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LETTER REPORT

TITLE: PROCEEDINGS OF THE NRC/ORNL WORKSHOP: RADIONUCLIDE SORPTION MODELING RELATED TO HIGH-LEVEL NUCLEAR WASTE REPOSITORY PERFORMANCE ASSESSMENT

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SUMMARY

A workshop was held to: (i) explore issues pertinent to the measurement and modeling of sorption processes in rock/groundwater systems that must be resolved to satisfy the regulatory process for high-level nuclear waste repository analysis, and (ii) help support the revision of the draft NRC Technical Position on sorption.

The conclusions of the workshop were:

* Understanding of sorption processes at candidate repository sites will be necessary for the analysis of release rates of the radionuclides which are important to predicting repository compliance with regulatory standards. Understanding will be necessary to conduct either accurate performance assessments under expected conditions, or conservative bounding release calculations for limiting conditions.

* Understanding of sorption processes requires an emphasis on

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laboratory programs to produce the information needed to support the modeling of radionuclide sorption and releases to the accessible environment. Field tests and natural analogues should be used to compliment laboratory studies. Various types of sorption models could be applied (e.g., simple chromatographic, thermodynamic, kinetic, etc.) for repository assessment. Although consideration of the various models was not a major workshop activity, an important conclusion was that laboratory data must be obtained which results in an understanding of the sorption processes regardless of the modeling approach selected.

* The experimental parameters that must be explored and quantified to achieve understanding of sorption were identified and prioritized at the workshop. The experimental programs should initially be designed to establish an understanding of sorption processes under site-specific geochemical conditions. These site-relevant measurements should then be supplemented by more detailed experimentation designed to provide an understanding of how sorption processes change when the geochemical conditions differ relative to those for the candidate repository site.

* There was a general recognition of the experimental and theoretical difficulties which may be experienced in gaining an accurate understanding of sorption processes. Uncertainty and sensitivity analyses for specific radionuclides, release scenarios, and candidate sites will be needed to help specify the required degree of understanding and to set research priorities.

1. PURPOSE AND GOALS

The workshop was planned to help support the NRC regulatory process for the candidate high-level nuclear waste repository sites through an evaluation of the technical and regulatory issues associated with modeling sorption[1] of radionuclides in repository systems. Such an evaluation is timely, because the DOE site projects are currently: (i) predicting radionuclide releases to the accessible environment that are consistent with regulatory requirements, and (ii) crediting sorption processes as an important barrier in achieving these releases (DOE 1986a, DOE 1986b). The workshop also will contribute to the development of the NRC Technical Position on sorption to be issued in final form

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[1] For the purposes of this report, the term "sorption" is used to describe any process which results in the binding of radionuclides to geologic solids. Other terms such as ion exchange, adsorption, and chemisorption are sometimes used to identify specific sorption processes. However, sorption reactions are difficult to characterize and we have chosen not to attempt to identify subclasses of sorption reactions in this report.

this fall (a draft Technical Position was issued in January 1986 for public comment). Concern over the current DOE methodology for modeling sorption provided much of the impetus for the workshop (see Section 4). Performance assessment calculations are being conducted by DOE with so-called conservative estimates of sorption values. However, these sorption values have often been estimated from empirical tests which provide insufficient understanding of the sorption processes. Without an understanding of the processes, the degree of conservatism cannot be quantified. Some of the problems or uncertainties associated with an empirical approach to the measurement of sorption data and the prediction of retardation, as well as the need for a better understanding of the sorption reactions or processes involved, have been previously expressed (Relyea et al. 1980; Barney 1981; McKinley and West 1981; Serne and Relyea 1981; NRC 1983; Katoh et al. 1984; Siegel and Erickson 1984; Tripathi et al. 1986).

2. CONCLUSIONS

The following sections summarize the consensus of the workshop participants. Selected pertinent references to previously published comments on specific items, supporting discussions, or examples of the effects of the parameters, are included. The discussion during the workshop fell into two general categories: (i) an evaluation and analysis of the role that geochemical sorption processes may play in the multiple barrier concept for high-level waste isolation, and (ii) a description and prioritization of the sorption parameters that must be quantified to accurately and defensibly model radionuclide retardation in rock/groundwater systems. There was general agreement at the workshop concerning the need to better understand sorption processes. However, unanimity did not exist with respect to the level of detail required or the prioritization of all sorption parameters. It seems probable that each radionuclide/site/release scenario analysis would have to be resolved on a case-by-case basis.

2.1 Sorption and the Performance of Repositories

Gaining a better understanding of sorption processes will be essential to extrapolation of sorption behavior with respect to changes in the chemistry of aqueous and solid phases over time and space, before modeling the future behavior of radionuclides in the rock/groundwater system of most sites will be possible. Obtaining an understanding of sorption processes to a degree adequate to support accurate or conservative predictions of radionuclide retardation will require appropriate theoretical concepts, valid mathematical models, and the supporting data required by the models. It is the responsibility of the DOE site projects to show an understanding of the sorption processes which is sufficient to satisfy the reasonable assurance aspect of regulatory standards, and additional experimentation and use of improved models by the DOE may be necessary to achieve that goal. Improvements are likely to be incremental over time. To maximize the benefit from the additional work, it will be important to recurrently evaluate and

prioritize the work needed to reduce the predictive uncertainties.

2.2 Sorption Parameters

A major accomplishment of the workshop was the compilation and prioritization of the parameters which should be measured or characterized in the DOE experimental program to develop an understanding of sorption processes. It seemed apparent to the workshop participants that a substantial body of new information would be required to achieve understanding of sorption processes, and that most of the new data would have to be developed as a result of laboratory experimentation. The following paragraphs briefly describe the geochemical parameters relevant to sorption which were identified during the workshop. The parameters are divided into two broad, and somewhat overlapping, categories: (i) those which were considered to be universally essential for modeling the sorption properties of one or more important radionuclides at one or more candidate repository sites, and (ii) those which should be evaluated because they might be essential under some situations.

2.2.1 Essential Investigations

Quantification of these parameters is considered essential for either site-specific tests or experiments designed to reach an understanding of sorption processes. For site-specific tests, the measurements must be made under test conditions that are relevant to the repository and the release scenario under evaluation. The geochemical conditions associated with some sites or release scenarios or repository aspects may be difficult to establish. This uncertainty can best be dealt with by conducting tests over ranges of parameter values. This approach will contribute to an understanding of the sorption process and will serve to identify the more important parameters. While the experimental program could initially emphasize site-specific conditions, the work must be extended to include detailed experimentation designed to establish an understanding of the sorption processes.

Solids - The solids used in the experimental program must be representative of the sorptive phase(s) in the waste package and along the release pathway(s) (Relyea et al. 1980; Barney 1981; NRC 1983). The major and minor site minerals must be characterized physically and chemically (Serne and Relyea 1981). This characterization may require identification of release pathways at the site, i.e. fracture flow, matrix diffusion, etc. (Neretnieks 1980), and physicochemical characterization of drill core and exploratory shaft samples (Relyea et al. 1980). Actual site materials or representative generic materials should be used in the sorption tests. Single mineral experiments can be used to identify specific reactive phases and help gain understanding of reactions (Palmer and Meyer 1981; Vandergraaf et al. 1984), while tests with repository rock may serve to better model site behavior. Different results may be obtained with tests using crushed vs intact rock, and these differences should be evaluated (Barney 1981). When crushed or intact rocks are used in the sorption experiments, the solid phases

exposed in these rock samples must be representative of the sorption phases along the release pathway. The surface area of the test material should be representative of the *in situ* system, or else the effects of the surface area/volume ratio of the tests should be quantified (Higgo and Rees 1986). If rocks have been pulverized for use in the experiments, understanding the effects of grinding, surface area change, and potential alteration of surface coatings on the sorption reactions will be important. Methods such as x-ray diffraction, scanning electron microscopy, as well as surface characterization analytical methods (e.g., ESCA, Auger, etc.) could be useful in understanding the surface phenomena (Relyea et al. 1980; Serne and Relyea 1981). The surface of powdered rock samples have been shown to be enriched in certain elements by factors of two or three, relative to the bulk rock composition (Soma and Seyama 1986).

Solutions - The solutions used in the site-specific tests must be representative of the altered groundwater released from the waste package to the repository site (Relyea et al. 1980; NRC 1983) and unaltered groundwater present in the far field environment which might mix with the groundwater released from the waste package. The alteration of groundwater composition due to radiolytic and thermal effects as well as the presence of waste package components in the engineered facility should be included in the test matrix (NRC 1983). Also, tests should be designed to investigate the effects of changes in groundwater composition due to geochemical reactions and mixing of groundwaters along release pathways. The possible effects of organics that could be present (McKinley and West 1981; Swanson 1983; Swanson 1984; Swanson 1985), or particulates which could form due to groundwater instability (supersaturation), waste package components, or radionuclide hydrolysis reactions should be considered (Relyea et al. 1980; Avogadro and Lanza 1982; Avogadro and de Marsily 1983). Because sorption reactions may be sensitive to the aqueous speciation or complexation of dissolved radionuclides (Siegel and Erickson 1984), the solution species released from the waste package should be identified and used in the tests, or actual solutions from waste form dissolution tests should be used. Some experiments to develop a better understanding of the sorption reactions must be performed using solutions containing only a few constituents of the groundwater. Systematic variations of the composition of these simplified solutions may yield significant insight into the sorption mechanisms occurring in the more complex systems. It also is important to determine if any solutions used for sorption experiments are oversaturated with mineral (e.g., silica or iron hydroxide) or radionuclide-containing solid phases.

Test Methodology - Both static and dynamic (closed and open) experimental methodologies must be employed (Relyea et al. 1980). Batch contact tests allow rapid screening of a number of parameters, while column chromatographic tests more accurately simulate flowing groundwater and allow evaluation of a greater variety of parameters (Relyea 1982). Other experimental methods such as staged experimentation, mixed-flow reactors (Walton et al. 1982; Theis and Kaul 1985), etc. may contribute to the interpretation of the site-

specific batch contact and column tests. A limited number of larger scale tests should also be conducted in the field to validate the predictions based on laboratory results (McKinley and Hadermann 1984). Experience has shown that sorption behavior in the field is not always similar to that measured in laboratory tests (Nyhan et al. 1985).

Conditions - For site-specific tests, the geochemical conditions during the test must be representative of the release pathway conditions and the release scenario under evaluation (Relyea et al. 1980). Thus, a range of pH values, redox conditions, pressures of CO_2, etc. should be determined and established to mimic the range of geochemical conditions along the release pathway from the waste package to the regulatory boundary of the proposed site. Since laboratory tests cannot duplicate the 10,000 year regulatory time-scale for prediction of repository performance, means of conducting accelerated tests should be explored which insure that the small-scale tests are dynamically similar to the real system. Both static and dynamic tests should be conducted to simulate repository conditions (Barney 1981; McKinley and Hadermann 1984). In general, ranges rather than single values of all conditions should be investigated to establish the sensitivity of the parameter to the experimental conditions. For experiments designed to increase the level of understanding of the sorption reactions, the conditions may need to be evaluated separately and thus may not necessarily reflect actual site conditions.

Important Release Scenarios and Radionuclides - It is obvious that all the above parameters cannot be quantified for all sites, potential release scenarios, and radionuclides in a time frame consistent with the repository development schedule. It will be necessary to judiciously identify those scenarios and radionuclides that are particularly critical to showing compliance with regulatory standards at the respective sites. The experimental programs would thus focus, at least initially, on these critial or key scenarios and radionuclides.

2.2.2 Parameters for Evaluation

The parameters included in this section are those which may, or may not, be important for some radionuclides and/or release scenarios at one or more sites and, therefore, should be initially investigated to see if quantification is necessary for defensible sorption predictions.

Competition - Competition among radionuclides and major groundwater constituents for sorption sites may be important (Serne and Relyea 1981; Katoh et al. 1984). The sorptive phases may have a limited number of active sorption sites and competition for these sites may decrease the sorption of some radionuclides. Conversely, some sorptive phases may be selective for specific radionuclides and, thus, may be unaffected by competition effects at low or trace loadings (McKinley and Hadermann 1984). Competition among radionuclides and major cation constituents of groundwater for complexing agents can also occur, as well as the formation of mixed-metal complexes among radionuclides (Grenthe et al. 1986). Sorption tests frequently are run with only one

radionuclide at a time, which does not allow evaluation of competition effects. To evaluate these phenomena, sorption tests should be run with several relevant radionuclides present in appropriate combinations and proportions (McKinley and West 1981).

Kinetics - Establishing kinetic relationships for sorption processes is important not only to addressing site behavior, but also to help understand the sorption processes and permit a better extrapolation of laboratory and field sorption measurements as a function of time. For example, detailed investigations into apparent sorption/desorption disequilibrium for some radionuclides has suggested that the presence of multiple species in batch contact tests may be the cause of the apparent disequilibrium (Higgo and Rees 1986). If actual sorption/desorption disequilibrium (hysteresis) occurs, the results from batch contact tests may be conservative, relative to the anticipated behavior in the repository system (Barney 1985). However, this conservatism cannot be demonstrated without an understanding of the processes involved. In some groundwater systems (especially fracture flow systems), the rate of sorption for some radionuclides may be slow relative to the groundwater residence time or to the rate of mass transfer of the radionuclide to the sorptive surface (Neretnieks 1980). In these cases, quantifying the kinetic relations will be essential for reliable estimation of radionuclide migration (McKinley and Hadermann 1984). Parameters which may have a critical impact on attaining local equilibrium include: seepage velocity, dispersion coefficient, distribution coefficient, sorption rate, and boundary conditions (Valocchi 1985).

Colloids/Particulates - Some radionuclides could be transported as coprecipitated components of non-radioactive colloids, sorbed onto existing colloids, or as colloidal radionuclide solids rather than as solution species, and these colloidal forms would migrate differently than solution species in the repository (Relyea et al. 1980; Avogadro and de Marsily 1983; McKinley and Hadermann 1984; Higgo and Rees 1986). Consequently, it may be necessary to consider non-equilibrium forms of radionuclides (Travis and Nuttall 1985). Both americium and plutonium, which likely will be important radionuclides for performance assessments due to the very low cumulative release limits in the EPA regulation, can be present in colloidal forms in many groundwaters (Olofsson et al. 1982; Nyhan et al. 1985; Mailen and Bell 1986). Other radionuclides may also have colloidal forms in some site groundwaters. Modeling of the transportation of colloidal particles in a porous medium is quite different from modeling solute transport (Bonano and Beyeler 1985a; Bonano and Beyeler 1985b). The nature of the interactions between the colloids and rocks is so different from solute-rock interactions that a new suite of experiments will be necessary to obtain the values of the parameters that describe phenomena such as London-van der Waals and electrokinetic effects. These effects should govern the capture of the colloidal particles by the rock (Guzy et al. 1983) and, hence, the release of radionuclides in this form.

Sorption Isotherms - The sorption value measured in tests may be dependent upon radionuclide concentration (Relyea et al. 1980).

Typically, the distribution coefficient[2] decreases at higher radionuclide concentration due to saturation of the sorption sites (McKinley and West 1981). To explore and quantify this effect, it is necessary to develop sorption data in the form of sorption isotherms (Serne and Relyea 1981; McKinley and Hadermann 1984).

Mineral Surface Coatings - Mineral surface coatings, if present, may play an important role in sorption (Jenne 1977). Characterization of the *in situ* coatings along release pathways should be conducted. In addition to identifying and characterizing these mineral surface coatings, it may be necessary to determine: (i) if the coatings remain chemically stable in the waste package-altered groundwater, (ii) the importance of these coatings as sorptive substrates relative to that for minerals which are present and have no coatings, and (iii) if the mineral surface coatings are sufficiently thick and continuous so as to isolate the host rock from the groundwater. Tests to explore the effects of surface coatings on sorption reactions may be important for quantification of the sorption to be expected at a given site (Ericksen 1985).

Thermodynamic Data - Additional and more accurate thermochemical data will be needed for certain aqueous complexes or species, and for solids containing elements of interest, to support modeling calculations and extrapolation of laboratory data to different geochemical conditions (Relyea et al. 1980; Serne and Relyea 1981). As site-specific tests are conducted, the needed data will become apparent. For example, the stability constant for a particular complex or a solubility product for a solid phase may be needed to elucidate the nature of a sorption reaction. As these needs become apparent, the data should be obtained.

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[2] The distribution coefficient, K_D, is defined as the ratio of the concentration of the sorbed element on the solid divided by the concentration in solution. The units are L/kg.

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4. BACKGROUND INFORMATION ON SORPTION AND DOE ANALYSIS OF CANDIDATE REPOSITORY SITES

Much of the impetus for the workshop, and the reason for many of the conclusions discussed earlier, was concern over the current DOE methodology for predicting sorption processes in performance assessments. DOE's use of a simplified ion exchange model (sometimes called the K_D model) to describe radionuclide migration in a repository seems inappropriate for many situations. The underlying theoretical concept, the data measurement methodology, and the mathematical modeling employed in the K_D approach are all items of concern (Tripathi et al. 1986). This section summarizes these concerns.

A high-level waste repository site will consist of several km^3 of rock surrounding the emplaced waste. In addition to physical isolation from the environment, release pathways through this large volume and mass of rock will, for some sites, also provide an extensive sorptive medium for retardation of solubilized radionuclides. Sorption is specifically cited in NRC regulations only in 10 CFR Part 60.122 as a favorable condition for siting criteria. However, most scientists have assumed that sorption by the repository host rock would play an important role in achieving site performance which is in compliance with the cumulative radionuclide release limits to the accessible environment specified in the EPA regulation 40 CFR Part 191. For many radionuclides, even poor to moderate sorption could result in retention of the radionuclides for times longer than those required for radioactive decay to reduce the concentration and release of radionuclides to acceptable levels.

The Environmental Assessment reports (EA's) issued by the DOE for the Yucca Mountain (DOE 1986b) and the Hanford Site (DOE 1986a) candidate repositories include an analysis of the retardation of solubilized radionuclides in groundwater by sorption onto the tuff at Yucca Mountain and the basalt at the Hanford Site. Little emphasis was given to sorption processes in the EA describing the candidate salt site (DOE 1986c). In the non-salt cases, the mobility of many radionuclides was calculated to be retarded by orders-of-magnitude, relative to the groundwater, as a result of sorption reactions involving the host rock. These predictions were based on application of the idealized chromatographic relationship (so-called K_D model) developed for saturated, porous ion exchange media

$$R_F = 1 + K_D(d/p) \ ,$$

where R_F is the retardation factor, d is the rock bulk density, p is the porosity of the rock, and K_D is an experimentally measured equilibrium distribution coefficient. This modeling approach may prove acceptable for some radionuclides at one or more sites. However, predictions of the mobility of some radionuclides in some geologic systems may prove to be inaccurate and non-conservative because sorption in some rock/groundwater systems may be poorly represented by this chromatographic model.

The following are important uncertainties in the application of K_D model to geologic systems (references are included to some similar statements of concern published by others).

(1) The radionuclide-transporting medium (groundwater) may not contact all of the reactive volume of the stationary sorbent phase (rock) due to: (i) the prevalence of fracture-flow hydrologic conditions, and/or (ii) slow diffusion rates of radionuclides into host rocks (Neretnieks 1980; McKinley and Hadermann 1984; Hadermann and Roesel 1985).

(2) Under dynamic conditions, the processes controlling sorption may not allow the latter to reach local equilibrium and, as a result, predictions of radionuclide releases made using a batch-measured K_D value may lead to overestimation of release time (Valocchi 1985) and underestimation of the amount released (Starr et al. 1985).

(3) The measured K_D value may not be independent of the radionuclide concentration and thus cannot be accurately modeled by a single value (Barney 1981; Serne and Relyea 1981; McKinley and Hadermann 1984).

(4) Radionuclides may undergo chemical reactions during migration, be present as multiple species, and competition between radionuclides or groundwater constituents may occur for the sorption sites (Serne and Relyea 1981; Siegel and Erickson 1984; Katoh et al. 1984; Higgo and Rees 1986). Such effects are not accounted for by the K_D approach.

(5) Unsaturated hydrologic systems, such as the Yucca Mountain repository formation, are difficult to properly model since the degree of contact between the groundwater and the sorptive phases may be unknown.

(6) Migration of colloidal radionuclides are difficult to model (Avogadro and de Marsily 1983; Travis and Nuttall 1985; Higgo and Rees 1986) and cannot be described by a K_D approach.

Many of the problems or uncertainties associated with the retardation predictions in the EA's stem from the experimental design chosen by the DOE laboratories for the sorption tests; for example, sorption data obtained using pulverized unaltered site rock that is not indicative of the solid phases that exist along the release pathway. The empirical test methodology used may be characterized as "process blind," and it provides little insight into the sorption reactions. Although significant amounts of empirical sorption data exist, the data have not been assembled to contribute to understanding and may not afford reasonable assurance that conservative values have been selected for modeling calculations.

Care will have to be exercised by the DOE to demonstrate that the uncertainties and limitations in the data and the K_D modeling methodology will preclude non-conservative predictions. In some cases, it seems possible that the K_D modeling approach may not even be adequate for estimation of bounding radionuclide retardation values under limiting conditions. As discussed earlier, the consensus of the workshop was that greater understanding of sorption processes will be necessary to support performance assessment modeling for some important radionuclides. This understanding will be important whether DOE attempts to calculate bounding values (the values must be demonstrably conservative and bounding) or accurate values using more comprehensive hydrogeochemical models.

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