RECENT DEVELOPMENTS IN THE INTEGRATED APPROACH TOWARD CHARACTERIZATION OF RADIONUCLIDE TRANSPORT, YUCCA MOUNTAIN, NEVADA

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ABSTRACT

The radionuclide migration program for the Yucca Mountain Site Characterization Project (YMP) includes studies of radionuclide solubility, sorption, diffusion, and transport. The study plans incorporate all possible parameters of investigation; decision-making strategies for prioritizing the parameters and evaluating their significance were developed in conjunction with the study plans. After definition of explicit research goals for each study, YMP evaluated the applicability of existing data and formulated experimental approaches for obtaining additional data. This resulted in development of individual testing strategies that were integrated into an overall strategy for the radionuclide migration program designed to provide input to credible performance assessments. The strategies allow for decision points at various steps of data collection and testing. They provide a streamlined process toward a defensible level of understanding of chemical retardation and transport processes that will be used to predict the mountain's ability to isolate waste.

INTRODUCTION

A major effort of site characterization activities at Yucca Mountain, Nevada, a site being studied by the U.S. Department of Energy (DOE) as a potential repository of high-level radioactive waste, is directed toward understanding the efficacy of the natural barrier to isolate waste. Characterization of potential aqueous radionuclide migration to the far field is a key step in that determination. At Los Alamos National Laboratory (Los Alamos), the integrated radionuclide migration program combines investigations to characterize the radionuclide transport processes, including solubility, speciation, sorption, diffusion, and dispersion, and studies to investigate components of the natural system affecting those processes, such as mineralogy, mineral stability and water chemistry (Fig. 1). Site models developed to support the determination of the natural barrier's efficacy must then support performance assessment modeling.

This paper focuses on the development of testing strategies for investigating the transport processes. Validation testing is an important component of the strategies and occurs at all levels of the program, from data collection to site and performance assessment modeling. The testing strategies are based on the philosophy that simplifications of the transport processes made for performance assessment models can be valid only if more detailed site models show that simplifications do not change results of the models.

BACKGROUND

Early in the program (from the late 1970's through 1987) an empirical approach was taken toward collection of geochemical data. This provided a distribution of values but could not be used to obtain mechanistic information and, without an extensive sampling program, did not provide confidence in the spatial distribution of values. A more mechanistic approach was proposed in 1988 (1) which would have potentially required as large a commitment of time and resources for an experimental program as would have the empirical approach. An approach was needed which would take advantage of the past empirical work and could also be used confidently in transport models, even in the face of incomplete site characterization data. Impetus for formulating a testable approach was provided when the Nuclear Waste Technical Review Board (NWTRB) recommended that YMP hold a workshop to address questions of radionuclide adsorption, especially the applicability of available data, the need for additional data, and the validity of such data in performance assessment models (2). As a result of the workshop, which was held at Los Alamos, NM in September, 1990, YMP sought to provide a simple yet scientific approach to answering questions of sorption. The plan that Los Alamos developed for YMP is referred to as the minimum K_d strategy.

The minimum K_d strategy takes the approach that radionuclides will be in contact with major minerals present in both matrix and fractures, regardless of flow path. A minimum K_d results from sorption of a radionuclide on a major mineral or rock with the lowest sorptive capacity in the most active water. The experimental steps used in answering questions along the strategy path focus on batch sorption experiments using the most active groundwater composition reasonably expected at Yucca Mountain. Ongoing pure mineral batch sorption experiments will obtain K_ds for minimum sorbing but major minerals for each rock unit, e.g. quartz and feldspar. The minimum K_d strategy is described in detail in Meijer (3).

Development of Testing Strategies for the Radionuclide Transport Program

From experience with the sorption studies, YMP recognized the need to develop other testing strategies and to integrate individual strategies into an overall defensible approach for the radionuclide migration program. The first step to developing individual testing strategies and an integrated radionuclide migration program strategy entailed evaluation of the DOE's Performance Assessment Strategy Plan (4). This plan describes a three-tiered approach to model development (Fig. 2).

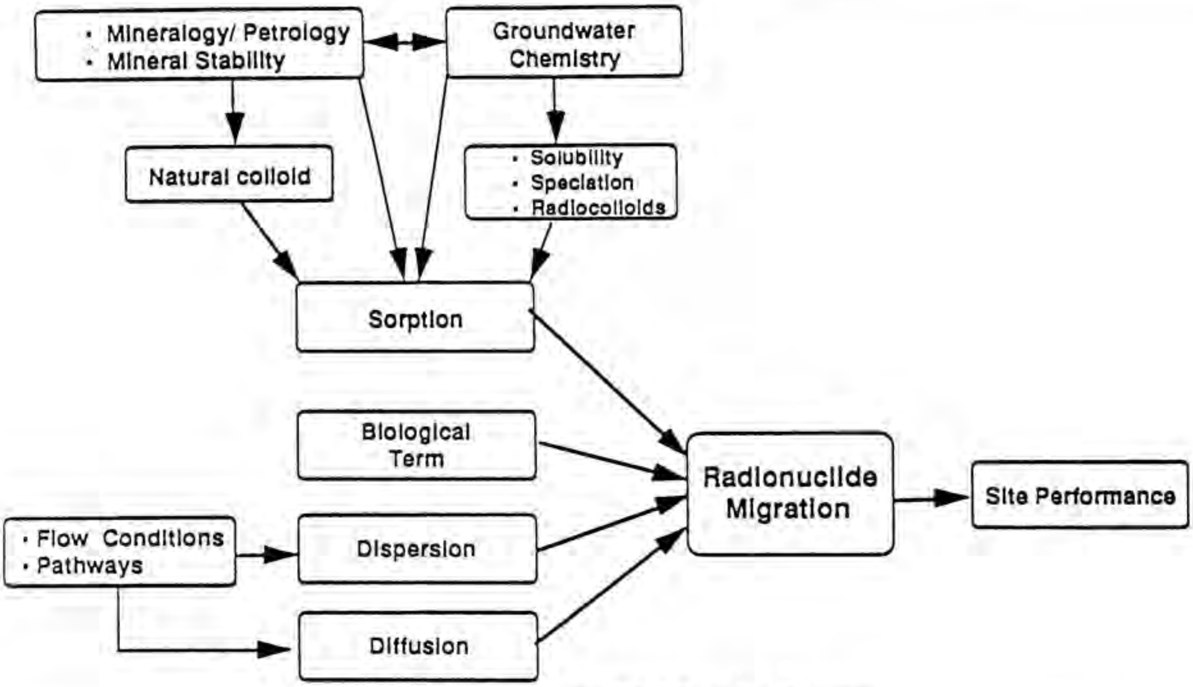

Fig. 1. Los Alamos Radionuclide Migration Program.

The foundation of the pyramid consists of multiple process-level models that are developed through site characterization studies for YMP. Examples of process-level models are conceptual models for sorption, solubility, and transport and their embodiment in detailed YMP computational codes such as TRACRN (5), which is based on TRACR3D (6); EQ3/6 (7); and TOUGH (8). These codes model processes at a level of detail not included in the current simplified total performance assessment models. The middle tier of the pyramid consists of a smaller number of subsystem models, such as waste package and engineered barrier models, and simplified transport models such as TOSPAC (9). At the apex are a few total system models such as the Total-System Analyzer of Wilson and others (10) and SPARTAN (11), which are designed to combine major subsystems simply and to allow probabilistic performance analysis.

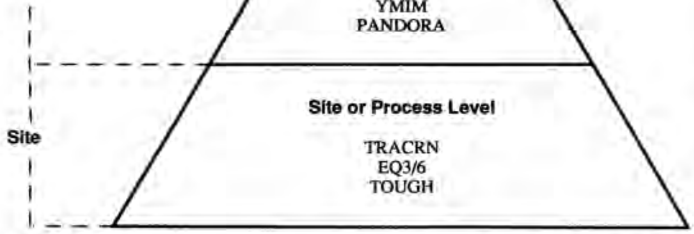

Fig. 2. Performance assessment strategy of the Yucca Mountain Site Characterization Project.

Site characterization research, coupled with process-level modeling, must develop a sufficient depth of understanding of processes such that simplifications by subsystem or total system models will still be valid. Site characterization builds confidence in this understanding by validation testing of the detailed models to the degree possible. It is in this context that the radionuclide migration program test strategy is described.

As shown in Fig. 3, knowledge of the site mineralogy, mineral stability, and water chemistry along with identification and modeling of potential changes in the natural state over time are needed to apply an understanding of radionuclide transport at Yucca Mountain. The radionuclide experimental effort includes characterizing solubility/speciation, sorption, and diffusion processes. Data applicability is determined in a dynamic system through column experiments. If laboratory validation testing is not supported by experimental results, then the characterization step begins again. This first step is critical because laboratory scale processes must be understood and limitations defined before results from larger scale transport tests can be understood. If data applicability is shown, field testing and/or use of analogues are now appropriate for continued validation testing of laboratory data and site models at the field scale.

Development of Testing Strategies for Specific Transport Processes

How are site conceptual models for specific transport processes developed and tested? Detailed study plans embodied in the YMP's Site Characterization Plan (1) discuss conceptual and experimental approaches, as well as modeling approaches, to studying processes such as radionuclide solubility and discuss alternate experimental paths that must choose from a range of testing alternatives. However, the study plans are not intended to call out explicitly how simplified models used in total system performance assessment can guide data collection. Prioritization of the work described in the study plans through testing strategies is needed, as is

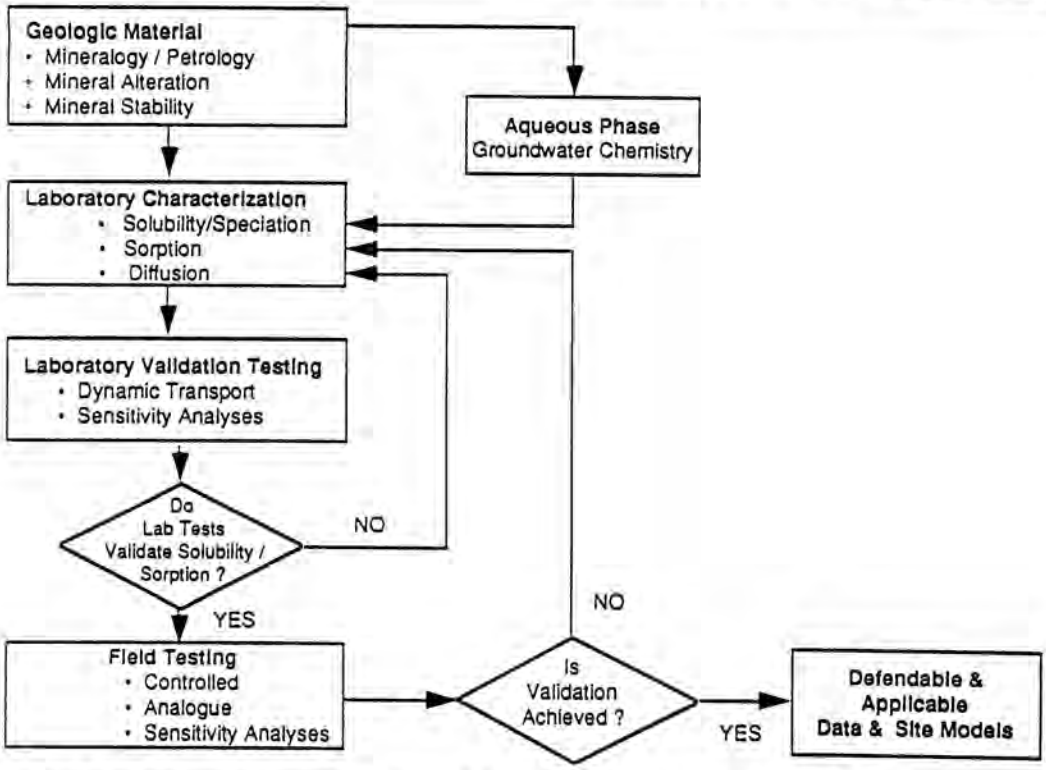

Fig. 3. Los Alamos strategy for radionuclide migration characterization and testing.

development of the process of iteration between model development and testing within characterization activities. This loop, which is often attributed to the performance assessment program, needs to occur at the site model and characterization level to support the necessary simplifications made in the subsystem or total-system-model levels. Frequently this iterative concept is only applied between the simplified total system or subsystem models and discrete data collection activities.

The testing strategies of the radionuclide migration program recognize limitations due to time and resources and allow for prioritization of data collection toward an applicable and defensible end. The testing strategy for radionuclide solubility and speciation is used as an example because it has been tested and carried through the cycle of Fig. 3. The radionuclide solubility and speciation study is divided into three major activities: 1) determination of empirical solubilities of key radionuclides; 2) identification of key radionuclide species, and 3) calculation of solubilities and speciation through modeling. Kerrisk (12) identified key solubility-limited radionuclides to study. The solubilities of Pu, Am, Np, U, Th, and Ra are being determined from both oversaturation and undersaturation at pH, Eh, and temperatures expected in the far-field. The solubility-controlling solid is also identified. Recent work is described by Nitsche (13) and Nitsche and others (14).

Identification of key radionuclide species also includes the determination of thermodynamic constants of formation. Use of photoacoustic/photothermal spectroscopy tools will help to identify the species at low concentrations (10^{-6} to 10^{-9}M). Thermodynamic modeling of radionuclide solubility and speciation calculates the number, type and distribution of species in solution, their solubility limits, and the solubility-limiting solid phases. These calculations are done over ranges of conditions expected at the Yucca Mountain site but not necessarily characterized in the laboratory. Exercising and testing the thermodynamic data base throughout study lifetime is crucial.

The radionuclide solubility and speciation strategy is shown in Fig. 4. If the aqueous nuclides reach the far-field, regardless of the waste package dissolution products and near-field processes, these nuclides will be controlled by the far-field environment. The strategy shows the important step of defining testing conditions (15). Temperatures of 25°C, 60°C, and 90°C, pH of 6, 7, and 8.5, and groundwater chemistries within reference compositional ranges found in the saturated zone in the Yucca Mountain region are considered. Radionuclide solubility limits and controlling solids are determined under these conditions. Other water will be considered when a "most active" groundwater chemistry is defined and the unsaturated zone water chemistry is determined. Radionuclide species are characterized concurrently and thermodynamic data is obtained. The thermodynamic data base is tested and updated. Validation testing occurs during data base testing, which determines whether observed and predicted solubilities or speciation agree, given the experimental conditions. Sensitivity studies are conducted to rule out the need to study all systems (ligand or alternate water chemistries) or condition ranges.

If the experimental and calculated solubilities and species do not agree, then additional characterization is needed. If compared results agree, the database is updated and is exercised over all anticipated conditions. The needed confidence level for agreement of experimental and calculational work remains a question to resolve, possibly through sensitivity analyses at a more systems-oriented model level. If release limits defined by regulations are met, then solubility-based compliance will be utilized for further analyses; otherwise,

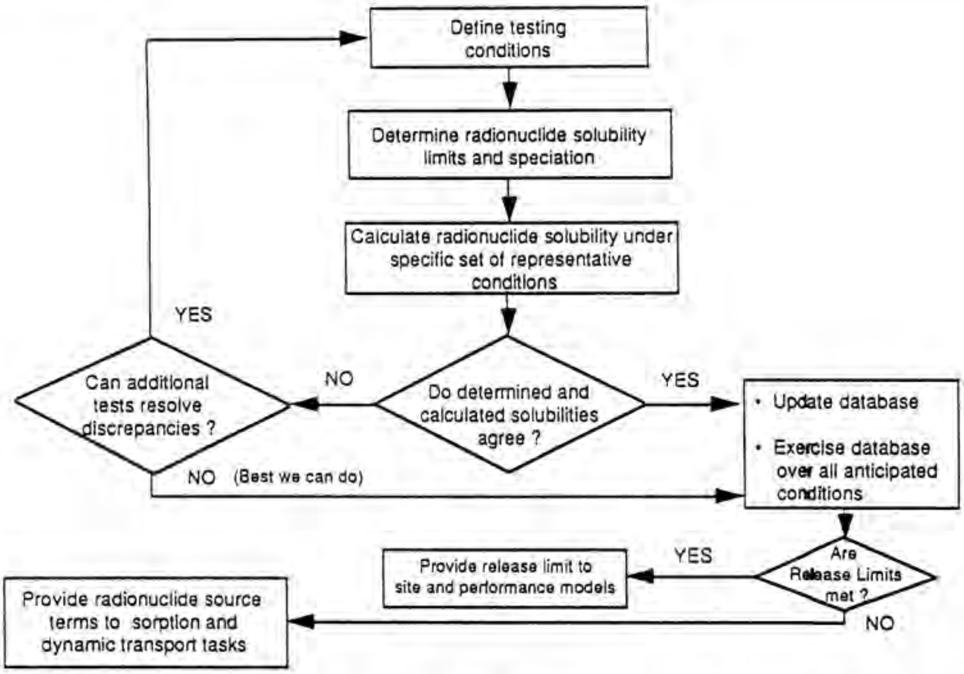

Fig. 4. Strategy for dissolved species concentration limit studies.

source terms will be provided to aid in radionuclide sorption characterization, modeling, and laboratory transport testing efforts.

The solubility/speciation strategy was applied to work done by Nitsche (13). Nitsche reported solubility data for Pu, Am, and Np in water from Well J-13 on the Nevada Test Site at temperature and pH ranges described above. His data were modeled and results showed that experimental and calculated solubilities for Pu, Am, and Np agreed fairly well, but experimental and calculated speciation results did not agree. Modeling results indicated phosphate species should form for Pu(IV) and Pu(VI) at pH 7 but these species were not observed. Given the small quantity of phosphate in J-13 water, phosphate data currently in the data base should be reexamined. Furthermore, much of the data used for modeling had to be assumed. Sensitivity analyses will focus the experimental effort on thermodynamic constants of formation for important species and should subsequently refine experiments to provide more realistic data.

Nitsche's results show the need to continually test and update the modeling data bases. Maintenance of the YMP thermodynamic data base (GEMBOCHS) entails evaluation and inclusion of generic thermodynamic data for radionuclides from the Nuclear Energy Agency Thermodynamic Data Base and inclusion of site-specific thermodynamic data. YMP has formed a working group of scientists to oversee the selection and inclusion of site-specific thermodynamic data for radionuclides to the data base, to ensure that the data are consistent, and to test the data base.

SUMMARY

The YMP's radionuclide migration program has developed testing strategies to prioritize efforts, ensure rigorous laboratory radionuclide characterization, and establish defensible validation testing of data and site models. This process provides the basis for necessary simplifications of subsystem and total system transport models and ensures confidence of applicability of site data in these models. The logic used in development of these strategies can be applied to many parts of the site characterization program to produce an integrated means of providing data that has been tested at various scales. Such methods will undoubtedly be valuable to increasing confidence in understanding and predicting performance of the site.

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