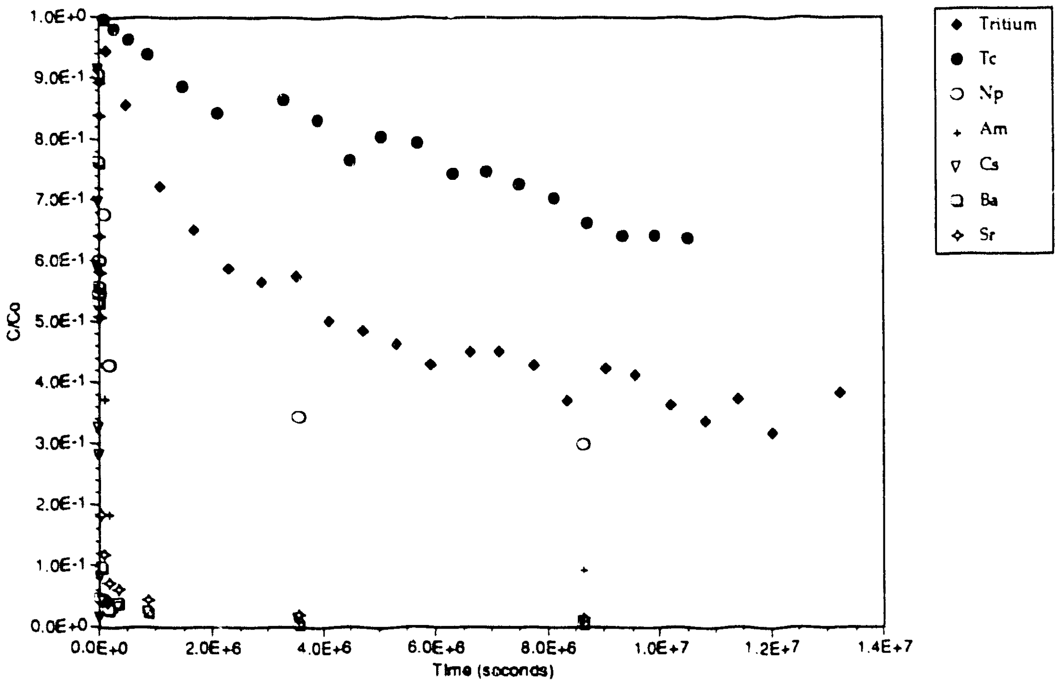
The radionuclides utilized for these experiments were H-3, Tc-95m, Np-237, Am-241, Sr-85, Cs-137, and Ba-133. A J-13 solution containing the radionuclides of interest was

Figure 5: Sample Rock Beaker



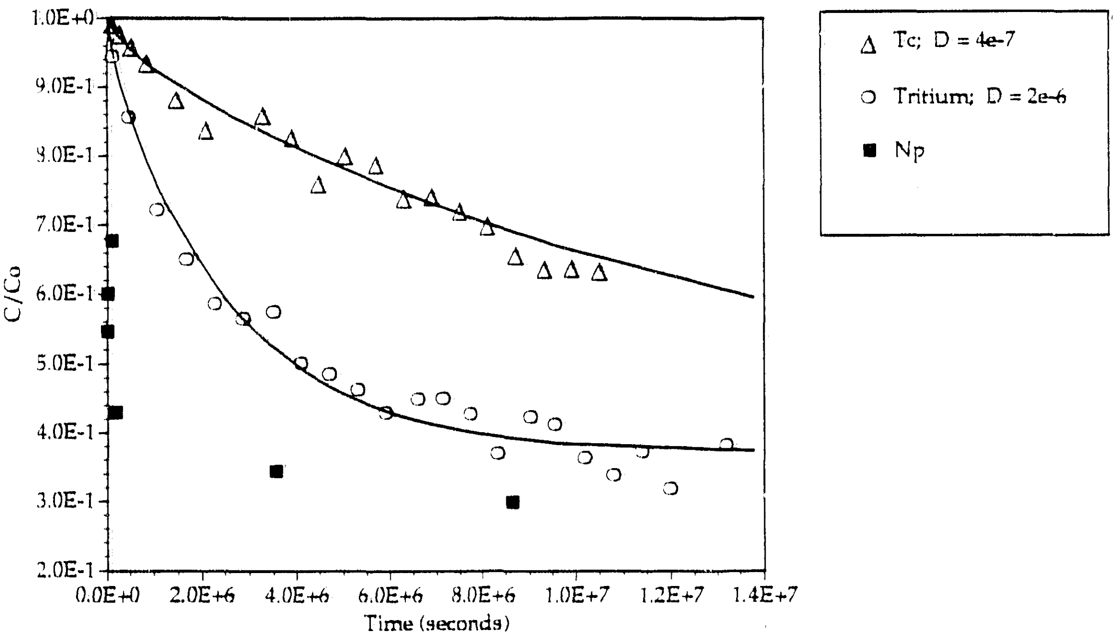
placed in the cavity of the rock beakers. Aliquots of the solution containing the radionuclides of interest were taken as a function of time and analyzed for radionuclide concentration. Figure 6 shows the concentration left in the solution in the cavity of the rock beaker (made with tuff G4-737) divided 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.

Figure 6: Diffusion Data for Rock Beaker G4-737



The diffusion data were fitted to the diffusion equation, given by equation 4, utilizing the transport code TRACRN.[15] Figure 7 shows the fits obtained (solid lines) utilizing TRACRN for non-sorbing radionuclides (H-3 and Tc-95m) in a beaker made of tuff G4-737. The diffusion coefficients obtained for the non-sorbing radionuclides in all the tuffs studied are given in Table 1. Large anions such as pertechnetate are exclude from tuff pores due to their size and charge. The diffusion coefficients obtained for non-sorbing radionuclides agree well with previous results.[16]

Figure 7: Fits (solid lines) to diffusion data for rock beaker G4 - 737



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 (K_d). In order to test this assumption batch sorption experiments were performed utilizing the tuffs studies. The batch sorption procedure consisted of pre-treating 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 pre-treated tuff, separating the phases by centrifugation, and determining the amount of radionuclide in both phases. The sorption coefficients (Kd) determined in this manner are given in Table 2.

The diffusion of the sorbing radionuclides could not be fitted assuming reversible,

Table 1: Diffusion Results

Tuff Sample	Major Minerals, %	Porosity	D (cm²/s)	
			HTO	TcO_4^-
G4-737	alkali feldspar, 68 cristobalite, 28	0.07	2.2×10^{-6}	3.9×10^{-7}
GU3-304 #1	alkali feldspar, 75 cristobalite, 25	0.06	1.5×10^{-6}	3.0×10^{-7}
GU3-304 #2			1.6×10^{-6}	3.0×10^{-7}
GU3-433	alkali feldspar, 76 cristobalite, 15	0.10	3.5×10^{-6}	
GU3-1119	alkali feldspar, 70 quartz, 19	0.10	2.0×10^{-6}	4.9×10^{-7}
Topopah Outcrop	alkali feldspar, 59 cristobalite, 23 quartz, 12	0.07	1.0×10^{-6}	1.0×10^{-7}

Table 2: Sorption Results

Tuff Sample	Major Mineral, %	K_d (ml/g)				
		Np	Am	Cs	Sr	Ba
G4-737	alkali feldspar, 68 cristobalite, 28	8	134	532	52	28
GU3-304	alkali feldspar, 75 cristobalite, 25	8		342	18	19
GU3-433	alkali feldspar, 76 cristobalite, 15	9	154	1264	20	61
GU3-1119	alkali feldspar, 70 quartz, 19	8	136	494	42	27
Topopah Outcrop	alkali feldspar, 59 cristobalite, 23 quartz, 12	9		465	20	25

instantaneous, and linear sorption. Figure 8 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 9) 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.[17] 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 intra-crystalline channels. This diffusion model yielded sorption coefficients that agree well with sorption coefficients determined with batch techniques for the cations Cs, Sr, and Ba.

Figure 8: Calculated Diffusion Data for Rock Beaker G4-737

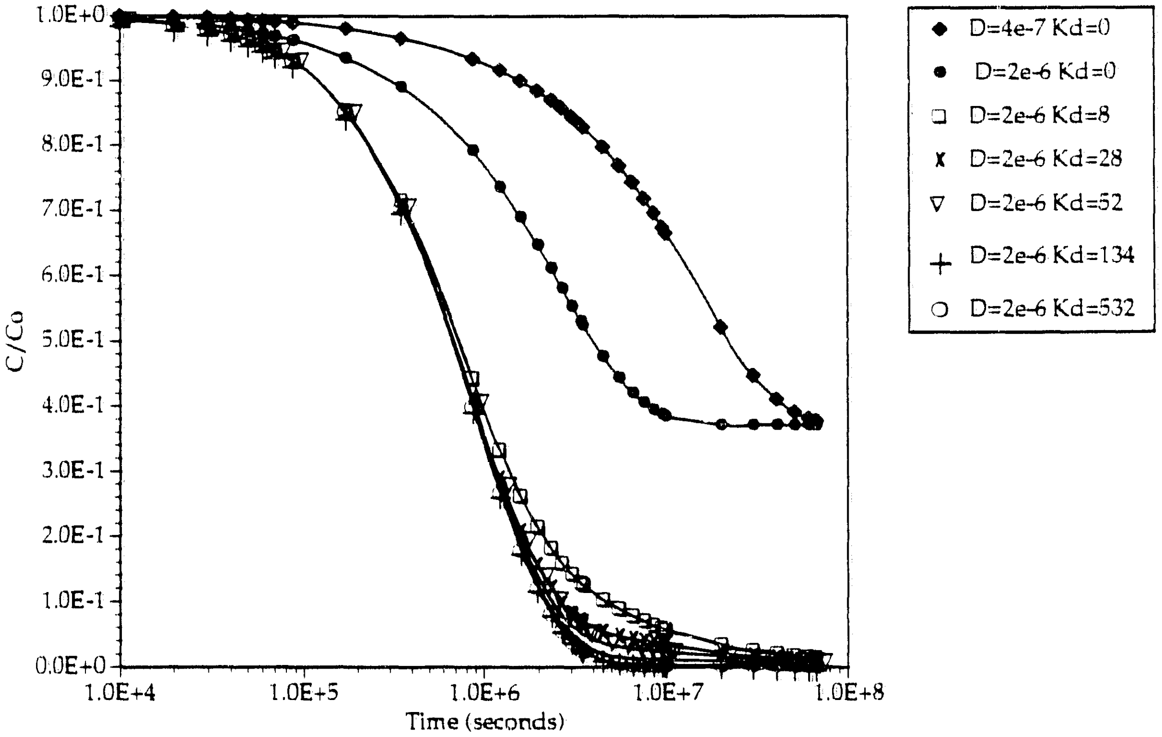
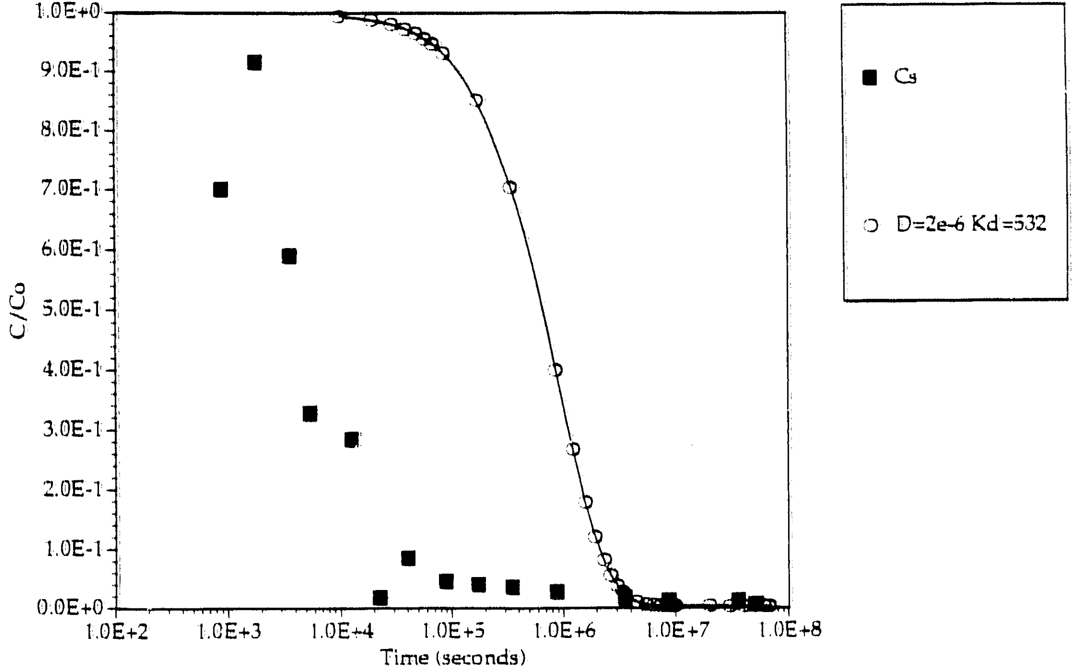


Figure 9: Comparison of Fit Obtained (solid line) with Actual Cs Data (solid squares) for Rock Beaker G4-737



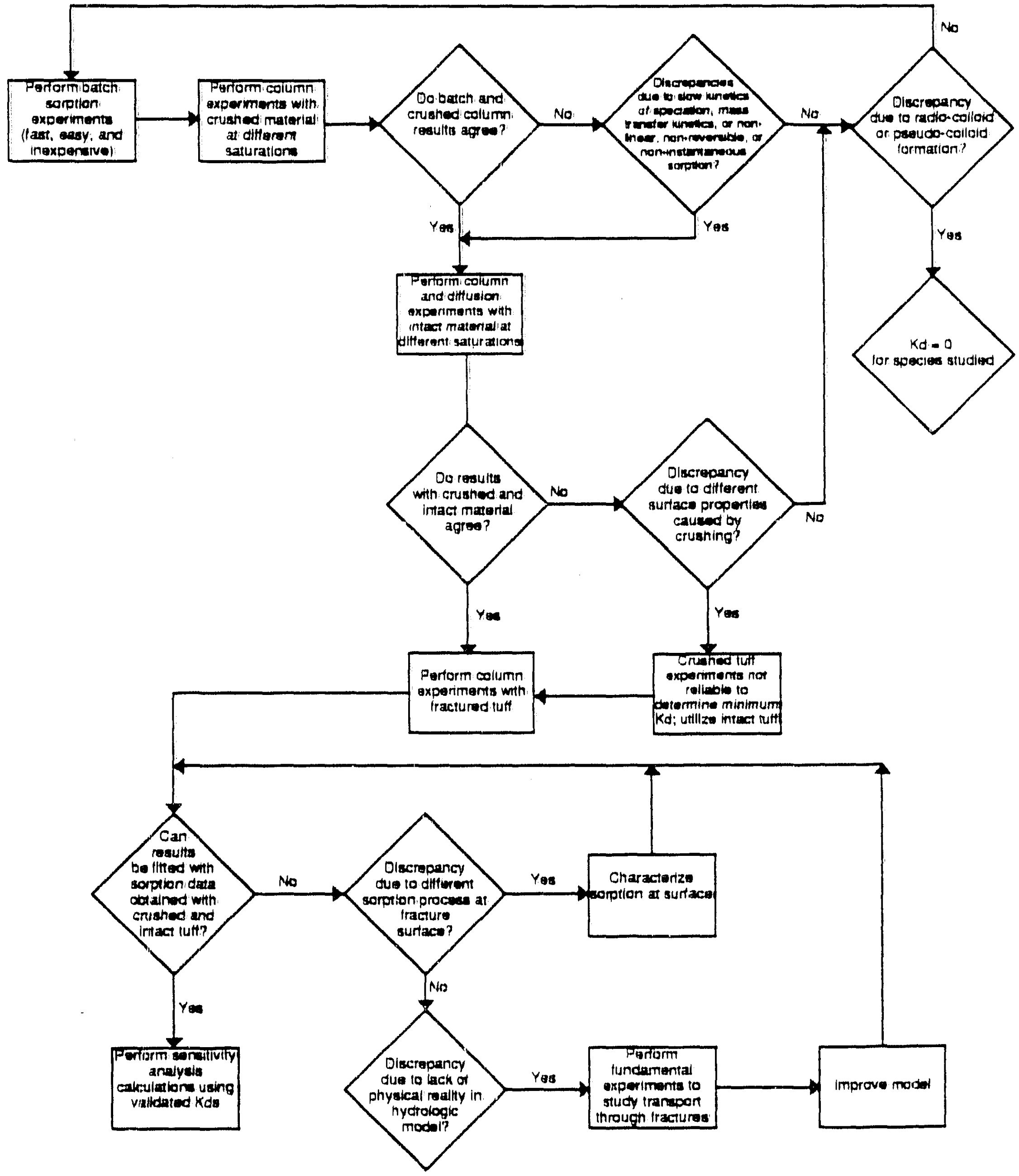
Strategy for Validation of Batch Sorption Data

Figure 10 shows the strategy to be utilized to validate batch sorption data utilizing laboratory transport experiments under advective and diffusive conditions. Batch sorption experiments will be performed to obtain sorption coefficients and identify sorption mechanisms. These experiments are fast, easy, and inexpensive as compared to column experiments. Results from column experiments utilizing crushed material will be compared with results from batch experiments (using equation 4). Discrepancies may be due to slow kinetics of speciation in solution, mass transfer kinetics, or sorption which is non-linear, non-reversible or non-instantaneous. Column experiments will also be sensitive to the formation of radio-colloids (such as Pu(IV) polymer) or pseudo-colloids (such as radionuclides adsorbed onto minerals suspended in groundwater).

Provided that the discrepancies between crushed rock columns and batch techniques can be resolved and that the sorption coefficient (given by the batch technique) yields a conservative value, the effect of crushing will be studied. Intact tuff will be utilized in column and diffusion experiments to assess whether batch sorption coefficients obtained with crushed material are applicable to transport calculations.

Varying the degree of saturation in column and diffusion experiments will allow

Figure 10: Strategy for Validation of Batch Sorption Data



assessment of the applicability of batch sorption coefficients (obtained under saturated conditions) to transport calculations under unsaturated conditions. Fracture tuff columns will be utilized to study transport through fractured tuff. The transport model describing vertical transport through fractures, diffusion from the fractures into the matrix, and sorption within the matrix (described by a batch sorption coefficient) will be tested.

Concluding Remarks

The validation of batch sorption data described in this paper has as its main objective the validation of the concept of utilizing a batch sorption coefficient in transport calculations. The fact that Figure 10 shows a $K_d = 0$ for colloids simply means that it is acknowledged that colloids may travel as fast as the groundwater and that colloid transport will probably not be treated utilizing the batch sorption K_d approach.

It has been postulated by calculations in the water chemistry efforts of YMP[18] that the amount of particulate matter in the groundwaters collected in the vicinity of Yucca Mountain is not significant enough to carry a sizeable amount of radioactivity; consequently, colloid transport is not important at this site. Since no conclusive evidence exists yet, the Dynamic Transport Column Experiments and the Retardation Sensitivity Analysis Studies of YMP are designing a colloid strategy in order to quantify the genesis of colloids (by natural processes and as a result of emplacement) as well as the transport of colloids through tuff.

Preliminary experiments in the field of colloids involved the determination of size distribution of Pu(IV) polymers[19] and the study of colloid transport of well-characterized synthetic colloids (in the size range from 0.1 to 10 micrometers) through saturated fractures.[6] No elution of the colloidal material was observed in the fracture experiments except in the case of the 1-micrometer colloids. Twenty-four percent of the input material in the case of the 1-micrometer colloids was eluted through the fractured tuff column. This observation is in agreement with the predicted size dependence of colloid filtration transport models.[20]

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