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Waste Isolation Safety Assessment Program Summary of FY-77 Progress

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WASTE ISOLATION SAFETY ASSESSMENT PROGRAM

SUMMARY OF FY-77 PROGRESS

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1. INTRODUCTION

The Waste Isolation Safety Assessment Program (WISAP) was initiated on October 1, 1976 at Battelle Pacific Northwest Laboratories to provide long-term safety information for the National Waste Terminal Storage (NWTS) Program. This multidisciplinary, multilaboratory program has the following technical objectives:

1. Provide the generic methods needed to assess the long-term safety of geologic isolation repositories.
2. Obtain the generic and site-specific data necessary to apply the methods.
3. Demonstrate the use of the methods for a specific site.
4. Apply the methods and the data to make safety assessments for sites of the NWTS Program.
5. Analyze societal acceptance issues and develop methods of communicating assessment results which enhance rational resolution of these issues.

A plan was developed to meet these objectives. Figure 1 shows the approximate timing of this plan.

Work in FY-77 can be conveniently classed according to whether it supported the development of the generic assessment method or the development of the generic data base. Table 1 lists the FY-77 activities in each

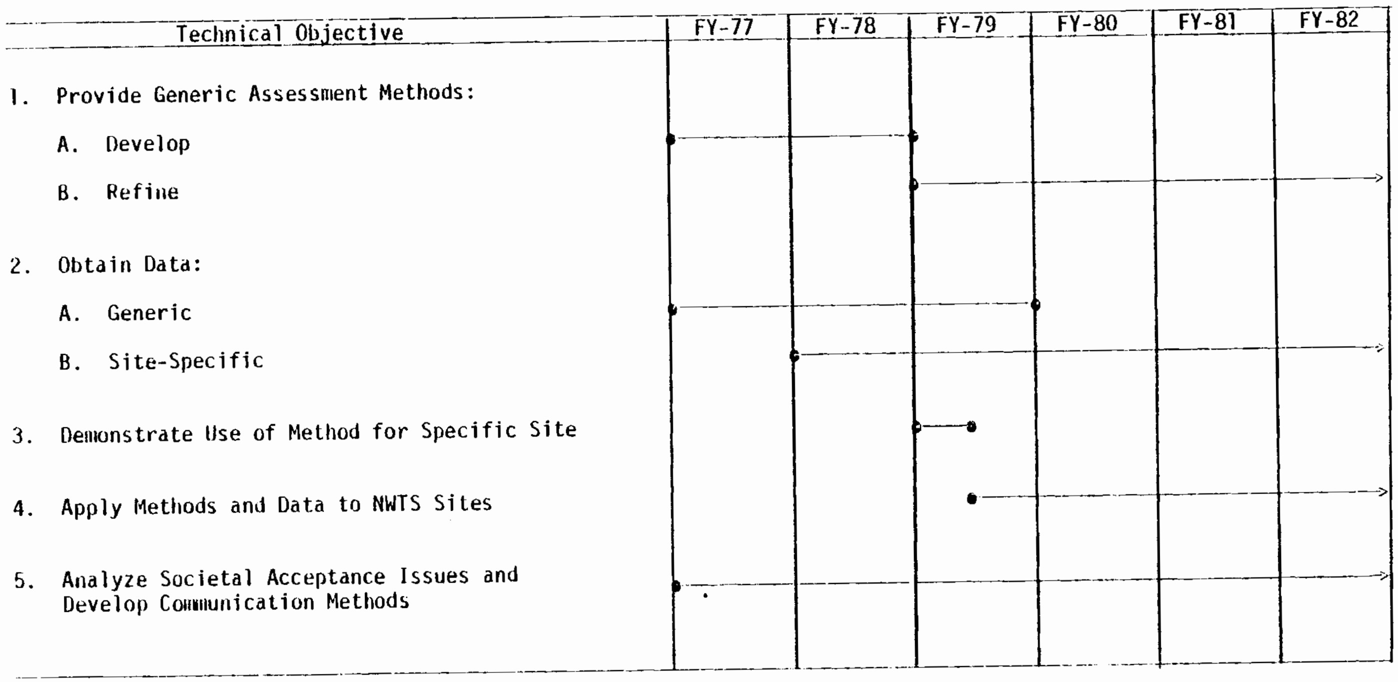

FIGURE 1. WISAP Schedule

TABLE 1. WISAP FY-77 Activities

Generic Assessment Method Development	Generic Data Base Development
1. Analysis of Potential Release Scenarios:	1. Waste Form Release Rate Data:
• Identification of natural and man-caused disruptive events and processes.	• Preparation of sample glass waste forms doped with long-lived radionuclides.
• Evaluation of potentially applicable analysis methods.	• Leaching of doped glass waste forms.
• Development of repository simulation analysis.	
	2. Radionuclide Geochemical Interaction Data:
	• Evaluation of sorption measurement methods.
2. Analysis of Potential Release Consequences:	• Sorption measurements for generic data bank.
• Evaluation of potentially applicable analysis methods.	• Analysis of generic sorption data.
	• Validation studies.
• Development of integrated computational modeling system for methods selected.	• Sorption mechanism studies.

of these classifications.

This is a highly abbreviated report of FY-77 activities. A comprehensive annual report will be issued at a later date.

2. GENERIC ASSESSMENT METHOD DEVELOPMENT

Safety assessments for geologic isolation have six elements: (1) identification of the events and processes which could potentially cause breach of containment at the repository cavity, (2) analysis of the ways these phenomena could combine to cause containment breaches, (3) quantitative description of the radioactive release scenarios, (4) prediction of radionuclide release rates from the repository cavity, (5) prediction of the transport of released radionuclides to man, and (6) prediction of nuclide interaction with man. Assessment method development activities related to the first three elements can be considered together under the term "Release Scenario Analysis", while those related to the last three elements can be considered together under the term "Release Consequence Analysis". Results of FY-77 work in this sector are reported below under the latter headings.

2.1 RELEASE SCENARIO ANALYSIS

2.1.1 Identification of Disruptive Events and Processes

We first identified phenomena that could either individually or in concert cause a breach in containment at a geologic isolation repository.[1,2] These were grouped into four classes: (1) natural, long period geologic processes, (2) natural, short period geologic events, (3) man-caused, short period events, and (4) repository-caused, long period processes. Phenomena in all of these groups are listed in Table 2. Many of the phenomena are interrelated and the distinction between short period and long period is sometimes unclear. Thus the classification shown here is somewhat arbitrary. Furthermore, we make no claim that the list of phenomena is all inclusive.

TABLE 2. Potential Disruptive Phenomena for Waste Isolation Repositories

Natural Processes	Natural Events	Man-Caused Events	Repository-Caused Processes
• Climatic fluctuations • Sea level fluctuations • Glaciation • River erosion • Sedimentation • Tectonic uplift • Tectonic subsidence • Volcanic extrusion • Igneous intrusion • Diapirism • Diagenesis • Motion on new or undetected fault • Dissolution • Aquifer flux variation	• Flood erosion • New or undetected fault rupture • Seismically induced fracturing • Seismically induced shaft seal failure • Meteorite	Improper Design/Operation: • Shaft seal failure • Improper waste emplacement Undetected Past Intrusion: • Undiscovered boreholes or mine shafts Inadvertent Future Intrusion: • Archeological exhumation • Weapons testing • Nonnuclear waste disposal • Resource mining (mineral, hydrocarbon, geothermal, salt) • Storage of hydrocarbons or compressed air Intentional Intrusion: • War • Sabotage Perturbation of Groundwater System: • Irrigation • Reservoirs • Intentional artificial recharge • Establishment of population center	Thermal, Chemical Potential, Radiation, and Mechanical Force Gradients: • Induced local fracturing • Chemical or physical changes in local geology • Induced groundwater movement • Waste container movement • Increase in internal pressure • Shaft seal failure

2.1.2 Evaluation of Safety Analysis Methods

FY-77 work included identification, description and evaluation of the safety analysis methods which are available to analyze the combined effect of the potentially disruptive phenomena and describe containment breach scenarios. Methods identified include expert opinion/Delphi, perspectives analysis, consequence analysis, stability analysis, fault/event trees, and Monte Carlo/Markov chain analysis. All of these methods are briefly described in the next six paragraphs.

Expert opinion is a qualitative technique in which a recognized body of experts determines whether an activity is acceptably safe or not. This was the "original" waste isolation safety assessment method applied in 1957 by the National Academy of Sciences in recommending that high-level wastes be stored in bedded salt formations.[3] Delphi is a formally structured method of obtaining expert opinion which produces an estimate of the certainty of the safety conclusion in addition to the conclusion itself.[4,5] This method has never been applied to the waste isolation safety assessment problem. Both of these methods have the advantage of simplicity but the disadvantage of subjectivity. This latter quality has become increasingly significant in the last decade with the growth of distrust of technology and technologists. In recent years expert opinion has usually only had weight when backed by a detailed exposé of the logic and data of a quantitative assessment.

Perspectives analysis is a qualitative technique for comparing one safety attribute of a given activity with a safety attribute of some other activity which is well understood or at least more familiar. This technique has been heavily used in the area of nuclear waste isolation to compare the potential hazard of the waste with the original ore from which it came and with other naturally occurring toxic materials.[6] As with the other qualitative safety assessment methods, perspectives analysis has the advantage of simplicity and the disadvantage of subjectivity. A particular difficulty with this technique is that conflicting conclusions can result from the same analysis depending on which attributes are compared and what assumptions are made in the comparison.

Consequence analysis is a quantitative method in which the safety assessment is based only on the consequences of assumed release scenarios. These release scenarios are simply postulated by expert opinion and no consideration is given to their likelihood of occurrence. This method has been applied to estimate the consequences of groundwater intrusion into and meteoritic exhumation of a waste isolation repository in particular but hypothetical geologic and biologic settings.[7,8] Consequence analysis has the advantage of estimating the quantitative impact of release scenarios and hence is a necessary part of all quantitative safety assessment methods, but when utilized by itself it has the disadvantage of biasing the safety assessment toward the low probability-high consequence release scenarios.

Stability analysis is a deterministic technique for analyzing whether important properties of the system of interest remain unchanged if the system is disturbed.[9] Steps in the analysis include the development of a system model, the development of mathematical models for important processes at work within the model system, and the determination of the system's response to mathematically imposed disturbances. Unstable scenarios are further analyzed to determine the expected conditions of system failure. This analysis technique has not been applied to geologic isolation safety assessments. Stability analysis seems well suited for analyzing the processes but not well suited for analyzing the events whose occurrences are difficult to model by continuum mathematics. Thus stability analysis is not a comprehensive technique but rather is a specialized method for analyzing a portion of the assessment problem.

Fault tree analysis is a deductive, deterministic technique by which the component failures leading to system failure can be logically deduced. Application of the technique yields combinations of basic events whose occurrence causes the undesired system failure events. These event combinations are then evaluated by various screening techniques to determine the high risk scenarios and their likelihood of occurrence. Historically the fault tree approach has received considerable attention in analyses of geologic isolation safety.[1,10] Event tree analysis is an inductive, deterministic technique which reverses the fault tree approach by starting with the basic failure events and working forward in time to display their logical propagation to system failure events.[11] There have been no applications of this technique to the assessment of geologic isolation safety. The fault and event tree techniques can be combined into one integrated analysis method. For both techniques a consequence analysis is performed for the identified system failures. Because of its "binary" logic the fault/event tree approach is well suited for quantitatively analyzing the short period events which either occur or do not occur; however, it is not well suited for analyzing the long period processes which occur continuously at some finite rate. Furthermore, because of their deterministic nature, fault/event trees are not well suited for analyzing the interaction of the many different possible sequences of the potentially disruptive phenomena.

Monte Carlo analysis and Markov chain analysis are both stochastic techniques for analyzing system failures when those failures are the result of sequences of events. A Monte Carlo analysis begins by listing the basic failure events and estimating their probabilities of occurrence. The analysis then steps through time assuming occurrence of these events according to their estimated probability distributions until a system failure occurs. By performing the simulation a large number of times, a distribution of failures can be determined. Consequence analyses are then performed on this failure distribution. A Markov chain analysis begins by specifying the possible states of the system and determining the probabilities for transition between those states. Implicit in the analysis is the assumption that the transitional probability between two states is independent of earlier states of the system. The analysis then steps through time assuming probabilistic occurrence of the transitional events until a system failure occurs. Extensive mathematical formalism exists to reduce Markovian processes to analytical functional forms. Neither of these techniques has been applied to geologic isolation safety assessments. Both techniques are well suited to quantitatively analyzing the discrete events, and because of their stochastic nature they are also well suited to analyzing the complex interations of these events. Both of these techniques have the potential disadvantage that a very large number of simulations may be necessary to produce adequate failure distributions for combinations of very low probability events. Monte Carlo analysis can be generalized to consider the continuous processes by updating the state of the system for their effect at each time step. Markov chain analysis, on the other hand, does not appear applicable to the continuous processes because the states of the system must be specified a priori. Furthermore, the transition between states of a geologic system seems likely to be very dependent on previous state transitions, in violation to the basic assumption of Markovian processes.

Our evaluation of these methods for the WISAP release scenario analysis method was based on three criteria: (1) the method should yield quantitative information about potential release scenarios, (2) the method should describe the many possible complex interactions and timing of the potentially disruptive phenomena, and (3) the method should analyze both discrete events and continuous processes. After applying these criteria we concluded that a generalized Monte Carlo analysis was the most applicable technique for assessing the safety of geologic isolation repositories and selected it as the base for construction of the WISAP assessment method, hereafter called repository simulation analysis.

2.1.3 Development of Repository Simulation Analysis

Repository simulation analysis is a combination of the expert opinion/Delphi, fault/event tree analysis, stability analysis, and Monte Carlo analysis techniques. During FY-77 the following steps for implementing the concept were defined:

1. Potentially disruptive natural and man-caused events and processes are listed (see Table 1).

2. The interdependencies of the processes are identified and their coupling relationships are quantified to the extent possiblè (e.g., worldwide warming trend, mean sea level increase, inundation of land over repository, renewed sedimentation and compaction, halokinetic diapirism, stress-induced overburden fracturing, groundwater penetration, and salt dissolution). This requires the expert analysis of geologists and geophysicists.
3. A stratified model of the site-specific geology is made using site exploration data.
4. The potential failure modes are identified.
5. Probabilities for the events and rates, durations, and lag times for the processes are determined by expert opinion from geologists and geophysicists.
6. Repository response curves (e.g., distance to the biosphere vs time) are derived for important system properties by simulating the combined effect of the continuous processes on the site-specific geologic model.
7. The occurrence probabilities of combinations of the events are determined by fault/event tree techniques.
8. The events and their combinations are allowed to occur according to their probability distributions (biasing techniques will be used to obtain proper statistics for very low probability events) on the model system, and their effect is superimposed on the repository response curves for the processes.
9. Containment failures are noted and the failure times are combined with information from the repository response curves to

determine the boundary conditions for subsequent consequence evaluation

A first generation computer code to implement repository simulation analysis was completed during FY-77.

2.2 RELEASE CONSEQUENCE ANALYSIS

2.2.1 Evaluation of Consequence Analysis Methods

During FY-77 we made an extensive compilation of computer models potentially applicable to describing the transport of radionuclides through the geosphere and biosphere and to calculating dose to man. Over 150 hydrologic, contaminant transport, water-dose, and air-dose models were identified.

Information about the hydrologic and contaminant transport models was obtained from a yet-to-be-published international assessment of such models being performed by the Holcomb Research Institute. Information about the water-dose and air-dose models was obtained from recent compilations in open literature. The hydrologic and contaminant transport models were categorized according to Table 3. The water-dose and air-dose models had considerable similarity and did not require a complex categorization.

Our evaluation of the hydrologic and contaminant models for the WISAP release consequences analysis modeling system was based on the following criteria.

1. The system should have a number of levels of modeling sophistication so that a range of data sophistication can be accommodated in a time and cost effective manner.

TABLE 3. Format for Categorizing Hydrologic and Contaminant Transport Models

FLOW →	TRANSPORT ONLY →	SATURATED FLOW SYSTEMS													
		ISOTHERMAL CASES (CONSTANT FLUID PROPERTIES)							ISOTHERMAL CASES (VARIABLE FLUID PROPERTIES)			NON ISOTHERMAL CASES			
		CONFINED AQUIFER			UNCONFINED AQUIFER			CONFINED AND UNCONFINED OR MULTIPLE AQUIFER (THREE DIM. AND BOUSSINESQ APPROXIMATE EQ.)	TWO FLUIDS WITH SHARP INTERFACE		COMBINED FLOW AND TRANSPORT WITH DISPERSION CONSIDERED	CONSTANT FLUID PROPERTIES	VARIABLE FLUID PROPERTIES		
					FREE SURFACE										
TRANSPORT ↓		ANALYTICAL AND/OR SERIES SOLUTIONS FOR IDEALIZED CASES	NUMERICAL SOLUTIONS FOR CASES WITH VERTICAL INFLOWS	NUMERICAL SOLUTIONS COMBINING STRESS AND ELASTIC DEFORMATIONS	WITH LEAKAGE AND/OR ACCRETION	WITH VERTICAL PARTIALLY SATURATED FLOW	DUPUIT-FORCHHEIMER APPROXIMATION (BOUSSINESQ EQ.)		ONE FLUID STATIC	BOTH FLUIDS DYNAMIC			FLOW BUT NO DISPERSION CONSIDERED	COMBINED FLOW AND TRANSPORT WITH DISPERSION CONSIDERED	
FLOW ONLY															
SUBSURFACE CONTAMINANT TRANSPORT — NON-REACTING CONTAMINANTS — ADVECTION ONLY (FLOW PATHS)															
SUBSURFACE CONTAMINANT TRANSPORT — NON-REACTING CONTAMINANTS															
CONTAMINANTS INVOLVED IN EXCHANGE AND CHEMICAL REACTIONS WITH ADVECTION AND DISPERSION — EQUILIBRIUM REACTIONS — ONE SPECIES — ADVECTION PLUS DISPERSION															
EQUILIBRIUM REACTIONS — CHEMICAL N SPECIES — NON-INTER-ACTING															
EQUILIBRIUM REACTIONS — CHEMICAL N SPECIES — INTER-ACTING															
KINETIC RATE REACTIONS — ONE SPECIES															
KINETIC RATE REACTIONS — CHEMICAL N-SPECIES — NON INTER ACTING															
KINETIC RATE REACTIONS — CHEMICAL N-SPECIES — INTER ACTING															
WITH RADIONUCLIDE DECAY CHAINS — EQUILIBRIUM REACTIONS — NON-INTER ACTING															
WITH RADIONUCLIDE DECAY CHAINS — EQUILIBRIUM REACTIONS — INTER-ACTING															
WITH RADIONUCLIDE DECAY CHAINS — KINETIC REACTIONS — NON INTER ACTING															
WITH RADIONUCLIDE DECAY CHAINS — KINETIC REACTIONS — INTER-ACTING															

2. The system should have the capability to model saturated flow through heterogeneous geologic media.

3. The system should have the capability to model hydrodynamic dispersion, radioactive chain decay, equilibrium sorption and time variant leach rates.

These criteria directly follow from programmatic direction to ignore near-field effects. These effects were addressed in other parts of the NWTS Program during FY-77. Particularly in the case of the hydrologic models, a number of models of essentially equal applicability according to these criteria were identified. In these cases we chose models which could be most efficiently adapted to our computing system. The characteristics of a number of the models chosen are shown in Table 4.

The WISAP system does not presently contain models to describe water flow in partially saturated systems. Such models will be added later if situations are identified where use of these models would lead to a substantially better prediction of release consequences. The contaminant models of the WISAP transport modeling system do not presently consider phenomena such as finite rate sorption, multiple chemical species of the same nuclide, chemical reactions between these multiple species, and irreversible mineralization. GETOUT and MMT will be extended to model these and other phenomena if the geochemical interaction data gathering activities of WISAP show that these phenomena are important for conditions of interest to geologic isolation safety assessments.

Our evaluation of dose models for the WISAP modeling system was based on the following criteria:

1. The system should model drinking water, terrestrial food, water

TABLE 4. Characteristics of WISAP Transport Models

HYDROLOGIC	
PATHS	Two-dimensional, analytical/numerical method, homogeneous geology, saturated flow[12]
VTT	Two-dimensional, numerical method, heterogeneous geology, saturated flow[13]
DAVIS FE	Three-dimensional, numerical method, heterogeneous geology, saturated flow[14]
CONTAMINANT TRANSPORT	
GETOUT	One-dimensional, analytical method, chain decay, single speciation, equilibrium sorption, constant leach rate, dispersion[15]
MMT	Three-dimensional, numerical method, single speciation, equilibrium sorption, time variant leach rate, dispersion[16]
WATER DOSE	
ARRRG	Drinking water, immersion, external shoreline, and aquatic food doses[17]
FOOD	Terrestrial food dose[18]
AIR DOSE	
KRONIC	Chronic external dose[19]
SUBDOSA	Acute external dose[20]
DACRIN	Chronic or acute inhalation dose[21]

immersion, external shoreline, and aquatic food doses.

2. The system should model chronic and acute air immersion and inhalation doses.

3. The system should calculate both individual and population doses.

In both the water-dose and air-dose cases a number of models of essentially equal applicability according to these criteria were identified. As with the hydrologic and contaminant transport models, we chose models which could be adapted most efficiently to our computing system.

2.2.2 Development of Integrated Modeling System

Work to convert all computer codes for the WISAP release consequence analysis system to a common computing language (FORTRAN) was begun in FY-77. We plan to integrate the system so that the output-input formats of interfacing codes are the same.

3. GENERIC DATA BASE DEVELOPMENT

Safety assessments for geologic isolation require data of six general types: (1) geologic and geophysical data for estimating disruptive event frequencies and disruptive process rates and durations, (2) waste form and container property data for estimating radionuclide release rates under the conditions of potential repository release scenarios, (3) geohydrologic data for estimating the origin, direction and velocity of groundwater flow for potentially disruptive scenarios, (4) geochemical data for estimating the degree to which radionuclides are retarded during groundwater transport through geologic media, (5) geographic and demographic information about sites of interest to the NWTS Program, and (6) biologic, biochemical and biophysical data concerning radionuclide transport processes in the biosphere. Data base development activities in FY-77 were concentrated on measuring radionuclide leach rates for glass waste forms and sorption properties for geologic media of interest to the NWTS Program.

3.1 WASTE FORM RELEASE RATE DATA

3.1.1 Preparation of Doped Glass Waste Forms

As a compromise between the reality of cracked glass monoliths and the powdered glass frequently studied in accelerated leach testing, we chose glass beads as the sample waste form for study. Two methods of preparing these samples were developed. A resistance heated unit was installed in a glove box for preparation of doped glasses, and an induction heated system was set up for hot cell use. Both units involve heating a comparatively large volume of glass to the melt temperature, continuously forming droplets at the end of a nozzle, and collecting the solid beads on a heated metal plate. The beads have a flattened hemispherical shape, well suited for a geometric determination of their surface area. The major value of this bead making approach lies in the ease and reproducibility of sample preparation.

Doped glass samples containing ^{99}Tc, ^{233}U, ^{237}Np, ^{239}Pu, ^{243}Am and ^{244}Cm were produced by these techniques and characterization comparisons with the undoped glass were made. The metallography, X-ray diffraction, and alpha autoradiography analyses show that the dopants were homogeneously distributed and that the doping/waste form manufacturing process does not significantly change the glass from its original form with the exception of small increases in porosity.

3.1.2 Leaching of Doped Glass Waste Forms

The leaching procedure for the doped glasses is based on the standard IAEA method and uses five different solutions: (1) salt brine based on the analysis of a deionized water dissolved core from the WIPP facility (300 g/ℓ NaCl), (2) high ionic strength calcium groundwater (2.206 g/ℓ $CaCl_2 \cdot 2H_2O$), (3) high ionic strength sodium groundwater (1.76 g/ℓ NaCl), (4) high ionic strength bicarbonate groundwater (2.52 g/ℓ $NaHCO_3$), and (5) deionized water. The experiments will be performed with both air and nitrogen atmospheres. The latter condition more closely simulates the anoxic conditions of some geologic isolation situations.

The experimental information obtained during FY-77 shows that the leach rates of all nuclide dopants are decreasing with time presumably as the resistance to diffusion of the radionuclides through the waste depleted glass matrix near the surface of the beads becomes increasingly dominant.

The high pH bicarbonate solution (pH = 9) shows higher leach rates for all nuclides than the low pH solutions. This is presumably caused by an increased dissolution rate of the glass matrix itself.

3.2 RADIONUCLIDE GEOCHEMICAL INTERACTION DATA

3.2.1 Evaluation of Sorption Measurement Methods

A number of methods can be used to measure the sorption of radionuclides by geologic media. During FY-77 we began investigations of these various methods to determine their precision, simplicity, time and cost effectiveness, and limitations. The methods studied included: (1) batch, static, shaking or jar tests, (2) low pressure, flow-through column experiments, (3) axial filtration, and (4) soil channel chromatography. Preparations were also made to study the high pressure, intact core, flow-through column method.

In the batch method a solution of known radionuclide concentration is placed in a container with geologic medium of known weight and/or surface area. The liquid phase is sampled until its radionuclide concentration, macro constituents, and pH are constant with time. The phases are then separated and counted. From this information the distribution of radionuclide between the two phases is calculated (K_d or K_a). This information can then be used together with knowledge about the bulk density, porosity, and fracture characteristics of the geologic medium in its <u>in-situ</u> state to estimate the retardation factor, K, for groundwater transport of that radionuclide (the quantity needed for the assessment model calculations) using either of the two equations below:

$$K \equiv 1 + \frac{K_d \rho}{\varepsilon} \quad \text{(unconsolidated media)} \qquad (1)$$

where:

$$K_d = \text{distribution coefficient} \left[\frac{\dfrac{\text{activity of nuclide in sorbed state}}{\text{mass of medium}}}{\dfrac{\text{activity of nuclide in dissolved state}}{\text{volume of pore space}}} \right]$$

$$\rho = \text{bulk density of medium} \left[\frac{\text{mass of medium}}{\text{volume of medium}} \right]$$

$$\varepsilon = \text{void fraction} \left[\frac{\text{volume of pore space}}{\text{volume of medium}} \right]$$

$$K \equiv 1 + K_a R_f \text{ (fractured media)} \qquad (2)$$

where:

$$K_a = \text{distribution coefficient} \left[\frac{\dfrac{\text{activity of nuclide in sorbed state}}{\text{surface area}}}{\dfrac{\text{activity of nuclide in dissolved state}}{\text{volume of fracture}}} \right]$$

$$R_f = \text{surface to volume ratio of fracture system} \left[\frac{\text{surface area}}{\text{volume of fracture}} \right]$$

Advantages of the batch method include simplicity, nominal cost, short duration, ability to perform many experiments simultaneously, ability to use crushed, chunks, shards, or rock tablets, ability to calculate K_d's on either a mass or surface area basis, ability to monitor pH, Eh, K_d time dependence, and reversibility. Disadvantages include inability to address multiple oxidation states and species, difficulty in solid-liquid phase separation, unrealistically high solution to solids ratios, disregard of hydrodynamic or flow effects, and alteration of geologic material by crushing and grinding.

In the low pressure column method a column of known length is loaded with the geologic medium of known density and porosity, and groundwater is pumped through the column at constant velocity. After steady-state is reached either a pulse or continuous injection of radionuclide solution is injected into the flowing groundwater. The experiment continues until the radionuclide discharges from the column. From the length of the column, the velocity of the water and the time for discharge of the radionuclide, the retardation factor (K) can be calculated. The distribution coefficient (K_d) is then back-calculated using Equation 1. The low pressure, flow-through column method permits the nuclide migration rates to be measured directly for porous materials without significant sample alteration. Reversibility, multiple oxidation states or species, and physical transport of colloids and fine particulates can be studied under realistic solution to solids ratios. Porous and fracture flow experiments are possible. Disadvantages include long duration experiments, inability to create practical flow rates in rock materials with low permeability, experimental phenomena such as channeling, greater difficulty in control of Eh and pH, and data reduction difficulties for non-ideal discharge curves.

The axial filtration method uses a rotating cylindrical filter which is enclosed in a coaxially-mounted pressure vessel. Solution flows into the outer chamber, through the filter into the cylindrical space within the filter, and finally out through a tube mounted on the axis of rotation. A slurry of the adsorbing geologic material is first placed in the outer (reaction) chamber and the system brought to steady-state. The radionuclide solution is then introduced into the system as either an impulse or step change in the inflow concentration and the radionuclide concentration in the outflow is recorded with time. The relative transport rates of the radionuclide with adsorbent present and absent are compared and the retardation factor is calculated. The distribution coefficient is then back-calculated using Equation 1. The principal advantages of this method are the ease and rapidity with which distribution coefficient determinations may be made and the capability to determine the reversibility of the sorption. Variable oxidation state or multiple species can be identified. Eh, pH can also be monitored and controlled. Disadvantages include the necessity of using unconsolidated geologic media and potential interpretation problems on unsymmetric data peaks.

In the soil channel chromatography method soils or ground rock material are slurried with water until moderately fluid and then applied to chemical chromatography plates using a conventional spreader. Strips of blotter paper are used as wicks for transporting the radionuclide solution to the layered soil in the channels. The radionuclide is introduced into the column chromatograph either through the feed solution or by spotting the radionuclide directly onto geologic media. The velocities of movement of both the eluting water and the nuclide are recorded. The retardation factor is calculated, and the distribution coefficient is back-calculated. Advantages of soil channel chromatography include the ability to rapidly compare the migration rate of nuclides through numerous geologic media, the ability to study several wetting and drying cycles nondestructively, and the ability to study multiple-oxidation states or species. Disadvantages include the need to use crushed materials, lack of control of flow rates, and the complexity in data reduction techniques.

3.2.2 Sorption Measurements for Generic Data Bank

During the year we made a large number of sorption measurements using the methods described previously for many nuclides and generic geologic materials. The nuclides investigated included Tc, I, Sr, Cs, Np, Am, Pu, Eu, Co, Ce, Sb, U, Nb and Ru. The geologic materials included limestone, dolomite, marble, sandstone, siltstone, shale, tuff, soapstone, anhydrite, galena, chalcopyrite, greenstone, granite, gneiss, basalt, rock salt, montmorillonite, illite, kaolinite, vermiculite, quartz, apatite, argillite, albite, anorthite, microcline, biotite, hornblende, enstatite, pyroxine, chalk and bentonite. These data are too extensive (over 300 sets of isotopes and conditions were investigated) to be reported in this summary document. We are establishing a data bank to collect and organize these data. Table 5 shows the information to be included with the data bank entries.

3.2.3 Analysis of Generic Sorption Data

Using information from the data bank about the geologic material properties, groundwater properties, and experimental conditions, it should be possible to develop empirical relationships between the measured sorption values and the independent experimental variables. Such relationships can then be used to predict sorption behavior in situations which are similar to (but not the same as) the situations investigated. We are investigating two statistical methods for accomplishing this task, nonlinear regression and adaptive learning networks.

Although our generic data base is not complete several preliminary comparisons can be made. Table 6 compares the strontium, cesium,

TABLE 5. Information Requirements for Generic Data Bank

Independent Variables			Dependent Variables	
Geologic Material Properties	**Groundwater Properties**	**Experimental Conditions**	**K_d or K_a**	**Uncertainty Limits**
Mineralogy:	pH	Solution Concentration		
Primary Minerals	Eh			
Secondary Minerals	Major Cations	Nuclide Valence State or Species Distribution		
$CaCO_3$	Major Anions			
Hydrous Oxides	Soluble Organic Composition	Contact Time		
Organic Content				
Cation-Exchange-Capacity	Soluble SiO_2	Measurement Method		
Anion-Exchange-Capacity	Soluble Minor Species			
Surface Area	Temperature			
Hydrology Properties:				
Porosity				
Water Velocity				
Percentage Saturation				
Surface Area to Volume Ratio for Fracture System				

TABLE 6. Preliminary Distribution Coefficients for Strontium, Cesium, Americium and Plutonium with Basalt and Plagioclase Feldspars Similar to Those Present in Basalt

Material	Experiment	Groundwater	pH	K_d Sr	K_d Cs	K_d Am	K_d Pu
Columbia River Basalt	Tablets (ANL)	Rock Equilibrated	-	-	-	250	150
	16-20 mesh (ANL)		-	-	-	83	88
	30-40 mesh (ANL)		-	-	-	130	120
	50-70 mesh (ANL)		-	-	-	180	130
Umtanum Basalt	Crushed (RHO)	0.003N $NaHCO_3$	8.6	-	300	-	-
			7.9	150	-	-	-
			8.8	-	-	230	20
Albite	Crushed (PNL)	0.03N $NaHCO_3$	9.4	550	530	-	-
			-	-	-	540	-
		0.03N $CaCl_2$	7.7	4	820	170	-
Anorthite	Crushed (PNL)	0.03N $NaHCO_3$	9.4	460	610	-	-
			-	-	-	3800	-
		0.03N $CaCl_2$	7.7	3	770	180	-

americium and plutonium sorption data for basalt with two plagioclase feldspars. Basalt contains labradorite, an intermediate composition plagioclase feldspar. The K_d for strontium appears to be sensitive to pH or HCO_3^- concentration perhaps due to greater sorption by carbonate precipitation. The K_d for cesium does not appear to be as sensitive as strontium to solution pH. The americium data of the three groups compares well but the ANL plutonium data is about five times larger than the RHO data. The equilibrium water composition of the ANL data has not been specified but it probably contains significantly less HCO_3^- ion which is capable of forming soluble complexes. Thus the lower K_d Pu found by RHO could reflect soluble carbonate complexation. Plutonium tracer concentration and/or speciation distribution could also be affecting the results. The pure mineral data for strontium suggest a strong effect of calcium concentration on strontium sorption.

Table 7 compares cesium sorption data from ORNL and PNL for montmorillionite with salt brine. The ORNL cesium K_d is about a factor of ten lower than the PNL data. This difference may be caused by the difference in radionuclide concentration utilized, and may support the conclusion that a few selective sorption sites can cause a large K_d for some nuclides at very low concentrations (such as those expected in geologic isolation release scenarios).

TABLE 7. Preliminary Distribution Coefficients for Cesium with Montmorillionite

Material	Experiment	Groundwater	Cesium Concentration	K_dCs
Montmorillionite	Crushed (ORNL)	4N NaCl	10^{-3} M	3-10
		0.6N NaCl	10^{-3} M	11-35
	Crushed (PNL)	5N NaCl	10^{-11} M	33
		0.03N NaCl	10^{-11} M	1000

Table 8 compares strontium, cesium, americium and plutonium sorption data for granite with a major mineral constituent, quartz. The ANL and RHO data for americium do not compare well, possibly because the RHO material had a significant amount of apatite. However, the ANL apatite data (taken under somewhat different conditions) do not support the large RHO value. Closer control of groundwater composition and Am concentration should be used to compare rocks of similar composition. The quartz and NTS granite data compare reasonably well for cesium.

Data were taken to compare the sorption of americium and plutonium for tablets and crushed materials. These measurements show higher sorption for the crushed materials and suggest that surface area can sometimes be an important parameter.

3.2.4 Validation Studies

An important consideration is the relationship between the results of short-term laboratory experiments and long-term *in-situ* behavior. We have collected thermodynamic data for elements with long-lived isotopes. Plots of nuclide activity in solution versus pH and Eh have been constructed. These data are being incorporated into a computer program to facilitate rapid calculation of nuclide activities for any groundwater composition. Future work may compare the laboratory results to the observed nuclide behavior at OKLO, NTS, and uranium ore deposits and to the waste management experience at many sites.

3.2.5 Sorption Mechanism Studies

A more complete understanding of the controlling sorption mechanisms for important nuclides in geologic environments relevant to waste isolation will increase our ability to predict radionuclide behavior at one site from data taken at another site. This in turn can result in time

TABLE 8. Preliminary Distribution Coefficients for Strontium, Cesium, Americium and Plutonium with Granitic Rocks, A Major Constituent, Quartz, and A Minor Constituent, Apatite

Material	Experiment	Groundwater	pH	K_d Sr	K_d Cs	K_d Am	K_d Pu
Granite (N.C.)	Tablets (ANL)	Rock Equilibrated	-	-	-	10	37
Granite (NTS)	Crushed (ANL)	Rock Equilibrated	-	-	-	21	2100
Quartz Monzonite Porphyry (NTS)	Crushed (RHO)	0.004N $NaHCO_3$ 0.007 $CaSO_4$	8.0	5.5	650	60,000	5400
Apatite	Crushed (ANL)	Apatite Equilibrated	-	-	-	9400	-
Quartz	Crushed (PNL)	0.03N $NaHCO_3$	9.4	350	37	130	-
		0.03N $CaCl_2$	7.7	0	34	0	-

and cost savings during subsequent site-specific investigations. We began investigations to study the effects of solution ionic strength on nuclide sorption, microstructural configuration of sorption sites on specific geologic materials, and controlled pH-Eh sorption experiments for Pu. These studies have not advanced far enough to yield significant results.

4. WORKSHOPS

Four workshops were held during FY-77 for the purpose of communicating WISAP results to and obtaining peer review from the technical community. The objectives and organizations represented at these workshops are described below.

4.1 WORKSHOP I

Date: February 23-24, 1977

Location: Battelle Pacific Northwest Laboratories, Richland, WA

Objective: Review of potentially applicable safety assessment methods and the proposed WISAP release scenario analysis method.

Organizations Represented: Battelle Pacific Northwest Laboratories, Electric Power Research Institute, Energy Research Group, Inc., Office of Waste Isolation, Science Applications, Inc., United Nuclear Industries, United States Geologic Survey, and University of Washington.

4.2 WORKSHOP II

Date: July 25-26, 1977

Location: Battelle Seattle Research Center, Seattle, WA

Objective: Review of hydrologic and contaminant transport models proposed for WISAP release consequence analysis system.

Organizations Represented: Battelle Columbus Laboratories, Battelle Pacific Northwest Laboratories, Boeing Computer Science, Inc., George Maddox & Associates, Intera Corporation, Lawrence Berkeley Laboratories, Lawrence Livermore

Laboratories, Office of Waste Isolation, Princeton University, Rockwell Hanford Operations, Rocky Mountain Arsenal, Sandia Laboratories, United States Geological Survey, University of Illinois, University of New Mexico, and University of Waterloo.

4.3 WORKSHOP III

Date: July 27-28, 1977

Location: Battelle Seattle Research Center, Seattle, WA

Objective: Review of geologic and geophysical data needs and availability for WISAP release scenario analysis method.

Organizations Represented: Battelle Pacific Northwest Laboratories, Energy Research Group, Inc., Lamont Geological Observatory, Lawrence Livermore Laboratory, Los Alamos Scientific Laboratory, Lousiana State University, Office of Waste Isolation, Ohio State University, Planetary Science Institute, Sandia Laboratories, Stanford University, TERA Corporation, United States Geological Survey, University of Illinois, and Western Washington State University.

4.4 WORKSHOP IV

Date: September 20-23, 1977

Location: Battelle Seattle Research Center, Seattle, WA

Objective: Review of FY-77 progress in obtaining radionuclide geochemical interaction data.

Organizations Represented: Adaptronics, Inc., Argonne National Laboratory, Atomic Energy of Canada, Ltd., Battelle Pacific Northwest Laboratories, Bechtel Corporation, Brookhaven National Laboratory, Georgia Institute of Technology, Lawrence Berkeley Laboratory, Lawrence Livermore Laboratory, Los Alamos Scientific Laboratory, Louisiana State University, Nuclear Assessments, Inc., NUS Corporation, Oak Ridge National Laboratory, Office of Waste Isolation, Pennsylvania State University, Rockwell Hanford Operations, Sandia Laboratories, Savannah River Laboratories, United States Geological Survey, University of California at Davis, University of Waterloo, University of Wisconsin, and Yale University.

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