JUL 12 1994

MEMORANDUM FOR: Margaret V. Federline, Chief
Performance Assessment
and Hydrology Branch
Division of Waste Management, NMSS

FROM: Michael J. Bell, Chief
Engineering and Geosciences Branch
Division of Waste Management, NMSS

SUBJECT: REVIEW OF STUDY PLAN 8.3.1.3.5.1. AND 8.3.1.3.5.2., REV.0, "DISSOLVED SPECIES CONCENTRATION LIMITS AND COLLOID BEHAVIOR."

Enclosed is the ENGB staff review of Study Plan 8.3.1.3.5.1. and 8.3.1.3.5.2. (Rev.0, "Dissolved Species Concentration Limits and Colloid Behavior") for use by PAHB. It includes one comment and six questions. The format used for the enclosure is consistent with Revision 2 of the "Review Plan for the NRC Staff Review of DOE Study Plan" dated March 4, 1993.

ORIGINAL SIGNED BY

Michael J. Bell, Chief
Engineering and Geosciences Branch
Division of Waste Management, NMSS

Enclosure: As stated

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Sections 8.3.1.3.5.1 and 8.3.1.3.5.2 Dissolved Species Concentration Limits and Colloid Behavior

COMMENT 1

This study plan does not address how the study is related to other DOE investigations in related areas such as spent-fuel dissolution or high-level waste glass leaching. Nor is the plan properly justified in its practical applications to the Yucca Mountain project.

BASIS

This study plan describes the procedures for investigations of the formation of actinide colloids. The investigation is restricted to the formation of colloids and speciation behavior that result from germane actinides in J-13 well water or other pertinent solutions. It does not include the degradation of waste form (i.e., spent-fuel dissolution and HLW glass leaching). Therefore, the proposed tests may not be realistic. More specifically, the actinide supersaturation for the formation of colloids is arbitrary in the proposed tests. During degradation of the waste form, many properties of waste packages and conditions of repository environments are responsible for the supersaturation of groundwater with actinides. They include dissolution behavior of waste form, precipitation behavior of secondary phases, and flow rates of groundwater. The solubility-limiting phases considered in this plan may not be the same as those from the waste form degradation. For instance, Uranyl Silicates were observed in the spent-fuel dissolution [1]; Clay minerals were observed in the HLW glass leaching [2].

It is generally known that three different processes are responsible for the formation of colloids [3,4]. The first is the condensation of dissolved individual ions, i.e., the colloids formed by putting together dissolved individual ions. This condensation is the process considered in this study plan and discussed above. The second is the dispersion of integrated materials, i.e., the formation of colloids which occurs by breaking up solid-phase materials. The third is the formation of pseudo-colloids by sorption and precipitation of actinides on the surface of groundwater colloids. Colloids of clay, silica or iron oxide are normally present in groundwater. In this study plan, the second and third processes are not considered. Although it is preliminary, the dispersion process may be a very important source of colloids from the degradation of waste form [5,6]. On the other hand, the groundwater colloids appear to be most important in the far-field transport of actinides.

The proposed repository site of Yucca Mountain is an unsaturated site. The immersion condition considered in this study plan may not represent the anticipated environmental condition around waste packages. Groundwater drips may be primarily responsible for the degradation of waste form. Under drip conditions, the characteristics of groundwater may deviate significantly from those proposed in this study plan.

RECOMMENDATION

The study plan should consider the following researches: (a) effects of waste form degradation on colloid formation and solubility limits, (b) alternate mechanisms for colloid formation including dispersion and sorption, (c) colloid

formation in unsaturated environments including drip conditions, and (d) practical applications of the study to the Yucca Mountain Project. These issues have been partly addressed in various documents:

Scientific Investigation Plan (e.g., Characterization of Waste Form);

Site Characterization Plan (SCP) (e.g., 7.4.3.1.1. Spent Fuel Dissolution Studies);

SCP Progress Reports (e.g., 2.2.2.7. Study 8.3.1.3.4.1 - Batch Sorption Studies. Activity 8.3.1.3.4.1.4. Sorption on Particulates and Colloids, or 2.6.3.3. Activity 1.5.2.2 - Characterization of the Glass Waste Form);

Other Study Plans (e.g., 8.3.4.2.4.1. Characterize the Chemical and Mineralogical Changes in the Post-Emplacement Environment); and

Yucca Mountain Site Characterization Project Waste Package Implementation Plan.

The study plan should consider interrelationship of these activities.

REFERENCES

[1] D. J. Wronkiewicz, J. K. Bates, T. J. Gerding and E. Veleckis, Uranium Release and Secondary Phase Formation during Unsaturated Testing of UO_2 at 90°C, J. Nucl. Mat., 190, 107 (1992).

[2] J. K. Bates, C. R. Bradley, E. C. Buck, J. C. Cunnane, N. L. Dietz, W. L. Ebert, J. W. Emery, R. C. Ewing, X. Feng, T. J. Gerding, M. Gong, W.-T. Han, J. C. Hoh, J. J. Mazer, M. Tomozawa, L.-M. Wang, and D. J. Wronkiewicz, ANL Technical Support Program for DOE Environmental Restoration and Waste Management: Annual Report-October 1990-September 1991, Report No. ANL-92/9, Argonne National Laboratory, Argonne, IL. 1992.

[3] S. Yariv and H. Cross, Geochemistry of colloids Systems, Spring-Verlag, New York, NY (1979).

[4] J. I. Kim, The Chemical Behavior of Transuranium Elements and Barrier Functions in Natural Aquifer Systems, Mat. Res. Soc. Symp. Proc., 294, 3 (1993).

[5] P. A. Finn, J. K. Bates, J. C. Hoh, J. W. Emery, L. D. Hafenrichter, E.C. Buck and M. Gong, Elements Present in Leach Solutions from Unsaturated Spent Fuel Tests, Mat. Res. Soc. Symp. Proc., 333, 399 (1994).

[6] J. K. Bates, J. P. Bradley, A. Teetsov, C. R. Bradley and M. B. Brink, Colloid Formation during Waste Form Reaction: Implications for Nuclear Waste Disposal, Science, 256, 649 (1992).

Sections 8.3.1.3.5.1 and 8.3.1.3.5.2 Dissolved Species Concentration Limits and Colloid Behavior

QUESTION 1

What methods will be used to evaluate uncertainties in determining solubility limits?

BASIS

Determination of solubility limits is difficult either theoretically or experimentally. Theoretically, it is not known whether species formed are in a true equilibrium state or in a metastable steady state of supersaturated solution. Experimentally, it is difficult to filter colloids completely to determine the concentration of dissolved ions [1].

RECOMMENDATION

This study plan should prescribe methods to evaluate uncertainties in determining solubility limits.

REFERENCE

[1] J. I. Kim, Chemical Behavior of Transuranic Elements in Natural Aquatic Systems, Handbook on the Physics and Chemistry of the Actinides, A. J. Freeman and C. Keller, ed., Elsevier Science, New York, NY.

QUESTION 2

What is the relationship of bulk charge as described in Section 3.4.4 to surface charge?

BASIS

"Bulk Charge" as used in Section 3.4.4 is not well defined. The importance of bulk charge over surface charge is unclear. Normally, surface charge is responsible for many characteristics of colloids such as colloid suspension.

RECOMMENDATION

The study plan should describe bulk charge and its relationship to surface charge.

QUESTION 3

What is the definition of "certain aspects" as used on page 9 of the report?

BASIS

Page 9 contains the following quotation, "Nonequilibrium or kinetic models will be used as needed to describe certain aspects of radionuclide solubility,". The use of the phrase "certain aspects" is unclear.

RECOMMENDATION

The study plan should provide examples or further explain what "certain aspects" will be described.

QUESTION 4

The study plan needs revision to reflect the current design options, particularly in relationship to maximum test temperatures.

BASIS

The maximum test temperature of 60°C in page 12 is too low for current design options of the waste packages [1].

RECOMMENDATION

The study plan should be revised to reflect the current design options.

REFERENCE

[1] H. A. Benton, MPC/Waste Package Technical Considerations, Presentation to The 63th Meeting of Advisory Committee on Nuclear Waste (ACNW), OCRWM-Management & Operation Contractor/BWFC, April 20, Bethesda, MD (1994).

QUESTION 5

The staff questions the use of microfilters for removing colloids in the test plans.

BASIS

The use of filters of <0.1 μm for removing colloids (page 28) is unclear. The NRC staff is not convinced that the use of approximately nm filters is appropriate because some colloids can be smaller than approximately nm.

RECOMMENDATION

The study plan should describe in more detail the purpose and predicted effectiveness of the filters.

QUESTION 6

What screening process will be used for the selection of actinides?

BASIS

The selection of actinides is not well focussed in page 30. Simple performance assessments may screen out most of radionuclides listed in page 30. The staff needs to better understand the screening process for actinides.

RECOMMENDATION

The study plan should describe in more detail the screening process for actinides before detailed experiments are conducted.