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SOLUBILITY EXPERIMENTS FOR THE NEVADA NUCLEAR WASTE STORAGE INVESTIGATIONS PROJECT

by

Jerry F. Kerrisk

Abstract

In response to the need for experimental data on the solubility of waste elements under conditions that are characteristic of a potential repository at Yucca Mountain, the Nevada Nuclear Waste Storage Investigations project has proposed a series of experiments to measure solubilities. This report outlines the experimental conditions that will be used for these measurements. Water compositions, temperatures, important waste elements, the importance of solids in their control of solubility, and the effect of radiation are discussed.

I. INTRODUCTION

The Nevada Nuclear Waste Storage Investigations (NNWSI) project is studying a site at Yucca Mountain in southern Nevada as a potential nuclear waste repository. As part of the geochemical site characterization of Yucca Mountain, the effects of solubility on limiting radionuclide transport are being investigated. Solubility, by providing an effective limit on the concentration of dissolved waste elements in water passing through the repository, can contribute to the overall assurance that the site will effectively isolate waste elements from the environment.

The repository at Yucca Mountain would be sited in the unsaturated zone. In this setting, water is expected to flow downward through the repository area and the remainder of the unsaturated zone until it reaches the static water level, where it will combine with water in the saturated zone. The concentrations of radionuclides in water in the vicinity of the repository

represent a source term for transport calculations that must be done to assess the performance of the repository with respect to the Environmental Protection Agency's (EPA) standard, 40 CFR 191 (EPA, 1985). The concentrations of radionuclides are also involved in determining the release of radionuclides from the engineered-barrier system; these release rates are limited by the Nuclear Regulatory Commission's (NRC) technical criteria, 10 CFR 60 (NRC, 1983). Radionuclide concentrations in water passing through the repository can be limited by two mechanisms, low dissolution rates of the solid waste or solubilities of individual waste elements. The relative importance of these two mechanisms is uncertain at this time. Solid-waste dissolution rates, by their nature, are strongly dependent on dissolution mechanisms. Solubility, on the other hand, can provide upper limits on radionuclide concentrations in solution that can be extrapolated to long-time behavior. For many waste elements, concentrations limited by solubility may be lower than those derived from solid-waste dissolution rates. The existence of independent limits (solid-waste dissolution rates and solubility) on radionuclide concentrations in water leaving a repository adds to the assurance that these limits are realistic.

The importance of solubility has been highlighted by the preparation of an NRC technical position entitled "Determination of Radionuclide Solubility in Groundwater for Assessment of High-Level Waste Isolation" (NRC, 1984). This technical position serves as guidance in the preparation of detailed plans for waste-element solubility experiments. It recommends that any site that elects to use solubility to limit waste-element release design experiments to determine the solubility under site-specific conditions. The experiments outlined in this report are meant to satisfy the requirements of this technical position.

Although solubility is a relatively simple concept, the solubility of a specific element may depend on many parameters. The parameters that will be considered in designing these experiments are:

1. water composition including pH and Eh,
2. temperature,
3. identity of the solid controlling solubility,
4. the presence of other solids, and
5. radiation effects such as radiolysis.

The ranges covered by these parameters for the site at Yucca Mountain are discussed in the following sections.

The object of this report is to define a group of experiments that will provide the solubilities of important waste elements under conditions that are characteristic of the Yucca Mountain site. The needs of this project, that is, solubilities that represent upper limits on waste-element concentrations, are of primary concern in defining the experiments. However, solubility data without an understanding of basic processes involved are of only limited value. That is why stress is also put on results that will aid in understanding and modeling solubility. An assessment of the important radionuclides in waste that would be stored at Yucca Mountain has already been performed (Kerrisk, 1985). The conclusions of that assessment will be used to select waste elements for solubility experiments. The conditions or parameters of the solubility experiments and the first groups of waste elements to be studied are outlined here.

II. WATER COMPOSITIONS

The compositions of natural waters found at and near Yucca Mountain have been studied for a number of years as part of the NNWSI project and other Department of Energy programs at the Nevada Test Site. The effects of waste emplacement on water composition have also been studied as part of the NNWSI project. Although these studies are not yet complete, the results presently available will be used to define expected water compositions.

A. Natural Water Compositions

A review of natural water compositions found at and near Yucca Mountain has recently been published by Ogard and Kerrisk (1984). At this time, there are no direct analyses of water from the unsaturated zone at Yucca Mountain. Water from one well, well J-13, was chosen as characteristic of water compositions expected to be found in the unsaturated zone near the repository. This well taps the Topopah Spring Member tuff, which contains the repository horizon, at the well location in Fortymile Canyon. If future tests associated with the construction of an Exploratory Shaft at Yucca Mountain indicate that unsaturated-zone water compositions are not similar to well J-13 water, additional water compositions will be considered. Even though well J-13 water is characteristic of unsaturated zone water, waste elements may encounter other water compositions along flow paths toward the environment, particularly in the saturated zone. For this reason, two other compositions that bracket the range of waters expected in the vicinity of Yucca Mountain have also been

chosen for solubility measurements. Well UE25p#1 taps the carbonate aquifer that underlies much of this area (Winograd and Thordarson, 1975). Well UE25p#1 water has about an order of magnitude higher ionic strength and total carbonate content than well J-13 water; it represents the most concentrated natural water expected in the vicinity of Yucca Mountain. In addition, solubility measurements will be done in a dilute neutral electrolyte (probably sodium perchlorate); these measurements will provide data for a relatively simple system (only hydrolysis products and oxide or hydroxide precipitates) as well as for scenarios in which dilute waters reenter the repository after the thermal period. Solubilities measured in these three compositions along with a model such as EQ3/6 (Wolery, 1983; Wolery, 1984) will allow calculation of waste element solubilities at intermediate compositions.

The pH of waters from wells J-13 and UE25p#1 fall within relatively narrow ranges (Ogard and Kerrisk, 1984). To cover the possibility that shifts in pH might occur and to provide a better understanding of the influence of pH on solubility, solubilities will be measured over a range of pH. The nominal pH of well J-13 water is 7; solubility measurements will be done at pH 6, 7, 8.5, and 10. The nominal pH of well UE25p#1 water is 6.7; solubility measurements will be done at pH 5.5, 6.7, 8.5, and 9.5. For the neutral electrolyte, solubility measurements will be done at pH 5.5, 7, 8.5, and 10. For the two well waters, the nominal pH will be maintained by controlling the CO₂ partial pressure over the water; this represents the natural state of the water. The most appropriate method of modifying the pH for measurements at values other than the nominal depends on the natural processes that might change water pH. Methods that maintain the total carbonate content of the well waters at their nominal values will be used in this work.

The Eh of essentially all natural waters found at and near Yucca Mountain is oxidizing (Ogard and Kerrisk, 1984). The exceptions are well H-3 and the packed-off Bullfrog zone of well UE25b#1. Waste-element solubilities are expected to be higher under oxidizing conditions than under reducing conditions. For this reason, all solubilities will be measured under oxygen-saturated solutions. An attempt will be made to characterize the Eh of these solutions.

B. Effect of Waste Emplacement on Water Compositions

Waste emplacement can affect water compositions in the vicinity of the repository by promoting reactions with the host rock or with the waste-package

and engineered-barrier materials. At this time, water from only one source, well J-13, is being used as characteristic of the unsaturated zone. Thus, reactions of well J-13 water with these materials at temperatures up to the maximum temperature expected at the repository are of interest in defining compositional changes.

1. Reactions with the Host Rock. A series of experiments studying reactions between well J-13 water and Topopah Spring tuff has been conducted by Lawrence Livermore National Laboratory (LLNL) as part of their waste-package program (Oversby, 1984). Although this work is still in progress, results of tests at 90 and 150°C for reaction times up to 72 days show only minor changes in water chemistry. These experiments support the position that well J-13 water is at or near equilibrium with the host rock. Based on the results of these experiments and the current understanding of water-chemistry controls at Yucca Mountain, no variations in well J-13 water chemistry are required in solubility experiments to account for reactions with the host rock. Increases in pH observed during these experiments were attributed to loss of CO₂; the variation of pH planned for well J-13 solubility experiments in Section A above will cover this phenomenon. No changes in water Eh are expected as a result of reaction with the host rock.

2. Reactions with Waste-Package or Engineered-Barrier Materials. Tests of borosilicate-glass and spent-fuel dissolution have been done as part of the Waste Package Task of the NNWSI project. These tests have used well J-13 water and have included stainless steel, the proposed NNWSI waste-package material. Analyses of water from these tests for components of stainless steel have shown Fe at approximately 0.1 ppm, Cu at approximately 0.03 ppm, Ni less than 0.03 ppm, and Ag, Be, Mn, Mo, and Pb less than 0.01 ppm (Oversby, 1985). The Fe result is in the range of Fe concentrations observed in wells J-13 and UE25p#1 waters (Ogard and Kerrisk, 1984). Both Cu and Ni have been found in well J-13 water at approximately 0.005 ppm (Claassen, 1973). Because of the small changes in water composition associated with the contact of well J-13 water with waste-package material, there are no plans to vary water compositions outside their natural range during solubility experiments.

Work on reaction of well J-13 water with the host rock and waste-package materials is still in progress. If significant changes in water compositions are observed from these experiments in the future, the solubility conditions outlined in this report will be reevaluated and changed as necessary.

C. Water Analyses

As part of the solubility experiments, samples of the water used will be analysed periodically. These analyses will be used to check the stability of the water as a function of time. Selected analyses will also be done for the major components in the water at the end of solubility experiments to check for changes in composition. Significant changes in water composition are not anticipated.

III. WATER TEMPERATURES

Water temperature can have an effect on waste-element solubility. By making solubility measurements over a range of temperatures that cover expected conditions at the repository and along flow paths to the environment, the effect of temperature can be defined experimentally and data needed to use thermodynamic models such as EQ3/6 can be obtained.

Temperatures of water from the tuffaceous aquifer at and near Yucca Mountain have been reported in the 23-38°C range (Benson, et al., 1983). The highest water temperature recorded in the area is for water from the carbonate aquifer (well UE25p#1) at 57°C (Craig and Robison, 1984). These temperatures are consistent with measurements of other water temperatures from wells and springs in the area (Claassen, 1983). The effect of waste emplacement will be to raise water temperatures in the vicinity of the waste. At this time, maximum temperatures with liquid water present are expected to be limited by the boiling point of water at Yucca Mountain, about 95°C (Braithwaite, 1985).

For well J-13 water, which has been selected as characteristic of vadose water in the host rock, solubility tests will be done at 25, 60, and 90°C. These temperatures cover the range from pre-emplacement temperatures to the maximum temperature where solubility would be important. For well UE25p#1 water, solubility tests will be done at 25 and 60°C. Water with this composition may be found along flow paths in the saturated zone, but is not expected to be found within the host rock. Maximum temperatures in the saturated zone under the repository are expected to be below 60°C (Johnstone, et al., 1984), and maximum saturated zone temperatures along flow paths away from the repository will also be less than 60°C. Solubility tests in the neutral electrolyte will also be done at 25 and 60°C.

IV. SOLIDS AND SOLUBILITY CONTROL

Solids enter solubility experiments in two ways. For an equilibrium solubility condition to exist for some element, a solid containing the element must be in contact with the solution. The most stable solid participating in reactions with aqueous species of the element that are rapid on the time scale of the experiment controls the maximum aqueous concentration of the element. Other solids of the element that would lead to higher solubilities under the conditions of the experiment should dissolve because they are less stable than the solid that controls solubility. Still other solids of the element that are more stable than the solid controlling solubility under the conditions of the experiment can exist, but may not participate because precipitation or dissolution kinetics are too slow. A common example of this situation exists with solid silica phases. The aqueous silica content of many natural waters is controlled by amorphous-silica or cristobalite solubility at silica contents well above the solubility of quartz, the most stable silica polymorph under the water conditions. Another way for solids to influence solubility experiments is for solids other than the one controlling solubility to be present. This can lead to sorption of elements participating in the solubility reactions or coprecipitation under some conditions.

Ideally, solubility experiments should approach steady state from oversaturation and undersaturation. Approach from oversaturation is usually done by preparing oversaturated solutions of the element and allowing precipitation to occur until a steady state is achieved; the solid formed must then be identified and characterized. Approach from undersaturation is usually done by preparing a characterized sample of the solid and allowing it to dissolve until a steady state is achieved. If equilibrium is attained, the aqueous concentration of the element under investigation should be the same as measured by undersaturation or oversaturation experiments.

For the solubility measurements outlined here, an attempt will be made to approach steady-state concentrations from both undersaturation and oversaturation. Experiments approaching steady-state concentrations from oversaturation will be done first and the solids that form will be characterized. Where possible, these solids will be prepared and used for experiments that approach steady state from undersaturation. This procedure has the advantage of not specifying the solid that controls solubility, but of allowing the system under investigation to determine the solid that will precipitate. There are no plans

to include other solids such as tuff from Yucca Mountain in the solubility experiments because they are not needed to control water composition. Adding other solids would only complicate interpretation of the experiments.

Phase separation represents a significant practical problem for solubility measurements, particularly with some of the actinides being examined here. Incomplete phase separation, that is, leaving some of the solid with the solution phase, would lead to overestimates of waste element solubilities. Phase separations should be as complete as possible for these experiments. In support of this philosophy, experiments to assess the effects of various phase-separation techniques (such as filtration and centrifugation) on the apparent solubility will be done. These experiments are not a major part of this program, but they are necessary to estimate the effects of incomplete phase separation on the measured solubilities.

For elements that can exist in various oxidation states, an attempt will be made to determine the oxidation states of the aqueous species in solution. This determination is not always possible, especially for elements with very low solubilities. This information, in conjunction with data on the solid, will aid thermodynamic or kinetic models of the experiments.

V. WASTE ELEMENTS

Kerrisk (1985) has recommended priorities for waste-element solubility measurements. Based on these recommendations, the Solubility Determinations Task of the NNWSI project has proposed solubility measurements for the following groups of waste elements:

1. Pu, Am, and Np,
2. U and Th,
3. Ra, Ni, and Zr, and
4. other waste elements.

The category of other waste elements will be specified in the future. Elements such as Sn, Pa, Se, Pb, and Pd are under consideration for solubility measurements. There are no plans at this time to attempt solubility measurements for C, Tc, Cs, Sr, or I. The solubility of C as CaCO_3 is well known. Solubility limits will not be employed for the other elements because of their large solubilities under the conditions expected at Yucca Mountain.

VI. RADIATION

Radiation can have an effect on the solubility of waste elements by altering the composition of the water or by influencing the crystallinity of the solids that form. The primary effects of gamma radiation will be a reduction in water pH and a trend toward more oxidizing conditions as long as air is present; secondary effects will be the production of nitrate (or nitrite) anions and the possible production of small quantities of organics from carbonate that is naturally present in the water. Gamma radiation will be most important early in the life of the waste and will only affect water near the waste. By 300 years after discharge, the total gamma power (watts) is about a factor of 500 below the level at 10 years after discharge (Croff and Alexander, 1980). Thus, by the earliest time containment would be breached (NRC, 1983), the effects of gamma radiation will be reduced considerably (Burns, et al., 1982). The effects of gamma radiation on waste-element dissolution and water composition are being studied in the Waste Package Task of the NNWSI project. The range of water compositions produced by gamma radiation is covered by the water compositions already considered (see Section II.A). No solubility measurements will be made in this task in artificial gamma fields.

Alpha radiation can affect water compositions in ways that are similar to the effects of gamma radiation. The primary effects of alpha radiation are a decrease in pH and a trend toward more oxidizing conditions in the water. Solids composed of alpha emitters tend to show self-irradiation damage to their crystal structure; solubility of solids such as the actinide oxides and hydroxides is affected in that amorphous solids are more likely to be the natural precipitation product (Rai et al., 1983; Nitsche and Edelstein, 1985). The effects of alpha radiation on solubility will be investigated by using radionuclides of the actinides that are present in spent fuel and high-level waste. Table I lists the specific activity in Ci/g of a number of the important actinide alpha-emitters. Table II lists the average specific activity of four actinide elements in spent fuel as a function of time after discharge from the reactor (Croff and Alexander, 1980). This quantity varies with time because the isotopic composition of the elements is a function of time. Based on these data, selected solubility experiments will be done with two isotopes of Pu (^{239}Pu and ^{242}Pu) and two isotopes of Am (^{241}Am and ^{243}Am)

TABLE I
SPECIFIC ACTIVITIES OF ALPHA EMITTERS

<u>Isotope</u>	<u>Half Life (years)</u>	<u>Specific Activity (Ci/g)</u>
^{238}Pu	87.8	17.1
^{239}Pu	2.41×10^4	0.062
^{240}Pu	6540	0.23
^{242}Pu	3.76×10^5	3.9×10^{-3}
^{241}Am	432	3.4
^{243}Am	7370	0.20
^{237}Np	2.14×10^6	7.1×10^{-4}
^{234}U	2.45×10^5	6.2×10^{-3}
^{235}U	7.04×10^8	2.2×10^{-6}
^{236}U	2.34×10^7	6.5×10^{-5}
^{238}U	4.47×10^9	3.4×10^{-7}

TABLE II
AVERAGE SPECIFIC ACTIVITY OF ACTINIDES IN SPENT FUEL

<u>Element</u>	<u>Specific Activity (Ci/g) for Various Decay Times (years)</u>			
	<u>100</u>	<u>1000</u>	<u>10000</u>	<u>100000</u>
Pu	0.38	0.11	0.083	0.029
Am	3.2	2.7	0.20	0.20
Np ^a	7.0×10^{-4}	7.0×10^{-4}	7.0×10^{-4}	7.0×10^{-4}
U	2.3×10^{-6}	2.7×10^{-6}	2.8×10^{-6}	2.8×10^{-6}

^aExcludes ^{239}Np , a beta emitter.

to test the effect of alpha activity on solubility. Previous solubility experiments in well J-13 water have been done using ^{242}Pu spiked with ^{238}Pu to a specific activity about 13 times the specific activity of ^{239}Pu (Nitsche and Edelstein, 1985). Because the range of alpha particles is so short, there are no plans to investigate the effects of alpha radiation on the solubility of waste elements that are not alpha emitters. The effect of alpha radiation on water composition has been considered in defining the ranges of water composition that will be employed for solubility measurements (see Section II.A).

VII. EXPERIMENTAL MATRIX

Based on the discussions in Sections I - VI, an experimental matrix has been prepared for the solubility measurements. Table III outlines the matrix.

TABLE III
SOLUBILITY TEST MATRIX

Water Compositions^a

<u>Water</u>	<u>pH</u>	<u>Temperature</u>
J-13	6, 7, 8.5, 10	25, 60, 90°C
UE25p#1	5.5, 6.7, 8.5, 9.5	25, 60°C
Neutral electrolyte	5.5, 7, 8.5, 10	25, 60°C

Waste Elements

Pu (^{239}Pu , ^{242}Pu),^b Am (^{241}Am , ^{243}Am),^b Np
U, Th
Ra, Zr, Ni
Others

^aWaters will be oxygen saturated.

^bSelected solubility experiments will be done with two isotopes of Pu and Am to test the effect of alpha activity.

VIII. EXPECTED EXPERIMENTAL RESULTS

The experiments described in this report are expected to provide the following results.

1. Steady-state concentrations of waste elements in well J-13 and UE25p#1 waters and in the neutral electrolyte solution at the temperatures indicated will be reported. Where possible, an identification of the oxidation states present in the aqueous phase will also be done.
2. The solids that control solubility in each water composition will be characterized. This characterization will include composition with a phase identification if possible.
3. Estimates of the efficiency of phase separation will be reported based on selected experiments.
4. Periodic checks of the water compositions before and after the solubility experiments will be done.

IX. MODELING SUPPORT

A Thermodynamic Modeling Subtask is an integral part of the Solubility Determinations Task. In addition to any direct use of the experimental solubility data in performance assessment analyses, these data will be used to test thermodynamic models of the solubility process. At the present time, the EQ3/6 chemical equilibrium code is being used to model waste-element solubilities in the NNWSI project (Wolery, 1983; Wolery, 1984). Solubilities calculated with EQ3/6 under the conditions of the experiments will be compared with measured solubilities as a method of verifying the thermodynamic calculations. If necessary, thermodynamic data for waste-element solids will be derived from the experimental solubility measurements for correction of existing data or for addition of new solids to the data base. In cases where existing thermodynamic data cannot model the variation of solubility with water composition or temperature, experiments to measure formation constants of important aqueous complexes will be proposed. The object of the Modeling Subtask is an understanding of the parameters that affect the solubility of waste elements under the conditions expected for a repository at Yucca Mountain. This modeling effort will be carried on in parallel with the experimental program in order to maximize the interaction of these efforts.

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