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Dear John:

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Sincerely,

[Signature]

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GKJ:bek

Enclosure

- cc w/o encl: Office of the Director, NMSS (Attn: Program Support Branch)
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EG/CR-4236, Vol. 3

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PROGRESS IN EVALUATION OF RADIONUCLIDE GEOCHEMICAL INFORMATION
DEVELOPED BY DOE HIGH-LEVEL NUCLEAR WASTE REPOSITORY SITE PROJECTS:
REPORT FOR APRIL - JUNE 1985

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ABSTRACT

Geochemical information relevant to the potential mobility of radionuclides at the Hanford Site (in basalt) and the Yucca Mountain site (in tuff), candidate sites high-level nuclear waste geologic repositories, being developed by Department of Energy (DOE) projects is being evaluated by Oak Ridge National Laboratory (ORNL) for the Nuclear Regulatory Commission (NRC). Neptunium(V) sorption isotherms in three different basalt/synthetic groundwater systems were initiated this quarter. Uranium(VI) sorption isotherms were completed with McCoy Canyon basalt and synthetic groundwater GR-2. The control of U(VI) solutions at a level of $\sim 10^{-4}$ mol/L is apparently dominated by the precipitation of sodium boltwoodite. Different apparent concentration limits for uranium were obtained with GR-2 and GR-4 solutions. The results suggest that uranium sorption/solubility behavior could be substantially different in the various basalt units likely to be encountered by groundwater during migration from the waste package to the environment. The EQ3/6 software package was implemented and tested on the ORNL computer system.

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PREFACE

This document is the eighth in a series of progress reports that are being issued by the Nuclear Regulatory Commission to describe the current status of an experimental program to evaluate the radionuclide geochemical information developed by the high-level nuclear waste repository site projects of the Department of Energy.

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1. EXECUTIVE SUMMARY

This project, supported by the Nuclear Regulatory Commission (NRC), is being conducted to evaluate the information and data acquisition methodology on the geochemical behavior of radionuclides that may be employed by the Department of Energy (DOE) high-level nuclear waste repository site projects in performance assessment calculations to show reasonable assurance of expected compliance with regulatory requirements. This project is focused on parameters that are important to the mobility of radionuclides in geologic media (primarily, sorption on host rock and apparent concentration limits in groundwater) under the anticipated geochemical conditions of the repositories. Initial emphasis was on information being developed by the Basalt Waste Isolation Project (BWIP) that is applicable to the Hanford Site candidate repository in the Columbia River basalts at Richland, Washington. Attention is now being shifted to evaluate information that is being developed by the Nevada Nuclear Waste Storage Investigations (NNWSI) project that is relevant to the Yucca Mountain site in tuff near the Nevada Test Site in Nevada.

Work to characterize the crystalline and amorphous materials in our samples of tuffs at Yucca Mountain has been initiated.

Determination of a series of Np(V) sorption isotherms with three basalt/synthetic groundwater systems under anoxic redox conditions at 60°C was initiated this quarter. The purpose of these tests is to investigate whether neptunium is likely to exhibit similar sorption behavior in different basalt units during migration from the waste package.

Uranium(VI) sorption isotherms were completed with McCoy Canyon basalt and synthetic groundwater GR-2 under anoxic redox conditions at 60°C for 50 d. The behavior of uranium solutions (10^{-4} mol/L) is largely controlled by sodium boltwoodite precipitation. An apparent concentration limit of $\sim 2 \times 10^{-5}$ mol/L uranium was observed. We previously reported a considerably lower apparent concentration limit ($\sim 8 \times 10^{-7}$ mol/L) with Cohasset basalt and synthetic groundwater GR-4 under parallel test parameters. It is possible that precipitation kinetics may be different in the two systems, or that other factors related to groundwater composition could be involved. In any case, it appears that uranium sorption/solubility behavior could be significantly different in different basalt units during groundwater migration from the waste package to the environment.

Version 3230B of the geochemical code EQ3/6 was implemented and tested on the ORNL computer system this quarter. In addition to the EQ3/6 code and its required supplementary code EQLIB, the codes MCRT and EQTL were also brought up on the ORNL computer. MCRT and EQTL provide for the compilation, documentation, and management of thermodynamic data files required to run EQ3/6. The successful conversion of the codes was confirmed by running test cases.

The objective of this project is to support the NRC staff analysis of geochemical information used by the DOE to predict the performance of candidate high-level waste (HLW) geologic repository sites. Under this project, both experimental and calculational activities are undertaken to evaluate pertinent geochemical values and the methodology used in the development of these values. The results of the work conducted under this project are compared with published information reported by the DOE site projects or others, and (where appropriate) concerns are expressed relative to the accuracy, conservatism, or relevance of the information for the prediction of the potential isolation performance of the candidate repository sites.

This project is primarily a laboratory activity because it is anticipated that the DOE may rely on experimentally measured apparent concentration limits and sorption distribution coefficients for key radionuclides. The experimental methodologies used by the DOE site projects, the selection and characterization of site materials used in the DOE experiments, and the DOE test results are being evaluated. The appropriateness of the materials selected and methodology employed to simulate repository geochemical conditions are being considered. The conservatism inherent in the laboratory approach is being evaluated. Attention is also directed toward geochemical modeling efforts both to support our laboratory work and to evaluate the data bases and calculational methodologies employed in the geochemical modeling used by the DOE. For example, the DOE site projects may elect to calculate some geochemical values, such as radionuclide solubility, without supporting laboratory measurements.

The DOE site projects may also use modeling methodology to extrapolate laboratory information to conditions of temperature and chemistry that are unexplored experimentally. These and other aspects of the geochemical methodology and values used by the DOE site project will be examined in light of the reasonable assurance criteria of the NRC regulation 10 CFR Part 60.

3.1 HANFORD SITE MATERIALS

The preparation and characteristics of the basalt samples and synthetic groundwaters, and comparison of the synthetic groundwater formulations used in the sorption experiments have been described previously (KELMERS 1985a).

3.2 YUCCA MOUNTAIN MATERIALS

Four core samples from three different tuff units (Topopah Springs, Calico Hills, and Bullfrog) from drill hole USW-G1 and a sample of Topopah Springs outcrop from Busted Butte were previously received from the U.S. Geological Survey (see KELMERS 1985c for a description of the samples). Work completed this quarter consisted of a preliminary petrographic examination of polished thin sections of these samples. This examination has revealed several important features of the tuffs. It was evident that all three tuffs are not only heterogeneous texturally, but also are very different from one another. Particularly noteworthy is the abundance of lithic fragments in the Topopah Springs and the Calico Hills tuffs, and to a lesser extent, in the Bullfrog tuff as well. Also, it is apparent that the phenocrysts in the three tuffs are different both mineralogically and texturally. Detailed petrographic and x-ray diffraction analysis of the samples, when completed, will be described in future reports.

3.3 BATCH CONTACT METHODOLOGY

As reported previously (KELMERS 1985c), the pH gradually increases in samples of J-13 groundwater, either synthetic or actual, when exposed to air. This increase in pH is a result of the loss of dissolved CO₂ to the atmosphere. Therefore, in long-term batch contact tests to measure sorption ratios, methods must be devised to establish a carbon dioxide partial

pressure above the test solutions that maintains the appropriate pH. In tests reported last quarter (KELMERS 1985c), an estimate of somewhat less than 5% CO₂ by volume was given as an appropriate composition of an air/CO₂ mixture required to maintain J-13 at about pH 7.

To enable us to control carbon dioxide composition, a glove box was set up and appropriate valves and flowmeters were installed so that various mixtures of air and CO₂ could be blended and put through the box in a once-through mode. A CO₂ monitor (PIR-2000 Process Gas Analyzer from Horiba Instruments) was installed so that measurements of the CO₂ concentration necessary to maintain the pH of J-13 groundwater could be initiated. The results of the tests, when completed, will be described in future reports.

4.1 SORPTION ISOTHERMS

During the report quarter, we began a series of Np(V) sorption isotherm determinations under anoxic conditions at 60°C for the synthetic ground-water/basalt systems: (1) GR-2/McCoy Canyon basalt, (2) GR-2/Umtanum basalt, and (3) GR-4/Cohasset basalt. We will report the results of these isotherm determinations when they are completed, and when we have established the maximum neptunium concentration that can be included in the isotherm experiments.

5.1 PUBLISHED SOLUBILITY AND SORPTION INFORMATION

A summary of the published information describing uranium solubility limits and sorption distribution coefficients relevant to the candidate repository site in the Columbia River basalts was given in a previous report (KELMERS 1984a). The sorption and solubility information has also been separately reviewed and assessed (BLENCOE 1985; KELMERS 1984b).

5.2 RESULTS

During the current report period, we completed a 50-d sorption isotherm for U(VI) onto McCoy Canyon basalt from synthetic groundwater GR-2 under anoxic conditions at 60°C. For Cohasset basalt and GR-4, we had previously observed significant increases in sorption ratios (Rs) when the duration of the test was increased from 14 to 50 d under anoxic conditions at 60°C (KELMERS 1985c). In some cases, the Rs values increased by a factor of ten; values of Rs ranged from 19.4 to 31.1 L/kg at the lower initial concentrations of uranium (10^{-7} mol/L), while an Rs value of 335 L/kg was observed at the highest initial concentration of uranium, 1.02×10^{-7} mol/L. This large Rs value was caused mainly by precipitation of an insoluble compound of uranium, sodium boltwoodite (KELMERS 1985c). In the 50-d experiments with the GR-4/Cohasset basalt system, values of Rs ranged from 115 to 669 L/kg at the lower range of uranium concentrations (10^{-7} mol/L), while a value of 1288 L/kg was observed for the highest concentration (10^{-7} mol/L). The large increases in Rs values in the 50-d experiments, compared to the 14-d experiments, were probably a result of increased precipitation of sodium boltwoodite at all concentrations of uranium, since we observed large decreases in concentration from all of the control samples (no basalt present) in the 50-d experiments.

The values of R_s for McCoy Canyon basalt in GR-2 for all conditions, anoxic and oxic, were in the range of 2 to 5 L/kg, but a value at 50 d and 60°C had not been previously determined. The R_s values for McCoy Canyon basalt are much lower than those observed for the GR-4/Cohasset system. We are now using much more careful oxygen removal techniques in our anoxic experiments than we did in last year's determinations with McCoy Canyon basalt. Anoxic 50-d experiments with McCoy Canyon basalt in GR-2 were, therefore, carried out to see whether these more stringent oxygen-removal techniques would cause the sorption ratios for uranium to exceed the 2 to 5 L/kg range that we had observed in all previous experiments with this system and to see whether increasing the duration of the experiment from 14 to 50 d would cause the same large increase in sorption ratio as was observed with the Cohasset/GR-4 system. The results of these experiments are shown in Table 5.1 and are plotted in Figs. 5.1 and 5.2.

For the three lowest concentrations, the values of R_s for the 50-d experiments are approximately twice as high as those for the 14-d experiments. At the higher concentrations of uranium, the value of R_s , 48 L/kg, is ~35% higher than the value from the 14-d test. The large losses of uranium from the control experiments suggest precipitation of uranium again, probably forming sodium boltwoodite, as we have already observed with some systems. However, we did not directly observe the formation of a precipitate because so little could have been formed; it would be difficult to see the precipitate in the translucent polypropylene test tubes. In other

experiments previously reported (KELMERS 1985b) formation of sodium bolt-woodite was observed to form from solutions of uranium (10^{-4} mol/L) in GR-2. The small increases in Rs at the two lower concentrations of uranium suggest that the more stringent oxygen removal techniques used in these experiments did not cause a large difference between the 14- and 50-d experiments.

Table 5.1 Sorption of uranium on McCoy Canyon basalt from GR-2.
(anoxic conditions, 60°C, 50 d)

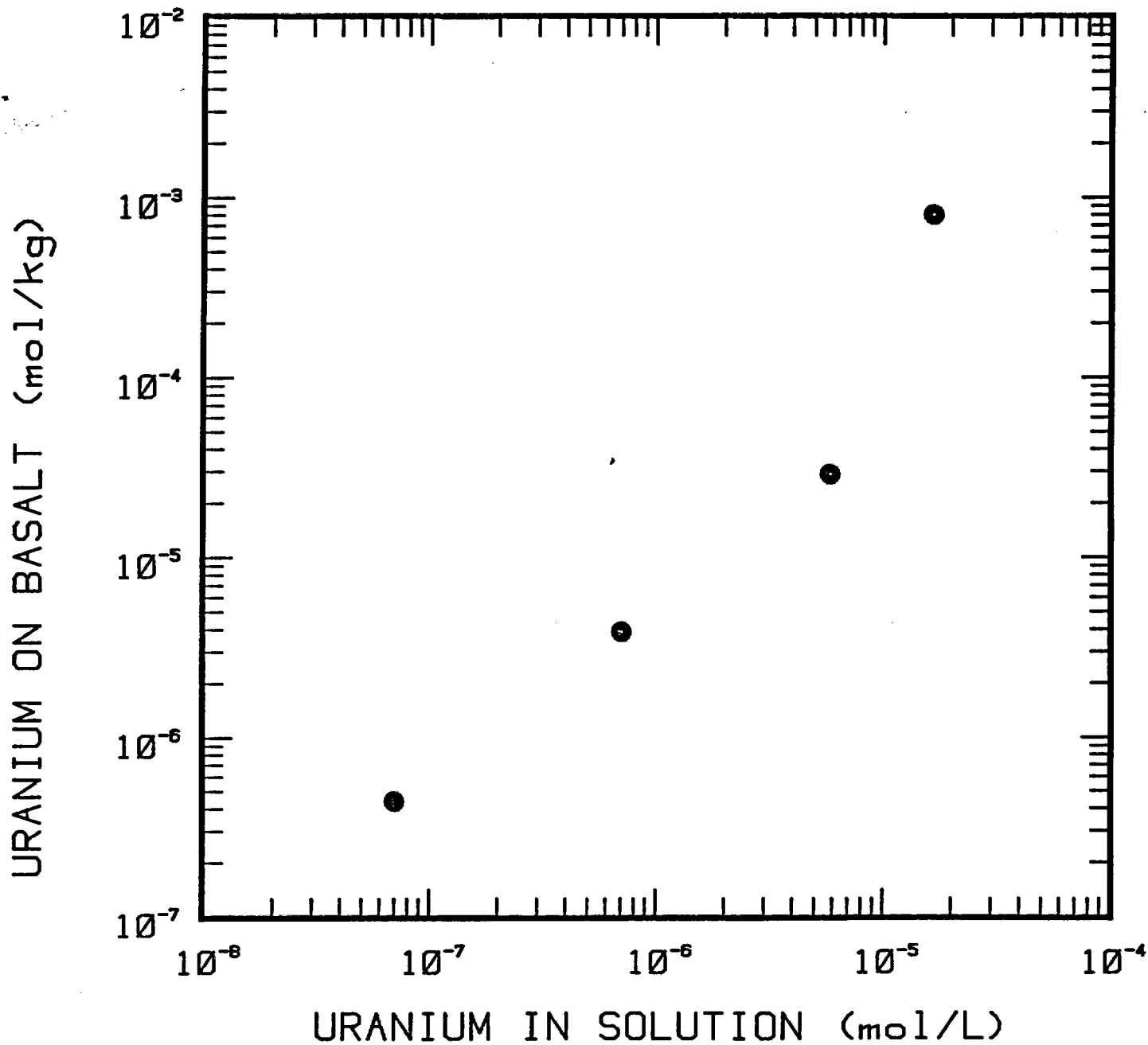
Initial uranium (mol/L)	Rs calc. ^a from standards (L/kg)	Rs calc. ^a from controls (L/kg)	% loss of uranium from control	Rs values ^b for 14 d (L/kg)
1.15×10^{-7}	6.3 ± 0.8	4.8 ± 0.7	9.0	3.9 ± 0.2
1.10×10^{-6}	5.5 ± 0.4	1.1 ± 0.3	29.5	2.5 ± 0.2
8.85×10^{-5}	4.9 ± 0.4	-3.9 ± 0.5	60.7	2.0 ± 0.4
1.00×10^{-4}	48.0 ± 1.7	-4.3 ± 0.3	90.9	35 ± 7

^aIn column two, values of Rs were determined by comparison of the final concentration of the uranium with the concentration of the standard solutions used initially. In column three, values of Rs were calculated by comparison with the control samples (no basalt present). These controls were equilibrated under the same conditions for the same length of time as the samples with basalt. For the two higher concentrations of uranium, the loss of uranium was greater in the controls than in the samples, and the resulting sorption ratios are, therefore, negative (see discussion below).

^bReported in Kelmers (1985a). All test conditions were the same except for the 14-d duration of the test. Initial concentrations of uranium in the the 14-d test were slightly different from those in the first column, but they were always within 15%.

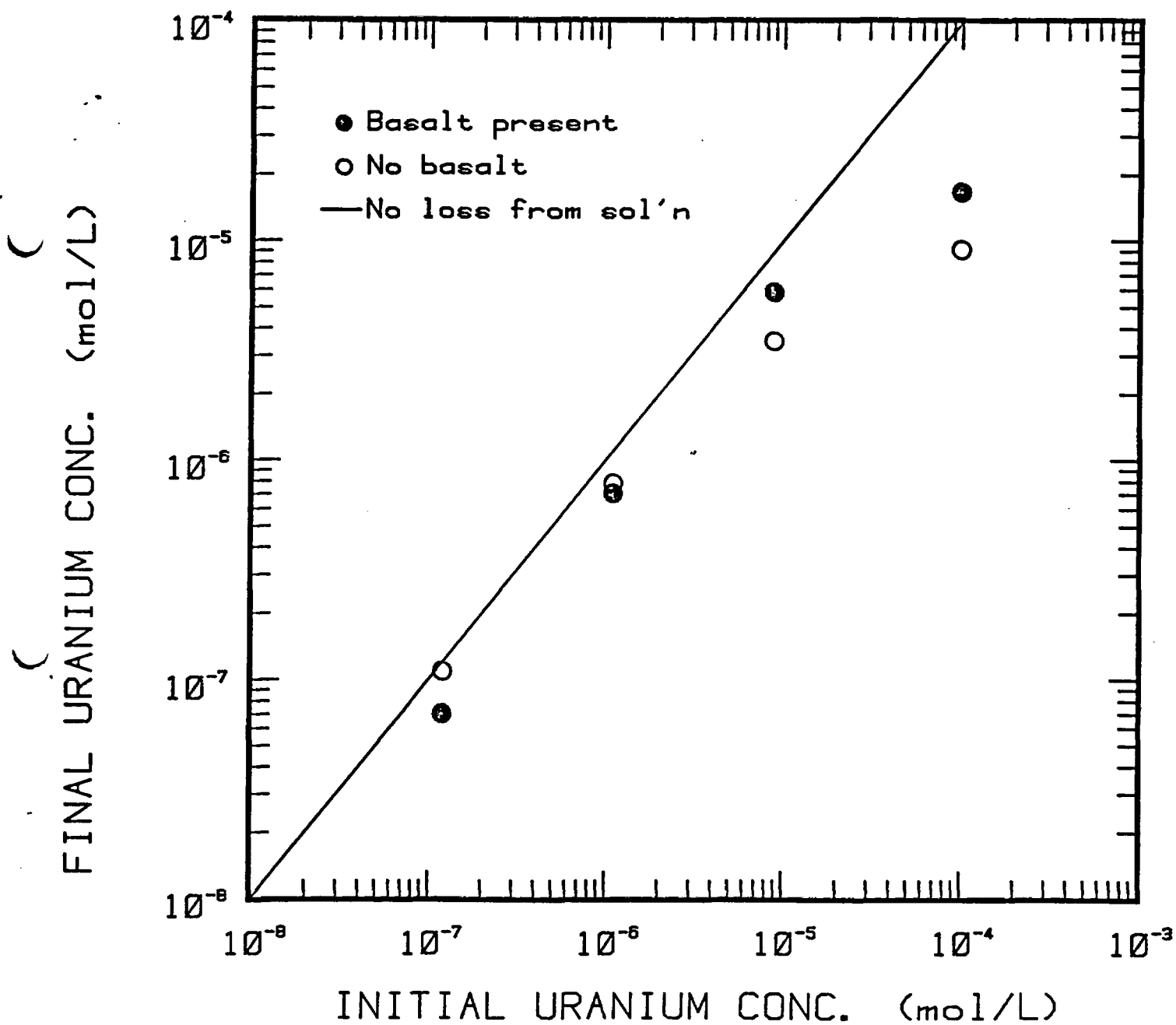
5.3 DISCUSSION

Our experiments indicate that the behavior of U(VI) solutions at concentrations of 10^{-4} mol/L at 60°C in synthetic groundwaters GR-2 and GR-4 is largely controlled by a precipitation process. As reported previously (KELMERS 1985c),



Sorption isotherms for uranium on McCoy Canyon basalt. Batch contact experiment anoxic conditions with synthetic groundwater GR-2 at 60°C. 50 days.

Fig. 5.1. Sorption isotherms for uranium on McCoy Canyon basalt. Batch contact tests, anoxic conditions with synthetic groundwater GR-2.



Initial and final uranium concentrations for sorption isotherm experiments with McCoy Canyon basalt in GR-2. Anoxic at 60C. 50 days.

Fig. 5.2. Initial and final uranium concentrations for sorption isotherm experiments with McCoy Canyon basalt in GR-2. Anoxic conditions at 60°C, 50 d.

x-ray diffraction analysis of the light-yellow precipitate formed from solutions of GR-2 and from GR-4, both initially containing 10^{-4} mol/L of U(VI), indicated that the solid formed was sodium boltwoodite, $\text{Na}(\text{UO}_2)(\text{SiO}_3)(\text{OH}) \cdot 5\text{H}_2\text{O}$.

Plots of sorption isotherms of uranium on basalt for the two systems GR-2/McCoy Canyon basalt and GR-4/Cohasset basalt indicate apparent concentration limits of uranium at the upper solution concentrations. For the GR-2/McCoy Canyon system under anoxic conditions, a limit of over 2×10^{-5} mol/L was indicated by the 14-d isotherm and slightly under 2×10^{-5} mol/L after 50 d (Fig. 5.1). [The 14-d isotherm was reported previously (KELMERS 1985a) and is given on Fig. 5.1 for purposes of comparison.] In the absence of basalt, the control experiments with GR-2 indicate lower concentration limits as indicated by the solution concentration of uranium at the highest initial concentration of uranium in Fig. 5.2. In this figure, a comparison is given between final solution concentrations for the control experiments and for the sorption tests with basalt. For the two higher uranium concentrations, the final control solutions contained less uranium than solutions with basalt.

For the GR-4/Cohasset system under anoxic conditions at 60°C , considerably lower concentration limits were observed. After 14 d the limit was about 3×10^{-6} mol/L, and after 50 d the limit was about 8×10^{-7} mol/L (KELMERS 1985a). The limits for the two systems vary by about a factor of thirty. At this time, it is not possible to give the reasons for this variation. Extensive studies of the equilibrium solubility of sodium boltwoodite and of the kinetics of the precipitation reaction would be necessary for a complete understanding of this reaction. It is possible, for example, that the equilibrium solubilities of sodium boltwoodite in GR-2 and GR-4 might

be similar, but that the kinetics of precipitation from solutions of GR-2 are much slower than those from GR-4. The lower concentration limits in the GR-2 control solutions shown in Fig. 5.2 compared to the solutions with basalt could also be a kinetic effect in that the presence of basalt might slow the precipitation reaction. Therefore, it would appear from our data that the equilibrium and kinetic properties of sodium boltwoodite formation should be investigated in more detail.

Information from the three candidate repository site projects (KERRISK 1985; EARLY 1985; ONWI 1984) suggests that the DOE will utilize the geochemical computer code EQ3/6 alone, or in conjunction with other available geochemical models, to help evaluate: (1) geochemical conditions within the geologic setting, disturbed zone, and waste package; (2) sorption and solubility/speciation processes; and (3) the alteration of host rock, packing/backfill materials, and waste package components. Therefore, to improve the capability of our projects to provide the NRC with independent evaluations of geochemical information obtained by the DOE, we have brought the EQ3/6 software package (WOLERY 1979) on-line at ORNL to supplement our existing available codes (e.g., WATEQ, MINTEQ, PHREEQE, GEOCHEM).

Version 3230B of the EQ3/6 software package (with instructions for upgrading EQ6U21 to EQ6U23) was implemented and tested on the ORNL computer system this quarter. In addition to the EQ3/6 code and its required supplementary code EQLIB, the codes MCRT and EQTL were also brought up on the ORNL computer system. MCRT and EQTL provide for the compilation, documentation, and management of thermodynamic data files required to run EQ3/6. The successful conversion of the codes was confirmed by running the test cases for each code that was included as part of the software package. However, a detailed evaluation of the codes will require running several test cases that test the range of input options for each code.

An improved version of the EQ3/6 software package is scheduled to be released near the end of 1985 (WOLERY, personal communication). When available, this new version of EQ3/6 (as well as other versions of existing codes, or new

codes) will be integrated into our current suite of codes. In this way, we can maintain an up-to-date capability to support the NRC evaluation of DOE geochemical information used to support the assessment of repository performance.

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