Determination of Radionuclide Sorption for High-Level Nuclear Waste Repositories

Technical Position
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DETERMINATION OF RADIONUCLIDE SORPTION FOR HIGH-LEVEL NUCLEAR WASTE REPOSITORY NRC TECHNICAL POSITION

1.0 INTRODUCTION

1.1 Purpose

The purpose of this Generic Technical Position (GTP) is to provide guidance to the Department of Energy (DOE) concerning the general information that the Nuclear Regulatory Commission (NRC) considers will be needed in order to assess the effects of radionuclide sorption upon performance of a high-level waste repository. Sections one and two of this document contain summaries of applicable regulations, definitions of significant terms and a discussion of the role of sorption in influencing repository performance. Section three contains the NRC position on sorption information needs, and section four contains the rationale behind each information need. This document does not prescribe specific methods for collecting the information. Instead, it is intended to provide general guidance to DOE that may prove useful in their preparation of detailed plans for gathering radionuclide sorption information.

1.2 Regulatory Framework

The Nuclear Waste Policy Act of 1982 (P. L. 97-425) defines the role of three Federal agencies in the national program for disposal of high-level radioactive wastes. The Environmental Protection Agency (EPA) has been responsible for developing "generally applicable standards for protection of the general environment from offsite releases from radioactive materials in repositories." These standards have now been issued as final regulations (50 FR 38066). The NRC develops and issues "...technical requirements and criteria that it will apply in approving or disapproving (i) applications for authorization to construct repositories; (ii) applications for licenses to receive and possess spent nuclear fuel and high-level radioactive waste in such repositories; and (iii) applications for authorization for closure and decommissioning such

repositories." In this way, it will implement the EPA standards. The Department of Energy (DOE) is responsible for collecting the data needed for site characterization and for constructing and operating a waste disposal facility in accordance with NRC regulations. Sections of the NRC Regulations, 10 CFR Part 60, to which information on radionuclide sorption may apply are the overall system performance objective for the geologic repository after permanent closure (60.112), the performance of particular barriers after closure (60.113), and the siting criteria (60.122).

1.3 Definition of Sorption

Sorption - one or more physicochemical processes, including ion exchange, adsorption, and chemisorption, but excluding precipitation of stoichiometric (fixed radionuclide composition) solid phases, in which the radionuclide is removed from a liquid phase by interaction with a solid phase or phases.

2.0 BACKGROUND

A geologic repository controls the rate of radionuclide release to the accessible environment by means of two major subsystems: (1) the geologic setting of the controlled area; and (2) the engineered barrier system. The geologic setting (site) is selected for its geologic, hydrologic, and geochemical attributes that enhance radionuclide isolation.

In order to compensate for the uncertainty in predicting the behavior of geologic systems over long periods of time, the Congress has mandated a multibarrier approach to ensure isolation of radionuclides. The NRC has adopted this approach in its licensing criteria. In general, this approach puts emphasis on: (1) engineered containment of radionuclides during the period of peak fission product decay, and (2) assurance of a low controlled release thereafter. During the period of engineered containment of the waste, the site

geology should provide sufficient backup for radionuclide isolation in case engineered containment is lost. Thereafter, the site geology should also have the capacity to limit the release of the long-lived radionuclides to the accessible environment so that 10 CFR 60.112, the overall system release performance objective (EPA standard), is met. The DOE endorses the multiple barrier approach and places primary importance on the capabilities of the natural system for waste isolation (DOE, 1984).

Radionuclide sorption parameters can be used in performance assessment models for characterizing the performance of a high-level nuclear waste site pursuant to 10 CFR 60.112. These parameters are difficult to determine precisely because future geochemical conditions cannot be known with complete certainty and laboratory tests may not accurately model site behavior. However, by experimentally investigating sorption parameters using site-specific phases and conditions, it should be possible to take account of many sources of uncertainty and make reasonable estimates of sorption along radionuclide release pathways.

2.1 Use of Sorption in Performance Assessment Analysis

When a liquid is flowing through permeable solid media, sorption retards the migration of the solute relative to the liquid flow. Some radionuclides require some retardation in the host rock to meet regulatory criteria. Experimentation can be used to estimate this retardation and, thus, quantify two aspects of repository performance. First, sorption experiments can be used to help identify which radionuclides can be sorbed on repository solids. Second, sorption studies can also be used to determine the ability of the subsurface repository (the engineered system and the geologic setting out to the accessible environment) to slow the migration of the radionuclides to the accessible environment. For example, parameters such as sorption or desorption

ratios[1], sorption capacities[2], and retardation factors[3] derived from these studies can be used to help quantify the ability of the subsurface repository to retard radionuclide migration.

2.2 Types of Sorption Experiments

In general, sorption experiments can be subdivided into two types: 1) closed systems; and 2) open systems (NEA Sorption Workshop, 1983; McKinley and Hadermann, 1984). Both approaches have been used to approximate one aspect of repository performance. For characterizing sorption phenomena, closed-system experiments, such as batch sorption tests, involve contacting radionuclide-free (or deficient) solids with a radionuclide-bearing solution for the duration of the experiment followed by analytical determination of the sorption ratio, R_s. Batch desorption experiments, on the other hand, involve contacting radionuclide-free (or deficient) liquid with radionuclide-bearing solids, followed by measurement of the quantity of radionuclide leached. Open-system experiments, such as flow-through column tests, involve the introduction of liquid solution at one end of a reaction vessel containing solid and the removal of the liquid at the other end. The solid material sorbs the radionuclide(s) and, as a result, retards the migration of the contaminant(s) relative to that of the liquid. A retardation factor, R_f, is determined in the open system experiments.

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1 Sorption or Desorption Ratio, R_s - the ratio of the concentration of radionuclide on or within the solid to that in the liquid (units are L/Kg).

2 Sorption Capacity - the maximum amount of radionuclide that can be sorbed on a unit mass of solid for a given set of conditions.

3 Retardation Factor, R_f - the ratio of the velocity of the liquid to that of the radionuclide in an open (flowing) system.

3.0 STATEMENT OF POSITION

It is the position of the NRC staff that the following information is essential to any assessment of the effects of radionuclide sorption upon the performance of a high-level waste repository.

1) Description of a testing program that details information to be gathered for evaluating sorption models and considers the range of proportions and compositions of phases under the various physicochemical conditions in the subsurface repository;

2) Characterization of solid and liquid experimental starting materials and products;

3) Determination of sorption and desorption isotherms for closed-system experiments by varying radionuclide concentrations up to an apparent concentration limit if appropriate;

4) Evidence for or demonstration of the applicability of sorption parameters to repository performance showing consistency of results among various approaches such as open and closed laboratory experiments, in situ field tests, and studies of natural analogues; and

5) Documentation of the magnitudes of experimental and conceptual uncertainties from all anticipated sources.

The rationale for these information needs is presented in section four.

4.0 DISCUSSION

It is the responsibility of the DOE to demonstrate that when it proposes to take credit for sorption, the sorption parameters used in performance assessment calculations do not underestimate radionuclide migration.

4.1 Testing Program

The purpose of a testing program should be to demonstrate that theoretical sorption models used to explain sorption in the repository are appropriate and crucial experiments that characterize sorption will not be overlooked. The testing program should be flexible enough to allow for changes in prioritizing experimentation as evidence is compiled from early studies.

The testing program should evaluate variables such as solid composition, mineralogy, and texture, liquid composition, proportion of phases, temperature, pressure, particle size, specific surface area, flow rate and regime (porous and fractured media), effective porosity, fracture distribution and geometry, time, and ionizing radiation, along with any other parameter shown in early experimentation to influence sorption. This information should pertain to the portions of the repository system in which the DOE will take credit for sorption processes in performance assessment. These parameters can be considered in a performance allocation strategy describing the kind and amount of testing necessary to determine their role in affecting sorption. The DOE can then effectively demonstrate its rationale for choosing some combinations of parameters for study and eliminating other combinations as inappropriate.

4.2 Characterization of Experimental Starting Materials and Products

In order to demonstrate the quality of the sorption data, the DOE will need information on the solids most likely to react with groundwater and thereby affect radionuclide concentrations and release rates. Thus, identification and

characterization of waste form, canister, backfill, seals, packing, and host rock primary and secondary phases occurring along paths the radionuclide-bearing groundwater will take as it flows away from the waste, is important to show that appropriate experimental starting materials have been used in the sorption studies. Characterization of the solids should include chemical, mineralogical, textural, particle size and surface area determinations, and any other aspects found to be important as indicated by early studies. Also, information will be needed on the effects of surface area of the solids/volume of liquid ratio (Higgo and Rees, 1986) and the stability of the minerals that can affect repository performance. For example, sorbing minerals such as clays and zeolites can decompose at repository temperatures to less sorptive minerals (Bish et al., 1982). If crushed solids are used, it is essential to show that laboratory experiments involving sorption on crushed solids (for example) are relevant to the repository site. The surfaces of crushed material may be significantly different from the surfaces of intact material, both porous and fractured. Grinding may expose the surfaces of solid phases different from those which groundwater would contact in a repository and/or may change the reactivity of the same mineral surfaces with dissolved radionuclides. The surface of crushed minerals can be enriched in certain elements by factors of two and three relative to the bulk rock composition (Soma and Seyama, 1986). Surface analytical techniques could be important for characterizing the solids.

Similarly, the range of groundwater compositions expected in a repository system should be considered in selecting the liquid starting materials. Generally, in the rock-dominated environments of a high-level waste repository, groundwater compositions can be affected by reactions with solids at various temperatures and pressures. Consideration of the range in water compositions used in experimentation should be based on the range of compositions (both inorganic and organic) of analyzed groundwaters at ambient conditions, the range of compositions calculated from solid assemblages assumed to have equilibrated with the groundwater, and the range of groundwater compositions

experimentally determined at the temperatures and pressures applicable to post-emplacement. The groundwater compositions chosen for sorption work will be applicable to the repository regions in which credit for sorption will be taken.

The applicability of synthetic starting materials to the conceptual model employed in developing the testing program should be considered. Failure to address the applicability of these materials to those of the repository might result in experiments that do not adequately simulate repository conditions. For example, the preparation of radionuclide-bearing groundwater commonly involves the addition of a small amount of acidified tracer to a synthetic solution simulating the natural groundwater. The resulting solution may be neither representative of solutions under repository conditions nor be stable.

In addition to information on the starting materials, it is also important to characterize the experimental products. Following the experiment, analysis of the liquid products should include the determination of major, minor, and trace element concentrations, along with pH and redox conditions (for example, Sanchez et al. (1985) show that the concentration of ligands and pH have a strong influence on the amount of plutonium sorbed onto goethite).

The extent of sorption of some dissolved radionuclides on engineered barrier materials and host rock can be strongly dependent on the redox potential (Eh) and acidity (pH) of the groundwater. For example, Benjamin and Leckie (1981) show that the sorption of Cd, Cu, Zn, and Pb on amorphous iron oxyhydroxide is strongly dependent on pH. The percentage of cation sorbed varies from approximately zero to one hundred with a change in pH of two units. Likewise, Kelmers et al. (1984) have shown that sorption ratios for neptunium and technetium are dependent on the redox condition of the system.

The characterization of solid products from sorption experiments is important to the extent feasible because, under the same physicochemical conditions,

different solid phases can have drastically different sorptive capacities for the same radionuclide. Characterization of the solids, although extremely difficult, is important in determining which reactions took place and how these reactions depend on experimental technique. In addition to determination of the composition of individual solid phases, characterization should include surface area and/or particle size measurements. Because sorption is predominantly a surface phenomenon, the surface area of the solid may strongly affect the experimentally determined sorption parameters. For example, neptunium sorption ratios increased two orders of magnitude as particle diameter decreased from 200 to 2 μm (Kelmers et al., 1984).

Radionuclides can exist in colloidal and particulate forms in aqueous systems in both the laboratory and the field. In these forms, radionuclides can migrate differently than when the radionuclides exist as dissolved species in the liquid (Relyea et al. 1980; Avogadro and deMarsily, 1983; McKinley and Hadermann, 1984). For example, due to their size, colloids may not pass through the small pores of rock matrix but be confined instead to larger fissures (EA, Yucca Mountain, 1986). Thus, consideration and analysis should be given to colloid and particulate formation and how these forms may affect repository performance.

4.3 Isotherm Development for Closed-System Experimentation

Because radionuclide concentrations are expected to vary in the repository and sorption parameters can be concentration-dependent, it is reasonable to design experiments to determine the effect of radionuclide concentration on sorption ratios. Sorption isotherms should be determined for the various geochemical conditions expected in the repository. The concentrations of radionuclides in the repository can range from zero to an apparent concentration limit under equilibrium conditions. The apparent concentration limit is the greatest radionuclide concentration that the liquid can maintain at equilibrium when the temperature, pressure, and moles of all other components in the liquid, n_j, are

held constant. Supersaturated conditions can occur if precipitation is kinetically unfavorable. The apparent concentration limit is controlled by the solubility (NRC, 1984) or precipitation kinetics of some stoichiometric (with respect to the radionuclide) solid phase. Figure 1, a schematic sorption isotherm, illustrates the relationship between concentration on the solid versus concentration in the liquid when all other parameters are held constant. Analysis of the liquid product can be used to monitor the constancy of the other parameters. Although this figure shows a linear sorption region, many sorbed species, including radionuclides, can show nonlinear relationships between the quantity sorbed and the solution concentration. Thus, sorption ratios can be dependent on radionuclide concentration (Serne and Relyea, 1982; Relyea et al. 1980).

Sorption isotherms should be determined up to an apparent concentration limit if possible, so that it can be shown that precipitation is not contributing to the sorption ratio. If precipitation instead of sorption has occurred in a single sorption test, retardation is not proven. In such a case, the concentration of radionuclide in the liquid would be limited by the solubility of the radionuclide-bearing solid, but insensitive to the presence of the other solids in the substrate. For example, if precipitation occurred in a batch test containing a nonsorptive solid, an 'apparent sorption ratio' could be determined. This 'apparent sorption ratio' could be erroneously used to calculate a retardation factor (see Section 4.4). However, if the liquid from this batch test is then decanted into a column containing the same nonsorptive solid, the concentration would be below the solubility limit (i.e., no additional precipitation would occur) and the radionuclide would travel down the column as fast as the liquid (no retardation). Thus, if precipitation is not disproved in a sorption test, credit cannot be taken for retardation of the radionuclide. Experimentally, it should be possible to determine an apparent concentration limit of a radionuclide in liquid in contact with solid. For example, at the same temperature, pressure, and n_j in the liquid, two sorption experiments with different concentrations of the same radionuclide in the

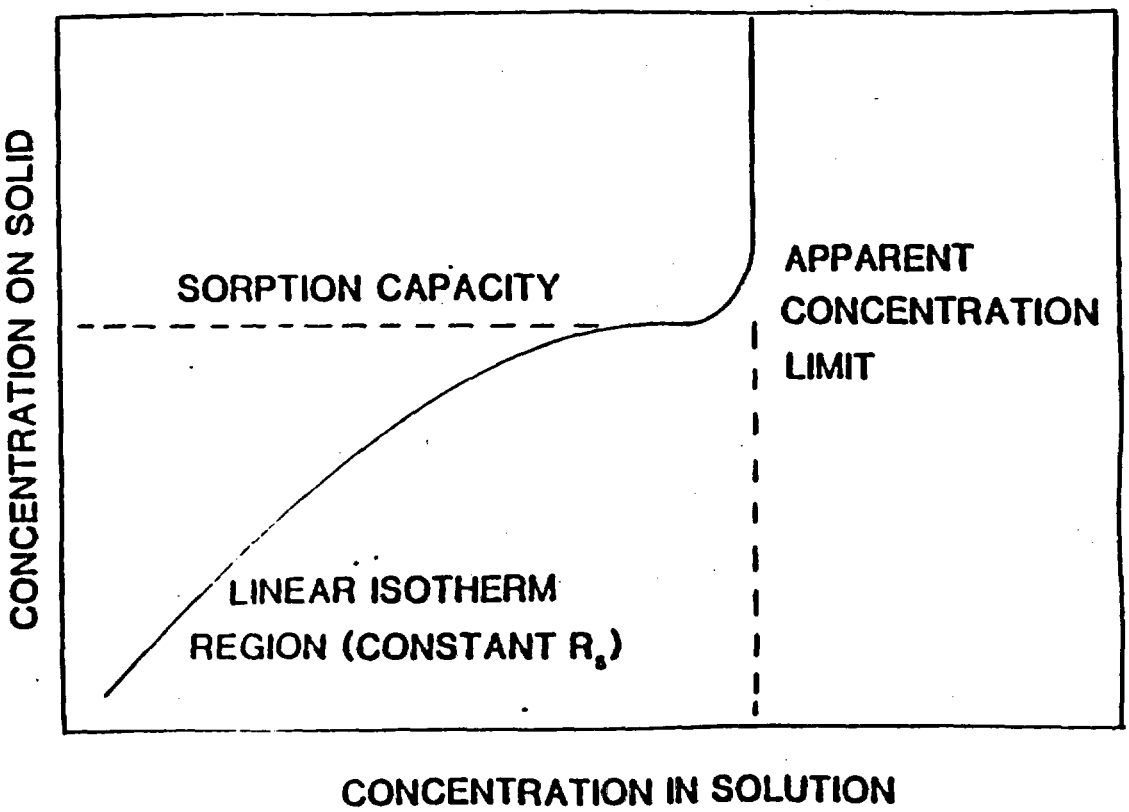

Figure 1. Schematic Sorption Isotherm.

liquid starting material should yield the same radionuclide concentration in the liquid products at the apparent concentration limit (see Figure 1).

In addition to sorption isotherms, the DOE will need desorption isotherms to adequately evaluate performance assessment. Although sorption parameters are normally less than desorption parameters and, thus, overestimate the radionuclide concentration in the groundwater under equilibrium conditions, radionulide migration scenarios can be imagined in which the lack of consideration of desorption would underestimate the migration of a radionuclide. For example, given two radionuclides with different sorption ratios, the radionuclide with the smaller ratio would travel faster than the other radionuclide and the peaks in radionuclide concentration would separate as they travelled toward the accessible environment. No sorption-site competition would occur. If, however, the radionuclide with the smaller sorption ratio undergoes slow desorption, it is possible that the two concentration peaks might coincide and competition for the sites could occur. The result of this site competition could be to accelerate the migration rate of the radionuclide with the larger sorption ratio.

4.4 Determination of Sorption Parameters by Multiple Approaches

If a sorption experiment could be designed that simulated all anticipated repository conditions, it would not be necessary to use multiple experimental approaches to determine sorption parameters. However, simulation of <u>all</u> anticipated repository conditions in sorption experimentation would be difficult and/or impractical. Some parameters or conditions cannot be simulated. Thus, extrapolation of these conditions to those expected in the repository is required. This extrapolation introduces uncertainty into the modeling of sorption parameters. Therefore, multiple approaches are important because they can lend support to, and reduce the uncertainties of, experimental results from studies in which some parameters are not site-specific. Some experimental parameters can be varied over a large enough range as to bound the

conditions anticipated in the repository. These parameters include surface area/volume ratio (SA/V), temperature, pressure, composition, and flow rate. Other parameters that often are not duplicated in the laboratory are scale, residence time, water/rock ratio, and flow characteristics, which can include saturated versus unsaturated flow and porous media versus fractured flow.

Experiments are designed so that measurable effects of physicochemical reactions can be monitored in a reasonable time. Section 60.101(a)(2) of 10 CFR Part 60 allows for the use of accelerated tests to demonstrate compliance with performance objectives and design criteria. At the relatively low temperatures anticipated in the repository, chemical reactions involving geologic materials can be extremely slow. In order to accelerate these reactions so that changes are measureable in experimental time, conditions other than those anticipated in the nuclear waste repository are sometimes imposed on the experimental system. For example, experiments have employed crushed solid material, high concentrations of solutes, agitation, catalysts, rapid flow rates, and elevated temperatures. In order to evaluate the appropriateness of experiments to repository conditions the effect of acceleration techniques should be determined.

In addition to accelerating reaction rates, laboratory experiments are designed so that the amount of material required can be handled reasonably. By scaling down systems of interest (repository size) to laboratory size, certain physical conditions must be altered. For example, the water/rock ratio in most repository systems is significantly less than one. However, in order to obtain enough water for analysis in laboratory experiments, the W/R ratio is ordinarily increased significantly. This technique makes the bulk chemistry of the experimental system different from that in the repository. The proportions of phases in experiments has been shown to affect radionuclide sorption parameters (Palmer et al., 1981; Rafferty et al., 1981; Meier et al., 1982). Thus, the effect of this technique on sorption parameters should be considered and analyzed. One can argue that in a fractured medium, with little porosity,

most of the rock will not be in contact with the groundwater. Consequently, water/rock ratios used in experimentation should be higher than those that take into account all the rock in a repository system. If this argument is used, however, it follows that the solid reactants should be predominantly fracture material and not bulk rock. Sorption experiments involving crushed bulk rock might have little applicability to sorption phenomena in fractured media.

One way to verify the applicability of experimentally determined sorption parameters to a repository system is to use multiple experimental approaches. [This approach was a recommendation of the Waste/Rock Interactions Technology (WRIT) Program (Serne and Relyea, 1982).] Using this approach, sorption parameters can be analyzed and compared. For example, the sorption ratio, R_s, obtained from batch experiments has often been used to calculate a retardation factor, R_f. The relationship between R_s and R_f is taken to be

$$R_f = 1 + \rho R_s(1 - \phi_e)/\phi_e$$

where ρ is the bulk density of the rock, and ϕ_e is the effective porosity. This relationship is based on equilibrium ion exchange theory as applied to porous media flow. However, due to the variety of processes that contribute to sorption, and a lack of equilibrium in either or both of the batch or flow-through experiments, the calculated R_f value may not equal the measured R_f value determined from flow-through column experiment. In such a situation further testing would be required to explain the discrepancy.

Comparison of the sorption and desorption parameters obtained from closed-system and open-system experiments is recommended. Generally, the sorption parameters (e.g., R_s) derived from closed-system experiments are equal to or greater than those derived from open-system tests using the same solid material (NEA Sorption Workshop, 1983). As a result, closed-system tests may

overestimate the effectiveness of a repository system to isolate radionuclides (Relyea et al., 1980), possibly due to particle abrasion in stirred closed-system experiments or the open-system experiments may underestimate the effectiveness of the repository due to the relatively short residence times (NEA Sorption Workshop, 1983). Other factors that can cause a discrepancy between the sorption parameters from open and closed systems are the presence in the liquid of multiple radionuclide species, colloids, and particulates. Varying physicochemical parameters such as temperature, fluid velocity, radionuclide concentration, time of reaction, and fluid composition may shed some light on the causes of the discrepancy between the two types of systems. Thus, a rationale for the difference in sorption parameters observed using different methods contributes to the overall certainty that can be assigned to the sorption parameters.

To demonstrate the appropriateness of experimental results in performance assessment calculations information on the flow characteristics expected in the repository will be needed. The flow characteristics of the groundwater can have a drastic effect on the applicability of laboratory-derived sorption parameters to repository performance. Most experiments use crushed material as a solid medium because it is easy to handle and characterize, and accelerates heterogeneous reactions. The application of crushed material to intact porous media may be adequate but not so when the natural system is fractured rock. Sinnock et al. (1984) and Nuttall and Ray (1981) have calculated that rates of radionuclide migration via fracture flow can be two orders of magnitude greater than that via porous media flow. Thus, for performance assessment calculations, consideration and analysis of flow regime can be of the utmost importance.

If groundwater flux in a repository is predominantly via fracture flow, sorption tests in the laboratory may not adequately simulate repository conditions. One method of further reducing the uncertainty caused by the inadequate simulation of various flow characteristics could be to perform in

situ tests on site-specific solid material (Serne and Relyea, 1982; Abelin et al., 1984; Neretnieks et al., 1982). The scale of these tests can be larger than that of the experimental tests but smaller than that of the repository. Furthermore, the in situ solid materials would certainly not have suffered the effects of handling (grinding, sieving, washing) required in laboratory tests. Time constraints, however, would still apply in these experiments. Comparison between the laboratory and field results can illustrate the usefulness of the different approaches, but the physicochemical conditions must be carefully monitored in the in situ tests to ensure a parallelism in the approaches. Therefore, in situ tests in conjunction with laboratory tests can be performed to reduce the uncertainties of extrapolation to the repository systems.

Although field tests can expand spatial scale over that which is normally handled in a laboratory, the time scale is still several orders of magnitude less than that of a repository. The study of natural analogues is a means of shedding some light on the migration of radionuclides in natural systems that have existed for long periods of time. Thus, their use is a way of dealing with the uncertainty associated with extrapolating short-term laboratory and field experiments to long-term performance of a repository. Common examples of systems used as natural analogues are ore bodies such as Oklo (Brookins, 1978) and the uranium deposits in the Northern Territory of Australia (Airey, 1983). Igneous intrusives have also been studied because they simulate anticipated thermal histories and alteration patterns. To be most useful, however, the natural analogue should include a process that is demonstrably equivalent to the same process present in the repository and have well-defined boundary conditions.

4.5 Documentation of Uncertainties

There are many sources of uncertainty in sorption studies. For example, sources of uncertainty stem from failure to duplicate anticipated repository conditions, improper extrapolation of experimental results to repository

conditions, incorrect experimental results, and analytical error. The failure to duplicate repository conditions can be caused by an incorrect understanding of the conditions, an inability to duplicate the conditions or an inadvertent improper experimental design. Likewise, improper extrapolation can also originate from improper experimental design. Incorrect experimental results can be caused by imprecision or misinterpretations of the data. Ways that uncertainties can be minimized are (1) the use of multiple techniques to determine repository conditions, (2) analyses to bound adverse impacts, (3) multiple experimental methods, and (4) the independent duplication of results. The characterization of uncertainties is important for determining the level of confidence placed on sorption parameters. The characterization of uncertainties is an essential element for the assessment of the effects of radionuclide sorption upon performance of a HLW repository.

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