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August 17, 1984

Mr. Everett A. Wick  
High-Level Waste Licensing Management Branch  
Division of Waste Management  
Office of Nuclear Material Safety &  
Safeguards  
Mail Stop 623-SS  
U. S. Nuclear Regulatory Commission  
Washington, D. C. 20555

WM-RES

WM Record File

A3164

A3167

ZNL

WM Project 10, 11, 16

Docket No.

PDR

LPDR (B.U.S)

Distribution:

X Wick

X LITTE

(Return to WM, 623-SS)

Dear Mr. Wick:

This is the monthly management report for the month of July for the programs entitled, "Review of DOE Waste Package Program," FIN A-3164 and "Waste Package Verification Tests," FIN A-3167. Included are the monthly highlight letters for the aforementioned programs. The breakdown of costs by task for each FIN is given on the attached computer summary sheets. Projections of costs by task for each FIN are given for FY 1984.

The labor, travel and overhead expenses associated with non-programmatic travel incurred by personnel assigned to NMSS FIN's for FY 1984 are as follows:

Costs (\$)

	<u>Month</u>	<u>Year-To-Date</u>
October	\$1,512.50	\$1,512.50
November	0	1,512.50
December	1,544.69	3,057.19
January	1,402.73	4,459.92
February	1,050.16	5,510.08
March	771.78	6,281.86
April	0	6,281.86
May	1,369.96	7,651.82
June	1,481.83	9,133.65
July	1,633.37	10,767.02

If there are any questions regarding format, distribution, or budget reporting, please contact Mr. A. J. Weiss, Administrative Technical Assistant, FTS 666-4473.

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PDR WMRES EXIBNL  
A-3164 PDR

Sincerely yours,

Walter Y. Kato  
Deputy Chairman

WYK/jw

Enclosures

- cc: C. Beckwith, NRC
- R. S. Brown, NRC
- R. E. Browning, NRC
- M. S. Davis, BNL
- D. Mattson, NRC
- D. C. Schweitzer, BNL

- P. Soo, BNL
- A. J. Weiss, BNL
- Document Control Center

REVIEW OF DOE WASTE PACKAGE PROGRAM  
(FIN/189a No.: A-3164)

P. Soo  
M. S. Davis

Monthly Letter Report, July, 1984  
Published: August 1984

Nuclear Waste Management Division  
D. G. Schweitzer, Head  
Brookhaven National Laboratory  
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## Task 1 - Review of DOE Waste Package Program

### Subtask 1.1 - Salt Repository Packing Material Program (E. Veakis)

This effort has been completed. A draft of the Final Report has been sent to NRC for review and comment.

### Subtask 1.2 - Granite Repository Program (E. Veakis)

This effort has been completed. A draft of the Final Report has been sent to NRC for review and comment.

### Subtask 1.3 - Spent Fuel Evaluation (E. Gause)

This task is concerned with the radionuclide containment and controlled release capabilities of spent fuel cladding in a repository environment. A draft report for this task is being prepared.

In the containment analysis, stress-corrosion cracking (SCC) and uniform corrosion of zirconium alloys have been evaluated as possible modes for breach of containment by the cladding.

Zircaloy specimens stressed in tension may crack in the presence of aggressive gaseous species, such as iodine, metal iodides ( $FeI_2$ ,  $AlI_3$ ) and cadmium at temperatures commonly found in water-cooled reactor cladding ( $\approx 300^\circ C$ ). It remains unclear what the operative mechanisms are in the laboratory tests and, more importantly, what the operative mechanisms are in a reactor environment, or what the identity of the responsible species is. Different mechanisms with different gas pressure and stress dependencies have been reported.

In the current work the main concern is that SCC failure processes which are not present during the above-ground storage period for spent fuel may become active in the repository due to the resumption of temperatures in the  $300-375^\circ C$  range, and the corresponding increase in gas pressure inside the fuel-cladding gap. We estimate that there would not be sufficiently high hoop stresses in the cladding to initiate cracks when compared to stresses causing failure in laboratory tests. However, inhomogeneities and defects in the cladding may provide enough localized stress to permit SCC to occur if a sufficient amount of the corrosive substance is present.

Two tables presented in the June Monthly Report showing the amount of gas inside fuel rods contained typographical errors. Corrected versions are given in Tables 1 and 2.

With regard to uniform corrosion, a review of published data reveals that weight gains at temperatures ranging from  $250^\circ C$  to  $400^\circ C$  in air, steam, brine, basaltic water, and high-pressure coolant would correspond to losses in cladding thickness in 300 years ranging from 0.008 to 1.2 mm depending on the test conditions. The typical cladding thickness is 0.6-0.8 mm. However, it is felt that a significant amount of time will be required to breach the overpack

Table 1. Amount and activity of gases produced in a BWR fuel rod irradiated to an average burnup of 27,500 MWd/MTU as a function of age.<sup>a</sup>

Gas -	At Discharge		300 Years After Discharge	
	Weight (g)	Activity (Ci)	Weight (g)	Activity (Ci)
H <sub>2</sub>	2.153E-04	2.076E00( <sup>3</sup> H)	9.792E-12	9.440E-08( <sup>3</sup> H)
He	7.006E-04 <sup>b</sup>	---	3.133E-02 <sup>b</sup>	---
Cl <sub>2</sub>	9.731E-04 <sup>c</sup>	3.124E-05( <sup>36</sup> Cl)	9.722E-04 <sup>c</sup>	3.121E-05( <sup>36</sup> Cl)
Br <sub>2</sub>	5.257E-02 <sup>d</sup>	N.R. <sup>f</sup>	5.259E-02 <sup>d</sup>	N.R. <sup>f</sup>
Kr	8.892E-01 <sup>d</sup>	2.244E01( <sup>85</sup> Kr)	8.317E-01 <sup>d</sup>	8.598E-08( <sup>85</sup> Kr)
I <sub>2</sub>	5.805E-01 <sup>d</sup>	7.527E-05( <sup>129</sup> I)	5.705E-01 <sup>d</sup>	7.619E-05( <sup>129</sup> I)
Xe	1.294E01 <sup>d</sup>	N.R. <sup>f</sup>	1.272E01 <sup>d</sup>	N.R. <sup>f</sup>
Cs	6.784E00 <sup>d,e</sup>	3.230E02( <sup>134</sup> Cs)	3.676E00 <sup>d,e</sup>	7.619E-05( <sup>135</sup> Cs)
		1.035E-03( <sup>135</sup> Cs)		2.502E-01( <sup>137</sup> Cs)
		2.503E02( <sup>137</sup> Cs)		
Rn	3.944E-15 <sup>d</sup>	4.754E-11( <sup>222</sup> Rn)	3.952E-12 <sup>d</sup>	6.084E-07( <sup>222</sup> Rn)
TOTAL		5.978E02 <sup>g,h</sup>		2.504E-01 <sup>i</sup>

<sup>a</sup>This table is adapted from ORIGEN-2 calculated values presented in ORNL/TM-6008 (1977) for a 8 x 8 assembly assumed to contain 63 rods. All volatile materials have not been included. For example, ≈8.0E-05 moles of C-14 are produced as fission and activation products. Some of this may be present in the form of gaseous compounds.

<sup>b</sup>This is the amount of He produced. It does not include He added during manufacture.

<sup>c</sup><sup>36</sup>Cl is formed as an activation product. Weight shown is only for <sup>36</sup>Cl.

<sup>d</sup>Weight shown includes weight of all isotopes present.

<sup>e</sup>Cs is a volatile fission product. The vapor pressure P (in torr) of Cs in the range 200-350°C can be calculated using the formula:

$$\log_{10} P = \frac{3833}{T(\text{in K})} + 6.949.$$

<sup>f</sup>N.R. indicates not reported.

<sup>g</sup>This activity represents ≈0.15% of the inventory at discharge.

<sup>h</sup>Total activity at 10 years from discharge due to these elements is 2.230E02 Ci. This represents ≈23.2% of the inventory 10 years after discharge.

<sup>i</sup>This activity represents ≈2.30% of the inventory 300 years after discharge.

Table 2. Amount and activity of gases produced in a PWR fuel rod irradiated to an average burnup of 33,000 MWd/MTU as a function of age.<sup>a</sup>

Gas	At Discharge		300 Years After Discharge	
	Weight (g)	Activity (Ci)	Weight (g)	Activity (Ci)
H <sub>2</sub>	1.918E-04	1.849E00( <sup>3</sup> H)	8.722-E12	8.408E-08( <sup>3</sup> H)
He	5.103E-04 <sup>b</sup>	---	2.781E-02 <sup>b</sup>	---
Cl <sub>2</sub>	7.828E-04 <sup>c</sup>	2.513E-05( <sup>36</sup> Cl)	7.828E-04 <sup>c</sup>	2.511E-05( <sup>36</sup> Cl)
Br <sub>2</sub>	4.897E-02 <sup>d</sup>	N.R. <sup>f</sup>	4.898E-02 <sup>d</sup>	N.R. <sup>f</sup>
Kr	8.348E-01 <sup>d</sup>	2.141E01( <sup>85</sup> Kr)	7.799E-01 <sup>d</sup>	8.201E-08( <sup>85</sup> Kr)
I <sub>2</sub>	5.422E-01 <sup>d</sup>	6.971E-05( <sup>129</sup> I)	5.309E-01 <sup>d</sup>	7.078E-05( <sup>129</sup> I)
Xe	1.199E01 <sup>d</sup>	N.R. <sup>f</sup>	1.199E01 <sup>d</sup>	N.R. <sup>f</sup>
Cs	6.221E00 <sup>d,e</sup>	3.494E02( <sup>134</sup> Cs)	3.277E00 <sup>d,e</sup>	---
		8.387E-04( <sup>135</sup> Cs)		8.402E-04( <sup>135</sup> Cs)
		2.346E02( <sup>137</sup> Cs)		2.345E-01( <sup>137</sup> Cs)
Rn	2.632E-15 <sup>d</sup>	2.879E-11( <sup>222</sup> Rn)	3.636E-12 <sup>d</sup>	5.598E-07( <sup>222</sup> Rn)
TOTAL		6.073E02 <sup>g,h</sup>		2.354E-01 <sup>i</sup>

<sup>a</sup>This table is adapted from ORIGEN-2 calculated values presented in ORNL/TM-6008 (1977) for a 15 x 15 assembly assumed to contain 204 rods. All volatile materials have not been included. For example, =6.0E-05 moles of C-14 are produced as fission and activation products. Some of this may be present in the form of gaseous compounds.

<sup>b</sup>This is the amount of He produced. It does not include He added during manufacture.

<sup>c</sup><sup>36</sup>Cl is formed as an activation product. Weight shown is only for <sup>36</sup>Cl.

<sup>d</sup>Weight shown includes weight of all isotopes present.

<sup>e</sup>Cs is a volatile fission product. The vapor pressure P (in torr) of Cs in the range 200-350°C can be calculated using the formula:

$$\log_{10} P = \frac{3833}{T(\text{in K})} + 6.949.$$

<sup>f</sup>N.R. indicates not reported.

<sup>g</sup>This activity represents =0.13% of the inventory at discharge.

<sup>h</sup>Total activity at 10 years from discharge due to these elements is 2.107E02 Ci. This represents =23.6% of the inventory 10 years after discharge.

<sup>i</sup>This activity represents =2.44% of the inventory 300 years after discharge.

and, at that time, the temperature of the spent fuel cladding will be substantially decreased. Since it is estimated that an order of magnitude decrease in corrosion rate occurs with every 50°C decrease in temperature, it is not likely that the cladding will fail by uniform corrosion processes during the 300-1000 year containment period.

Progress to date is shown in Figure 1.

Response to NRC Comments on Trip Report to the American Ceramic Society Meeting in Pittsburgh, April 30-May 2, 1984 (T. Sullivan)

The NRC Program Manager commented on the subject trip report in the May Monthly Letter Report and noted the inconsistency in measuring pH at 25°C when the expected ambient temperature in the proposed BWIP repository is 55°C. This discrepancy was not discussed in the paper reviewed but it seems that measurements were taken at 25°C for experimental convenience. Data given in the BWIP Site Characterization Report, reproduced below as Figure 2, show that increasing the temperature by 30°C causes a pH decrease of about 0.5, assuming that the pH is controlled by the dissociation of silicic acid.

Task 2 - General Technical Assistance

WAPPA Code Evaluation (C. Pescatore, T. Sullivan, C. Sastre)

A draft of the final report on WAPPA code evaluation has been completed and has been internally reviewed. After retyping, a copy will be sent to NRC for comment.

Data File for Waste Package Evaluation (C. Sastre)

Work continued in specifying a procedure to establish a data file for waste package performance information.

For the purpose of defining the data structure for the data base, a complicated example was selected from the leaching literature (PNL-3172, 1983).

In testing the reduction of the Westsik results to a form suitable for machine storage, it was found that attempting to reduce each data set to a single record would lead to an awkward and complicated structure. Therefore, it was decided to assign to each data set a group of files which were cross referenced. A main file will be used with author and title information, as well as experimental conditions, a comment file with descriptive information, and one or more data files in the form of number arrays.

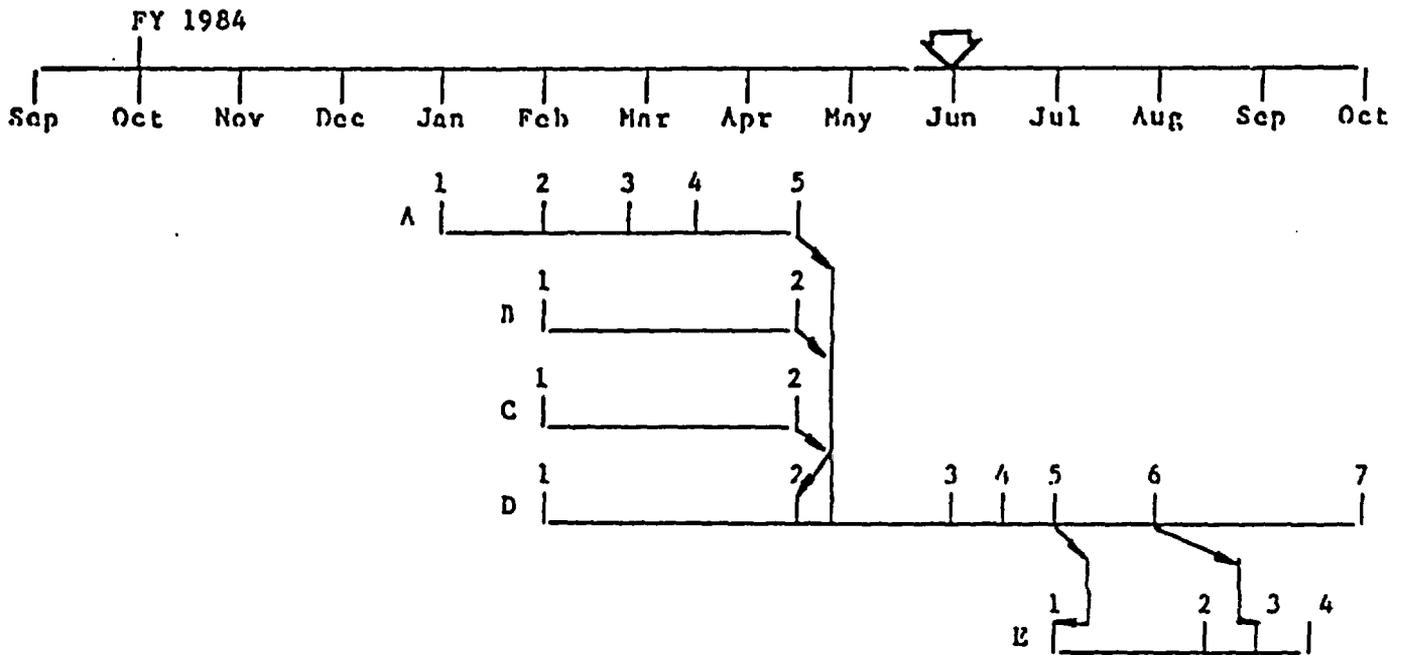
Reference

PNL-3172, "High Temperature Leaching of an Actinide-Bearing Simulated High Level Waste Glass," J. H. Westsik, Jr. and others, Pacific Northwest Laboratory, 1983.

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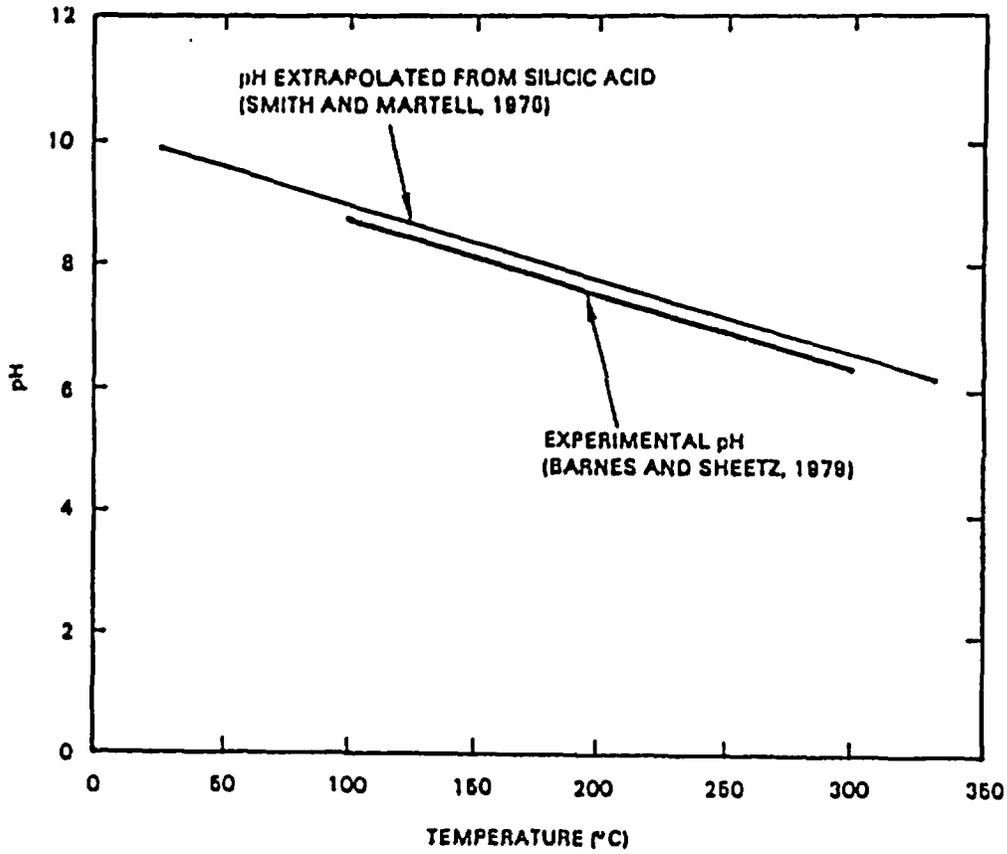
Figure 1

Milestone Chart for FIN A-3164  
Subtask 1.3 - Spent Fuel Evaluation



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Milestone Chart for FIN A-3164  
Subtask 1.3 - Spent Fuel Evaluation

- A1-A2 Collection of data to update previous FIN A-3164 effort on near-field repository conditions.
- A2-A3 Selection of relevant near-field environmental conditions and preparation of a brief account of the rationale for their selection.
- A3-A4 Typing and QA of list of environmental conditions and discussion of selection rationale.
- A4 Submission of list and report to NRC (3-15-84).
- A4-A5 Discussions with NRC on submitted report.
- A5 NRC approval of environmental conditions (on or about 4-15-84).
- B1-B2 Collection of information on failure rates for nuclear fuel.
- C1-C2 Collection of information on in situ testing of spent nuclear fuel.
- D1-D2 Collection of information on cladding material degradation and failure with emphasis on Zircaloy in repository-like conditions.
- D2 On receipt of NRC approval for environmental conditions, begin analysis degradation and failure modes of cladding materials with emphasis on corrosion of Zircaloy. Information on spent fuel failure rates and on in situ testing will also be included in the analysis.
- D3 Completion of cladding containment analysis. Preliminary Draft Report submitted for typing.
- D4-D5 Perform QA of preliminary Draft Report.
- D5 Draft Report submitted to NRC (7-1-84).
- D6 Receipt of NRC comments on Draft Report.
- D6-D7 Response to NRC comments. Revision of Draft Report.
- D7 Final Report submitted to NRC (9-30-84).
- E1-E2 Using information from Draft Report begin preparation of Program Plan tests to confirm the analysis in the Draft Report.
- E3-E4 QA of Program Plan. Comments from NRC on Draft Report will be considered during the QA.
- E4 Submission of Program Plan to NRC for design of tests to confirm analysis of Draft Report (9-15-84)



RCPD107-66

Figure 2. Relationship between estimated pH resulting from the dissociation of silicic acid and pH measured in the Hanford basalt/groundwater system between 100° and 300°C.

REVIEW OF WASTE PACKAGE VERIFICATION TESTS  
(FIN/189a No.: A-3167)

P. Soo  
M. S. Davis

Monthly Letter Report, July 1984  
Published: August 1984

Nuclear Waste Management Division  
D. G. Schweitzer, Head  
Brookhaven National Laboratory  
Associated Universities, Inc  
Upton, NY 11973

Task 1 - Evaluation and Identification of Performance Verification Testing for Tuff Repository Packing Material (E. Veakis, H. Jain)

A draft of the final report sections pertaining to test methods for performance verification of tuff packing material for a spent fuel waste package has been completed and is being typed at the time of writing. Many of the recommended test methods are similar to those identified in earlier BNL work for the bentonite/clay type basalt repository packing material. These include tests for mineralogical stability, mechanical stability, groundwater migration and radionuclide sorption under aqueous conditions. Such test methods are applicable for the period when the tuff repository has sufficiently cooled to permit ingress of liquid groundwater.

As described in previous Monthly Letter Reports, the groundwater initially approaching the waste package may be highly concentrated in dissolved salts. Therefore, testing is recommended to evaluate the range of groundwater chemical composition which might affect the alteration or sorption properties of the packing.

Additional test methods are also outlined for packing material evaluation for the initial period after repository closure when steam conditions will prevail around the waste package for several hundred years. These include mineralogical stability and radionuclide sorption and migration under such steam conditions.

Figure 1 shows progress to date in this effort.

The NRC Program Manager has requested clarification of several items in this task with respect to BNL's suggested test methodologies for the tuff repository program described in the May Monthly Letter Report. One concerns the logic for cooling a crushed tuff test column which would be used to evaluate solute concentration changes in J-13 water (caused by precipitation of dissolved salts at the boiling point as liquid water changes to steam) and the subsequent resolution of these salts as the repository cools and liquid water passes over the deposited materials. The description in the May Report did not mean to indicate that the column would be cooled to room temperature to check the chemistry and pH of the modified water. The test would involve cooling the column to a temperature which was just below the boiling point so that the water which has just passed through the region of prior salt deposition may be collected and characterized. This water can be characterized for pH temperature using available instrumentation. The high temperature ionic concentration of the water can be measured by adding a known quantity of acid to keep the constituents in solution as it is cooled to room temperature for analysis. The chemistry and pH of the water should then be a close representation of the groundwater at temperatures just below the boiling point. Thus, one would expect that this modified water composition will be representative of water that first contacts the waste package when the boiling point isotherm intersects it. Based on work reported by LLNL, the waste package will encounter liquid water during the 200-800 year period after repository closure.

Another comment by the NRC Program Manager concerned the selection of an appropriate tuff-surface-area to water-volume ratio for the column tests. Such a selection is very difficult since the water will be percolating through the tuff horizons via interconnected pores and fractures. These cannot be easily simulated in the laboratory using small-scale apparatus. Therefore, crushed tuff columns would have to be used in which the tuff particles would typically be in the 0.5-1.0 cm range. The logic for this is to use a large tuff surface area to allow easier equilibrium with the percolating solution. Water chemistries subsequently measured would, therefore, represent a bounding condition which would probably be similar to an actual repository condition where water percolation rates are slow, thereby encouraging water/rock equilibration.

A final NRC comment concerned the definition of thermally-altered tuff packing material which could be used for packing-water-radionuclide interaction tests. Since the container temperature could be as high as 250°C (Hockman, J. N, 1984) long-term aging studies (several years) may be carried out at this temperature to quantify mineralogic changes, and to prepare samples for interaction tests. Samples may be periodically removed to check when alteration is effectively complete. Higher and lower temperature tests may also be carried out so that there is some understanding of the effects of temperature and time on the nature and degree of alteration. In effect, time-temperature-alteration diagrams could be prepared to serve as a basis for estimating packing material mineralogy at the waste container interface as a function of time. Such diagrams may enable these mineralogic compositions to be simulated using high-temperature, short-time alteration procedures.

#### Reference

Hockman, J. N. and W. C. O'Neil, "Thermal Modeling of Nuclear Waste Package Designs for Disposal in Tuff," Waste Management '84, Tucson, Arizona, 1984.

#### Assessment of Problems Associated With the Use of Constant Distribution Coefficients (Kd's) for Radionuclide Sorption Calculations (T. Sullivan)

A review was conducted on the limitations associated with the use of constant Kd values for calculating radionuclide sorption behavior on geologic materials. These limitations should be recognized in order to assess potential problems should DOE elect to use such calculational procedures in their work. Appendix 1 is a summary of BNL's review.

### Task 2 - General Technical Assistance

#### DOE Environmental Assessment (EA) Report Review

Working drafts of the DOE EA reports for salt, basalt, and tuff repositories have been received via the NRC Program Manager. Staff at BNL are currently reviewing the sections pertaining to waste package performance and collecting cited references for additional study.

Work is currently being initiated to establish in detail the quantity and relevance of available data on waste package performance to determine whether performance assessment calculations given in the EA reports are valid, and if additional data will be needed from DOE to support the EA findings on waste package performance.

Comparison of Leaching Data on SRL-131 Composite Glass Reported by MCC and SRL (P. Soo)

At the request of NRC a comparison was made of leaching data on SRL-131 waste glass obtained from the Materials Characterization Center and Savannah River Laboratory. Appendix 2 gives the results of this study.

Task 3 - Stress Corrosion Cracking Studies on Container Materials for a Tuff Repository (H. Jain, P. Soo)

The C-ring specimens from 304L, 316L and 321 stainless steel and Incoloy 825 as-received tubing are currently being fabricated. Sensitization of these materials was achieved by heat treatment at 600°C for 100 hours followed by furnace cooling. The oxidation resulting from this heat treatment was removed by polishing the surfaces.

Preliminary tests have been completed to determine the relationship between the stress at the C-ring apex as a function of the strain applied by the sample's nut and bolt loading procedure. This information is needed to stress the specimens to the predetermined value.

Although a chemical analysis of reference J-13 groundwater has been specified by DOE subcontractors, there is no procedure specified for its preparation in the laboratory. The J-13 water used by NNWSI is obtained from the actual site rather than prepared artificially. Therefore, a solution preparation procedure is being developed at BNL. For the ten times concentrated groundwater, it should be mentioned that some of the species (e.g. SiO<sub>2</sub>, divalent ions) may be already present near their saturation limit at room temperature. However, further concentration may be possible at the test temperature to be used (100°C). Therefore, we plan to add ten times the quantity of salts needed for reference J-13 groundwater and let the salts equilibrate at 100°C. The final solution will be analyzed for its chemical composition.

All necessary apparatus for this study has been received from the vendors and the first batch of samples for the 12-month corrosion test will be fabricated in early August. Testing will be initiated shortly thereafter.

Task 4 - Determination of Local Conditions Appropriate for Low-Carbon Steel High Level Waste Containers in a Basalt Repository (E. Gause)

The objective of this program was to determine the chemical environment that will be present within high level nuclear waste packages emplaced in a basalt repository. For this purpose, low-carbon 1020 steel (a current BWIP reference container material), synthetic basaltic groundwater and a mixture of bentonite and basalt were exposed in an autoclave to a specific repository condition after sealing (150°C, 1500 psi) in a gamma radiation environment with a dose rate of  $3.8 \pm 0.5 \times 10^4$  rad/h. The experiment consisted of three test phases.

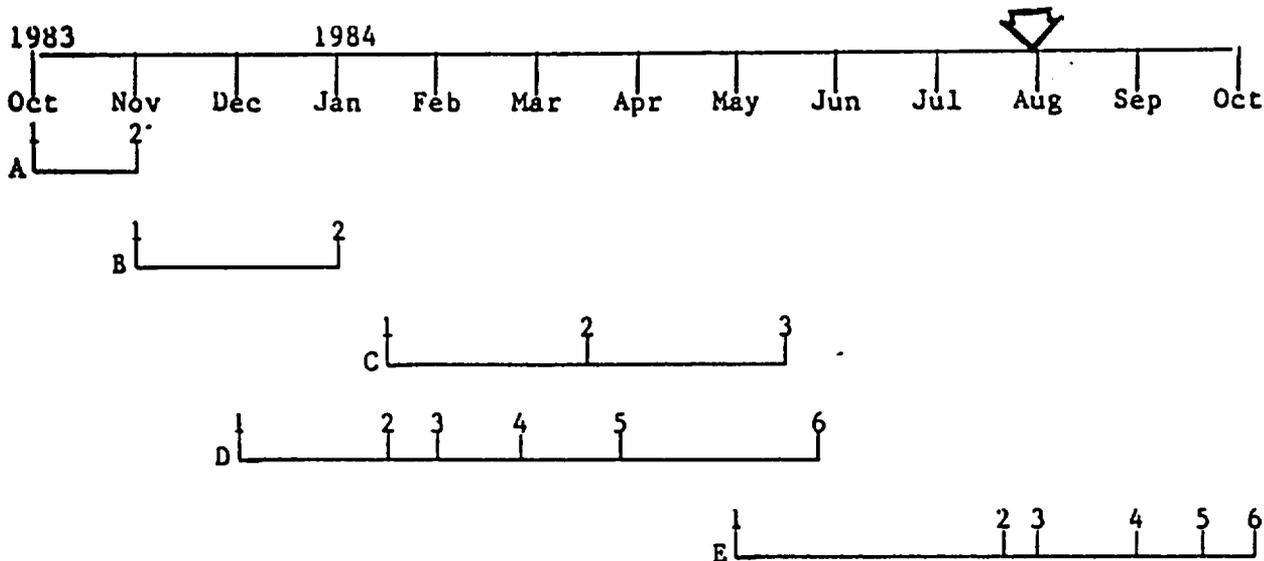
The Phase I test involved a two-month irradiation test in an argon environment. The Phase II test is a similar test in a methane environment in the presence of radiation. These two tests were followed by a Phase III control test which is similar to the Phase II study but which was conducted in the absence of radiation.

The draft report incorporating results of the three phases of the test was submitted to the NRC on June 18, 1984. Comments have not been received and no work has been done on this Task during July. Progress to date is shown in the attached Milestone Chart, Figure 2.

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Figure 1

Milestone Chart for FIN A-3167  
Task 1 - Test Verification Report on Crushed Tuff Packing Material



LEGEND

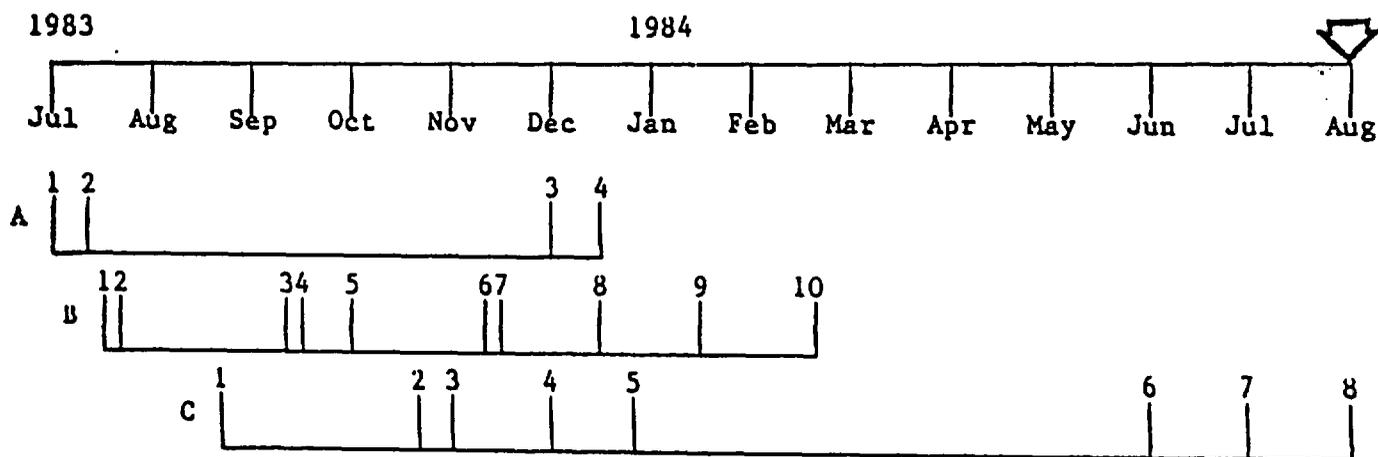
- A1-A2 Obtain from prior FIN A-3164 reports information on tuffaceous repository environmental conditions. These will include temperature, litho-static/hydrostatic pressures, water flow rates and chemistry, Eh/pH as a function of time during the pre- and post-closure periods. Update with new data from DOE tuff repository program.
- B1-B2 Determine through review and analysis the physical/chemical changes in crushed tuff packing arising from the thermal/irradiation environment, and interactions with waste glass/spent fuel, container corrosion products, and groundwater radiolysis species. Specify conditions for crushed tuff packing materials tests.
- C1-C2 Review available ASTM, MCC and other packing material test methodologies to ascertain procedures for measuring the containment capabilities of crushed tuff packing.
- C2-C3 Review available ASTM, MCC and other packing material test methodologies to ascertain procedures for measuring the controlled release capabilities of crushed tuff packing.
- D1-D2 Write section on testing parameters pertinent to testing of crushed tuff packing material, completing activities A1-A2 and B1-B2.
- D3 Complete typing of Draft Report.

- D3-D4 Complete QA and review of Draft Report, revise manuscript and submit Draft Report to NRC on 2-20-84.
- D5 Receive written comments on Draft Report from NRC.
- D5-D6 Incorporate NRC comments into Final Biannual Report and submit camera-ready copy to NRC on 5-20-84.
- E1-E2 Write section on recommended test methodologies pertinent to measuring the ability of crushed tuff packing to meet the NRC containment and controlled release criterion. Specify any inadequacies in currently available tests and detail improved tests which may be used to assess the performance of crushed tuff packing.
- E3 Complete typing of Draft Report.
- E3-E4 Complete QA review of Draft Report, revise manuscript and submit Draft Report to NRC on 8-20-84.
- E5 Receive written comments on Draft Report from NRC.
- E5-E6 Incorporate NRC comments into Final Biannual Report and submit camera-ready copy to NRC on 9-30-84.

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Figure 2

Milestone Chart for FIN A-3167  
 Task 4 - Effort on Determination of Local Corrosion Conditions  
 Appropriate for Low Carbon Steel HLW Containers in a Basalt Repository  
 (Revised 4/2/84; supersedes previous milestone charts)



LEGEND

- A1 Submit addendum to test specifications (submitted 5/10/83) to incorporate use of methane in a three-phase test system to NRC for approval (6/30/83).
- A2 Receive NRC approval for addendum to test specifications (7/8/83).
- A3 Submit test specification for the Phase III experiment to the NRC (12/20/83).
- A4 Receive NRC approval to conduct Phase III experiment (12/30/83).
- B1 Begin two-month Phase I experiment in radiation environment using argon overpressure (7/11/83) (EXPERIMENT WILL BE TERMINATED SHOULD A SAFETY HAZARD DEVELOP).
- B2 Begin examination of system components for pre-test condition (7/15/83).
- B3
  - a. Complete Phase I experiment (9/9/83).
  - b. Begin measurements of solution parameters (pH, DO, Cl<sup>-</sup>, SO<sub>4</sub><sup>-2</sup>, Fe) and gas analyses from Phase I experiment (9/9/83).
- B4 Begin two-month Phase II experiment in radiation environment using a methane overpressure (9/12/83) (EXPERIMENT WILL BE TERMINATED SHOULD A SAFETY HAZARD DEVELOP).

- B5 a. Complete measurements of solution parameters (pH, DO, Cl<sup>-</sup>, SO<sub>4</sub>-2, Fe) and gas analyses from Phase I experiment (9/30/83).
- B5 b. Complete examination of steel surfaces and packing material mineralogical content for pre-test and post-test samples from Phase I experiment (9/30/83).
- B6 a. Complete Phase II experiment (11/11/83).
- B6 b. Begin measurements of solution parameters (pH, DO, Cl<sup>-</sup>, SO<sub>4</sub>-2, Fe) and gas analyses from Phase II experiment (11/11/83)
- B7 Begin two-month Phase III experiment in a non-radiation environment using a methane overpressure (11/14/83) (EXPERIMENT WILL BE TERMINATED SHOULD A SAFETY HAZARD DEVELOP).
- B8 a. Complete measurements of solution parameters (pH, DO, Cl<sup>-</sup>, SO<sub>4</sub>-2, Fe) and gas analyses from Phase II experiment (12/16/83).
- B8 b. Complete examination of steel surfaces and packing material mineralogical content for pre-test and post-test samples from Phase II experiment (12/16/83).
- B9 a. Complete Phase III experiment (1/13/84).
- B9 b. Begin measurements of solution parameters (pH, DO, Cl<sup>-</sup>, SO<sub>4</sub>-2, Fe) and gas analyses from Phase III experiment (1/13/84).
- B10 a. Complete measurements of solution parameters (pH, DO, Cl<sup>-</sup>, SO<sub>4</sub>-2, Fe) and gas analyses from Phase III experiment (2/17/84).
- B10 b. Complete examination of steel surfaces and packing material mineralogical content for pre-test and post-test samples from Phase III experiment (2/17/84).
- C1 Issue a Draft Interim Report on findings to date in Phase I experiment (8/17/83).
- C2 Issue a Draft Interim Report on findings to date in Phase II experiment (10/19/83).
- C3 Submit Draft Final Report on Phase I experiment to NRC for comment (10/31/83).
- C4 Receive NRC comments on Draft Final Report on Phase I experiment (12/1/83).
- C5 Issue a Draft Interim Report on findings to date in Phase III experiment (12/22/83).
- C6 Submit Draft Final Report on Phase I, Phase II and Phase III experiments to NRC for comment (5/31/84).
- C7 Receive NRC comments on Draft Final Report on Phase I, Phase II and Phase III experiments (6/29/84).
- C8 Submit Final Report on Phase I, Phase II and Phase III experiments to NRC for publication (7/31/84).

BROOKHAVEN NATIONAL LABORATORY  
M E M O R A N D U M

DATE: June 18, 1984  
TO: Files  
FROM: T. Sullivan  
SUBJECT: Potential Problems with Using the Constant Kd Approach in Radionuclide Transport Calculations in the Near Field of a Nuclear Waste Repository.

1. INTRODUCTION

It is well known that nuclides released from the waste form into the contacting solution may be adsorbed by the surrounding packing materials or the host rock. This will decrease the transport of these nuclides through the repository. One approach used to account for this effect is to reduce the transport parameters through division by a retardation coefficient, R, defined as:

$$R_i = 1 + \frac{\rho}{n} \left( \frac{dS}{dC} \right)_i, \quad (1)$$

where  $\rho$  is the density of the solid;  $n$  is the porosity of the solid;  $(dS/dC)_i$  is the ratio of the time rate of change of the amount of mass sorbed,  $S$ , on the solid versus the time rate of change in solution concentration,  $C$ ; and the subscript  $i$  denotes the "i"th nuclide. It is emphasized that  $(dS/dC)$  is zero when steady-state behavior is obtained and therefore retardation does not influence steady-state transport.

Equation (1) illustrates that the greater the rate of uptake of solution species, the greater the retardation of transport. A typical uptake curve is presented in Figure 1. At low solution concentrations the amount sorbed increases linearly with concentration. As the solid approaches its saturation limit,  $S_{max}$ , with respect to the given solution species the rate of uptake and therefore retardation decreases. This curve is known as a sorption isotherm.

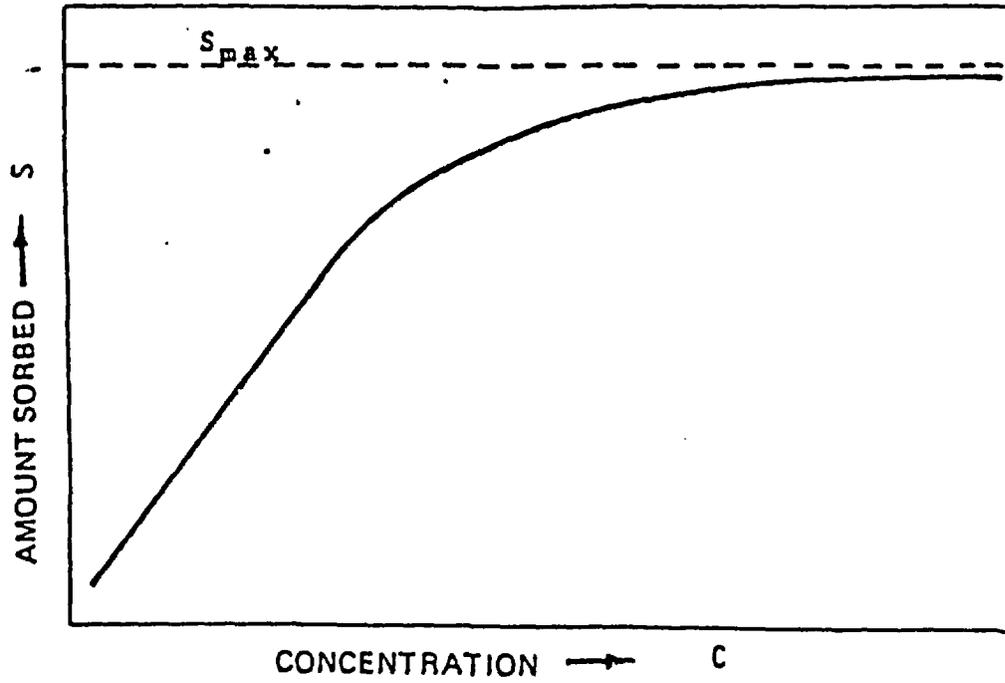


Figure 1. Typical partition of a solute between a porous solid and water (adapted from Ref.[1]).

To estimate the uptake of solution species, batch sorption experiments are conducted in which a solid is immersed in a solution initially containing a known concentration of a given species. After equilibrium is reached, the final solution concentration, C, can be measured and the amount sorbed on the rock can be inferred through a mass balance. The ratio of these two quantities is known as the distribution coefficient,

$$K_d = \frac{S}{C} . \quad (2)$$

For solution concentrations far below amount required to saturate the solid, this ratio is often found to be constant. In this case,

$$K_d = \frac{S}{C} = \frac{dS}{dC} , \quad (3)$$

and the distribution coefficient can be used directly in the expression for the retardation coefficient, Equation (1), to give

$$R = 1 + \frac{\rho}{n} K_d . \quad (4)$$

Currently, use of Equation (4) to represent retardation of species transport is the most common approach and has been used for BWIP calculations. [2] However, there are several issues that must be addressed before the constant  $K_d$  approach can be shown to be acceptable. Section 2 of this report will identify these issues. Section 3 will make recommendations on how to improve this method and will draw conclusions as to the applicability of the constant  $K_d$  approach.

## 2. LIMITATIONS OF THE CONSTANT $K_d$ APPROACH

The determination of a distribution coefficient,  $K_d$ , is an empirical attempt to combine the interactions of all of the complex sorption phenomena into a single parameter. In general, sorption onto a solid depends primarily on solution composition, Eh, pH, temperature, and solid composition. As any one of these parameters changes the distribution coefficient will change.

The near field of a waste repository is defined as the region in which substantial changes in temperature, pressure, and Eh are expected to occur due to the presence of the radioactive waste form. In the case of temperature, changes will continue for thousands of years. Thus, the  $K_d$  values measured in controlled isothermal conditions may not be relevant to near field repository conditions. Furthermore, the dependence of distribution coefficients on temperature is strong and not completely understood from a theoretical viewpoint. Also, because of experimental difficulties associated with determining distribution coefficients at temperatures greater than 100°C, the majority of data is at temperatures of 60°C or lower. However, the temperature in the near field will exceed 60°C because of the heat released from the waste. Since there are no reliable theoretical methods to extrapolate the distribution coefficients, more high temperature data is needed. It should be noted that data in the BWIP Data Package for Reference Solubility and  $K_d$  values<sup>[3]</sup> are based on measurements at 60°C and will not be relevant for calculations near the waste package.

Another problem with the  $K_d$ 's currently measured is that they are obtained from single component experiments. That is, the solution is spiked with only one species. Near the waste package, the solution will contain all of the actinides, rare earths, alkaline earths, and alkalis found in the waste form and many of these elements will be near their solubility limit. Under these conditions, it is not clear how the sorption of the different species will vary from sorption in a single component system. However, it is highly probable that it will be different. It is well known that solution chemistry influences the sorption process.

## 2.1 Evaluation of Interpretations of Experimental Data

An examination of the data from which  $K_d$  values are obtained illustrates the complexity of the sorption phenomena and allows more doubts about the validity of the constant  $K_d$  approach to form. Figures 2 - 5 are plots of the amount of material sorbed on various basalts from the BWIP site versus concentration in solution, a simulated BWIP groundwater. This data is taken from the report titled, "The Sorption Behavior of Selected Radionuclides on Columbia River Basalts." [4]

Figure 2 represents the data for uranium and examination of this log-log plot shows that  $K_d = S/C$  is not a constant and therefore should not be used in the retardation coefficient, Equation (1), in place of  $dS/dC$ .

The fact that

$$S \neq K_d C \quad (5)$$

for constant  $K_d$  was recognized by the researchers and they attempted to fit the data to an isotherm of the form

$$S = k C^N. \quad (6)$$

This is known as the Freundlich isotherm and has been widely used to fit sorption data. Taking the logarithm of both sides in Equation (6) gives:

$$\log S = \log k + N \log C. \quad (7)$$

Thus, on a log-log plot of  $S$  versus  $C$  the data should fall along a straight line of slope  $N$ . However, this is not the case for the uranium data and therefore the Freundlich isotherm is not appropriate for these data. Nevertheless, the Freundlich isotherm was used and the resulting fit was extremely poor. This is displayed in Table 1 which compares the measured amount of uranium sorbed on the basalt versus the predicted amount sorbed using the Freundlich isotherm. Differences as large as an order of magnitude between the two are found and differences of a factor of two are common.

Figure 2.  
 Uranium Sorption Data  
 T = 23°C, GRI Groundwater  
 Eh = -0.4V  
 page A-4 RHO-BWI-40-48

U - Umtanur Basalt  
 P - Panna Basalt  
 E - Fels Basalt  
 U<sub>i</sub> - Proposed Freundlich Isotherm for Umtanur Basalt Data.

$$S = 0.0203 C^{0.924}$$

Suggested Freundlich Isotherms

U -  $S = 0.0203 C^{0.924}$   
 P -  $S = 8.123 \times 10^{-3} C^{0.8195}$   
 E -  $S = 7.13 \times 10^{-4} C^{0.7343}$

S

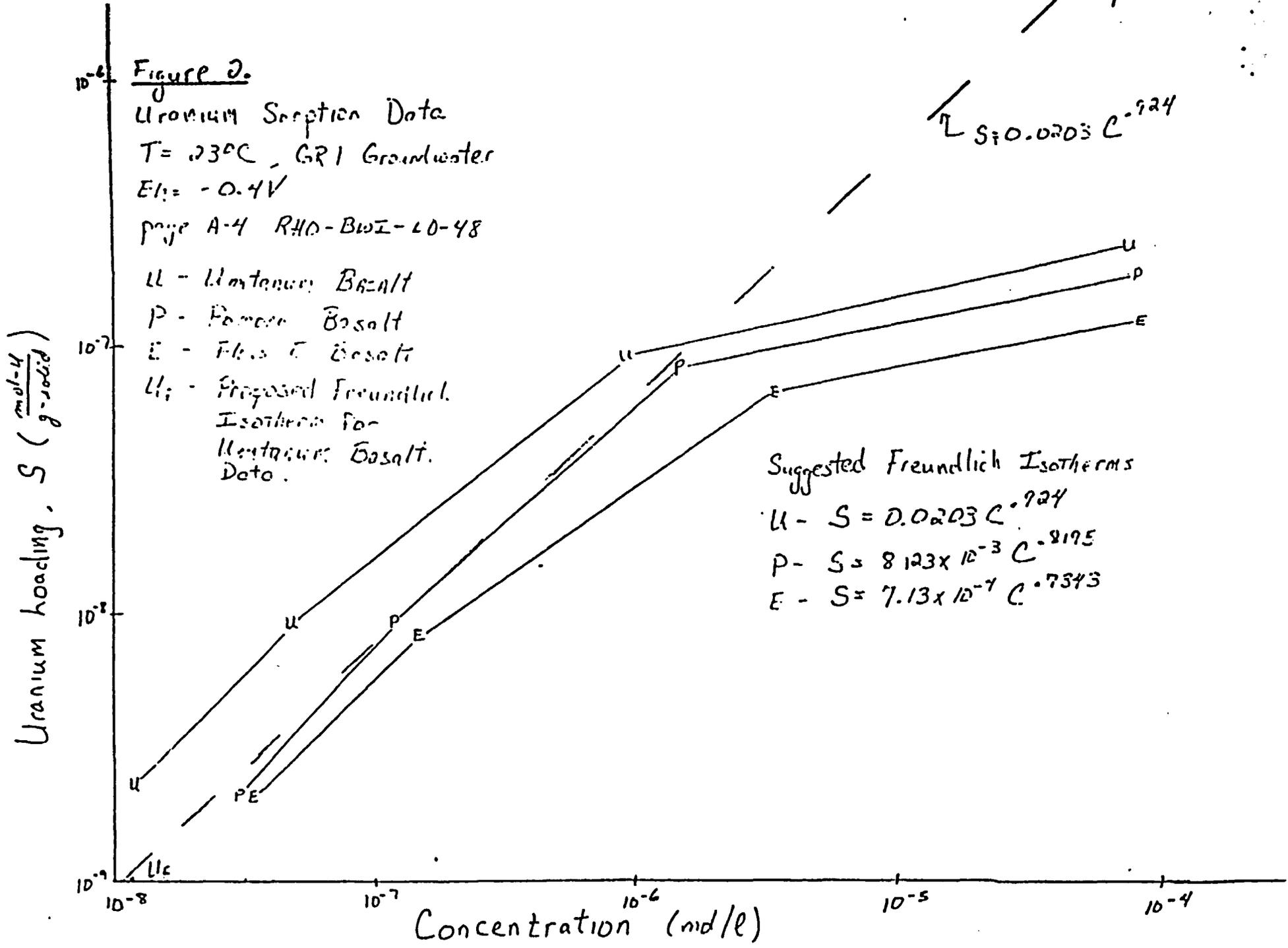


Table 1

Comparison of the measured amount sorbed,  $S_M$ , versus the predicted amount sorbed,  $S_F$ , using the Freundlich isotherm  $S_F = 0.0202877 C^{0.92369}$ . The data is for uranium being sorbed on uranium basalt at 23°C in GR-1 groundwater. Data from page A-4 and A-5 of RHO-BWI-LD-48.

$C$ (mol/l)	$S_M$ ( $\frac{\text{mol-U}}{\text{g-solid}}$ )	$S_F$ ( $\frac{\text{mol}}{\text{g-solid}}$ )	$(\frac{S_M - S_F}{S_M}) \times 100$
$7.74 \times 10^{-5}$	$2.26 \times 10^{-7}$	$3.23 \times 10^{-6}$	-1333
$9.69 \times 10^{-7}$	$9.03 \times 10^{-8}$	$5.66 \times 10^{-8}$	37
$4.57 \times 10^{-8}$	$9.54 \times 10^{-9}$	$3.37 \times 10^{-9}$	65
$1.23 \times 10^{-8}$	$2.33 \times 10^{-9}$	$1.0 \times 10^{-9}$	57

Figure 3 represents plutonium sorption data at two different temperatures. Again, Freundlich isotherms were used to fit the data. It is interesting to note the two curves follow basically the same trend but the differences in the fitting coefficients are very substantial. In fact, the fit at 60°C predicts the rate of uptake will increase faster than the solution concentration with increasing concentration whereas the opposite is true at 23°C. This effect may be caused by experimental error or it may be real over the concentration range in the experiment. The main point is that the fitting parameters are sensitive to the data. Therefore, small errors in the data may lead to large errors in the empirical fit. For this reason, the empirical fits should not be extrapolated outside the range of available data.

Figure 4 displays radium sorption data and shows the amount sorbed increased faster than the solution concentration. This trend held true for tests in umtanum basalt at either 23°C or 60°C in GR-2 simulated basaltic groundwater. This behavior is not understood and illustrates that sorption is a complex phenomena. The distribution coefficients for the data are presented in Table 2 and they show an increase of a factor of 5 between the high and low values. To gain a better understanding of the sorption of radium, data should be taken over a wider range of solution concentrations.

Figure 5 displays cesium sorption data and again the data does not follow a straight line. In this case, the researchers fit the data to a Dubinin-Radushkevich (DR) isotherm which has the form:

$$S = S_{\max} e^{-K(RT \ln(1+1/C))^2} \quad (8).$$

where:

S is the amount sorbed to the solid (mol/g-solid),

$S_{\max}$  is the maximum sorption capacity of the solid,

R is the universal gas constant,

T is the temperature in °K,

C is the equilibrium solution concentration ( $\frac{\text{mol}}{\text{l}}$ ), and

K is an empirical constant, greater than zero, which can be related to the mean energy of sorption.

6

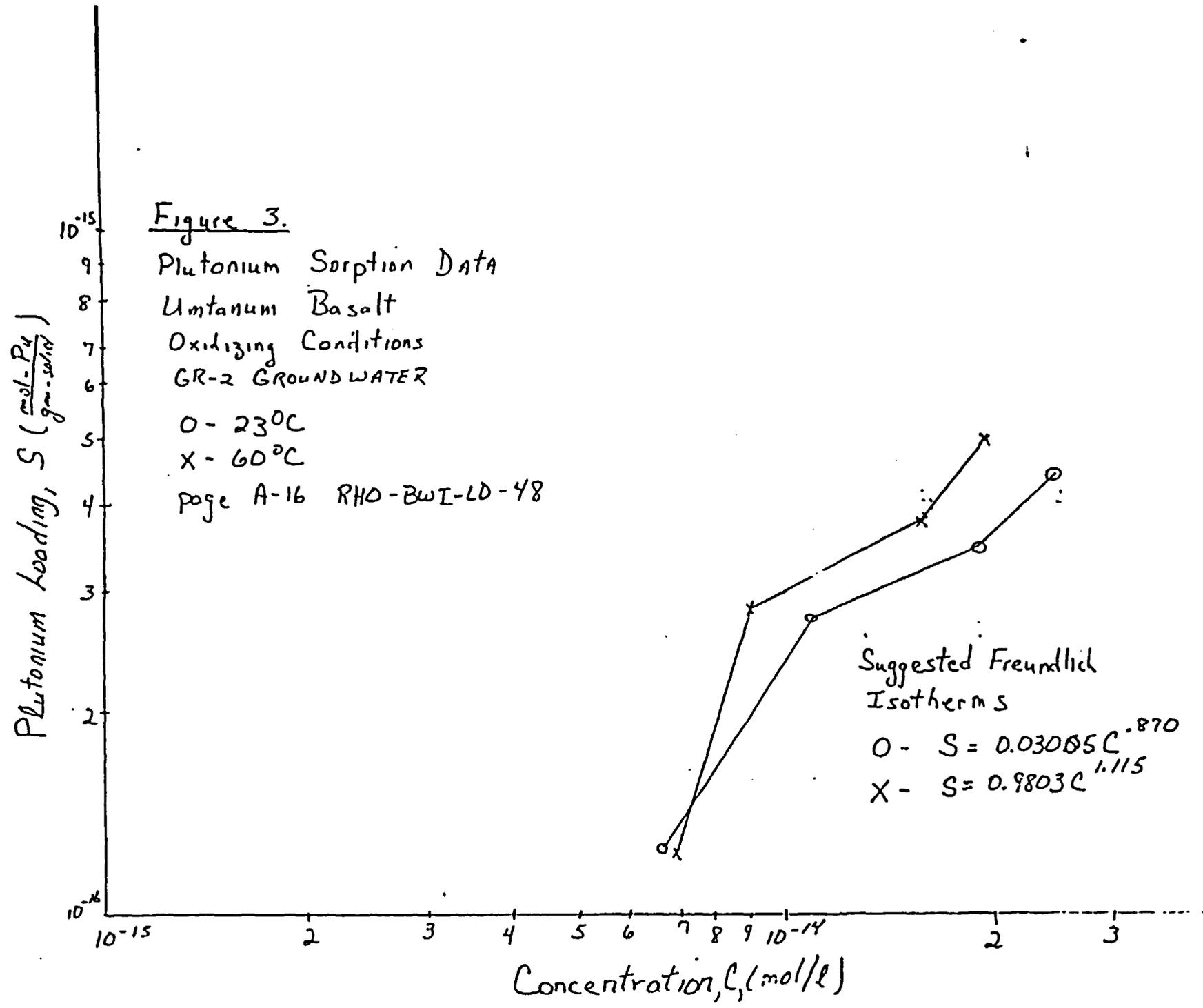


Figure 4

Radium Sorption Data

Umtarum Basalt

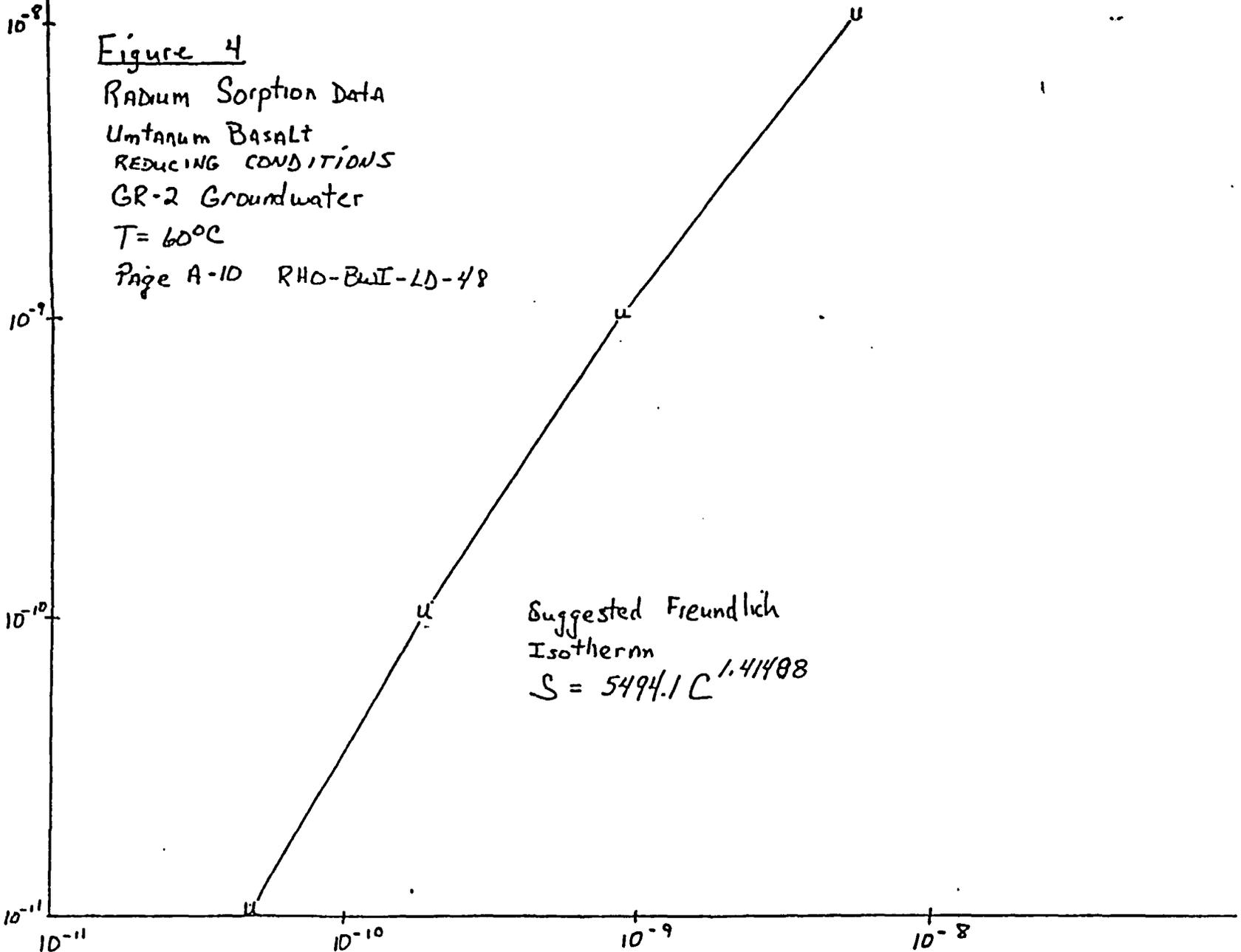
REDUCING CONDITIONS

GR-2 Groundwater

T = 60°C

Page A-10 RHO-BWI-LD-48

Radium Loading,  $S$  ( $\frac{\text{mol-Ra}}{\text{g-soln}}$ )



Suggested Freundlich  
Isotherm

$$S = 5494.1 C^{1.41488}$$

Concentration,  $C$  (mol/l)

Figure 5

Cesium Sorption Data

GR-2 Groundwater

Oxidizing Conditions

T = 60°C

Page A-6 RHO-BWI-LD-48

U - Umtanum Basalt

E - Flow E Basalt

P - Pomona Basalt

Suggested DR Isotherms

$$S = S_{max} \exp \left[ -\beta (RT \ln(1 + \frac{1}{C}))^2 \right]$$

$$U - S = 1.8E-05 \exp \left[ -0.0031 (RT \ln(1 + \frac{1}{C}))^2 \right]$$

$$E - S = 1.8E-05 \exp \left[ -0.0032 (RT \ln(1 + \frac{1}{C}))^2 \right]$$

$$P - S = 2.8E-05 \exp \left[ -0.0030 (RT \ln(1 + \frac{1}{C}))^2 \right]$$

Cesium loading, S (mol-Cs / gm-solid)

10<sup>-6</sup>

10<sup>-7</sup>

10<sup>-8</sup>

10<sup>-9</sup>

10<sup>-10</sup>

10<sup>-11</sup>

10<sup>-12</sup>

10<sup>-12</sup>

10<sup>-11</sup>

10<sup>-10</sup>

10<sup>-9</sup>

10<sup>-8</sup>

10<sup>-7</sup>

10<sup>-6</sup>

10<sup>-5</sup>

10<sup>-4</sup>

Concentration, C (mol/l)

Table 2

Distribution coefficients,  $\frac{S_M}{C}$ , for radium. The test used umtanum basalt as the solid at 60°C in CR-2 groundwater with an Eh of -0.15V

<u>C(mol/l)</u>	<u><math>S_M \left( \frac{\text{mol-Ra}}{\text{g-solid}} \right)</math></u>	<u><math>\frac{S_M}{C} * 1000 \left( \frac{\text{mL}}{\text{g}} \right)</math></u>
6.12E-9	1.10E-8	1800
8.95E-10	1.13E-9	1262
1.90E-10	1.12E-10	590
4.70E-11	1.12E-11	238

Again the fit is not precise and differences between the measured amount sorbed and the fit varied by as much as a factor of two. Later, it will be shown that the DR isotherm does not follow the trends of the data and was an inappropriate choice for the isotherm.

In summary, several points concerning the data,  $K_d$ 's, and the empirical fits need to be emphasized.

First, although not presented in this note, in many cases for a given species the amount sorbed is linearly proportional to the solution concentration over a limited range of concentrations. However, this is generally not true over the entire range of solution concentrations. Under certain conditions, as the solution concentration increases it is found that the amount sorbed increases at a slower rate. That is,  $dS/dC$  increases at a slower rate and the distribution coefficient,  $S/C$ , decreases. Under some conditions uranium and cesium exhibit this behavior as presented in Table 3. It should be kept in mind that near the waste form boundary, the solution concentration will be close to their solubility limits. Therefore, the region where  $dS/dC$  is small may be important to modeling transport because retardation, Equation (1), is a function of  $dS/dC$ . In other cases, radium for example, the ratio  $S/C$  increases with increasing concentration and again using a constant  $K_d$  is not an appropriate model.

Second, although dignified by the title isotherm, the Freundlich isotherm is only an interpolating function. It does not supply any physical information on the sorption phenomena and applies only within the range of experimental data points. The DR isotherm may, if used properly, provide physical insight into the sorption process, however, it also acts primarily as an interpolating function.

Finally, the purpose of fitting the data to an isotherm is unclear. A valid use for the proposed isotherms would be to differentiate them and use the result to obtain a retardation coefficient via Equation (1). However, this is not done. The isotherms are not used for any purpose as  $K_d$  is assumed constant.

It is stressed that before using any isotherm to obtain  $dS/dC$ , the isotherm/empirical fit must be checked to insure it is accurate and matches the trends in the data. As previously mentioned, this is not the case for the uranium and plutonium data being fit to a Freundlich isotherm. Similarly, the DR isotherm does not match the trends of the cesium data. In particular, the

Table 3

Distribution coefficients for cesium and uranium. Uranium data is for sorption on umtanum basalt in GR-1 groundwater at 60°C and an Eh of -0.30 volts. Cesium data is for sorption on umtanum basalt in GR-2 groundwater at 60°C in oxidizing conditions.

## Uranium Data

<u>C(mol/l)</u>	<u><math>S_M(\frac{\text{mol-U}}{\text{g-solid}})</math></u>	<u><math>\frac{S_M}{C} * 1000 (\frac{\text{mL}}{\text{g}})</math></u>
$3.99 \times 10^{-5}$	$6.01 \times 10^{-7}$	15
$8.00 \times 10^{-7}$	$9.20 \times 10^{-8}$	115
$6.28 \times 10^{-8}$	$9.37 \times 10^{-9}$	149
$1.37 \times 10^{-8}$	$2.31 \times 10^{-9}$	168

## Cesium Data

$3.39 \times 10^{-4}$	$6.61 \times 10^{-6}$	20
$6.31 \times 10^{-7}$	$9.33 \times 10^{-8}$	148
$2.63 \times 10^{-9}$	$9.77 \times 10^{-10}$	371
$2.34 \times 10^{-11}$	$9.77 \times 10^{-12}$	417
$3.63 \times 10^{-12}$	$1.58 \times 10^{-12}$	435

cesium data indicates that  $dS/dC$  decreases as concentration increases. This is reflected by the decreasing ratio  $S/C$  as presented in Table 4. However, for the DR isotherm this is not the case. Differentiating the expression for the DR isotherm gives,

$$\frac{dS}{dC} = \frac{S}{C} \left[ 2K(RT)^2 \ln \left( 1 + \frac{1}{C} \right) \right] / (1+C), \quad (9)$$

where the variables have been defined after Equation (8). Examination of Equation (9) indicates that the DR isotherm has the following properties:

$$\begin{aligned} \lim_{C \rightarrow 0} \frac{dS}{dC} &= 0, \\ \lim_{C \rightarrow \infty} \frac{dS}{dC} &= 0, \\ \frac{dS}{dC} &\geq 0, \quad 0 \leq C \leq \infty. \end{aligned} \quad (10)$$

These properties indicate that  $dS/dC$  will obtain a maximum for some non-zero concentration and the isotherm has an S-shape. Thus, before using a DR isotherm the data should show  $S/C$  as an increasing function at low concentrations, reaching a maximum and then decrease. This behavior is reflected in the  $S/C$  values obtained using the DR isotherm as presented in Table 4. From Table 4 it is clear that the choice of the isotherm and fit to the data can greatly alter the estimate of  $S/C$  which is directly proportional to  $dS/dC$ , the key parameter in the retardation coefficient. In this case, the  $S/C$  values obtained from the DR isotherm range from a factor of 2 lower to a factor of 2 higher than the measured  $S/C$ .

Table 4

Comparison of measured and empirically fitted distribution coefficients, S/C, for cesium. Cesium data is for sorption on uranium basalt with GR-2 groundwater at 60°C and oxidizing conditions. The empirically fitted distribution coefficients are determined from the DR isotherm used to fit the data,

$$S = S_{\max} \exp \left[ -K(RT \ln \left( 1 + \frac{1}{C} \right))^2 \right]. \quad S_{\max} = 1.8E-05, \quad K = 3.1E-03.$$

$C \left( \frac{\text{mol}}{\text{g}} \right)$	$S_M \left( \frac{\text{mol-Cs}}{\text{g-solid}} \right)$ (measured)	$S_{DR} \left( \frac{\text{mol-Cs}}{\text{g-solid}} \right)$	$\frac{S_M}{C} \times 1000 \left( \frac{\text{mL}}{\text{g}} \right)$	$\frac{S_{DR}}{C} \times 1000 \left( \frac{\text{mL}}{\text{g}} \right)$
3.63E-12	1.58E-12	1.2E-12	435	343
2.34E-11	9.77E-12	1.18E-11	418	504
2.63E-09	9.77E-10	1.69E-9	371	643
6.31E-07	9.33E-8	1.42E-7	148	225
3.39E-04	6.61E-6	3.95E-6	20	12

### 3. RECOMMENDATIONS AND CONCLUSIONS

Retardation of solute transport through a porous medium through sorption onto a solid is a valid concept and has been demonstrated in column infiltration experiments. However, before the results of batch sorption studies can be used to model retardation, the following points should be considered. Sorption is a complex phenomena and for this reason experimental measurements should be conducted over the entire range of concentrations expected in the repository. For near-field calculations, emphasis should be placed on multi-species testing with concentrations near the solubility limits of the nuclides because this is the expected condition near the edge of the waste form. Even this data may not truly represent the actual conditions in the repository due to radiolysis effects and interaction of the groundwater with the canister. Assuming the experimental conditions provide a reasonable estimate of the repository conditions, the data should be used to evaluate  $dS/dC$  for each species which will determine the retardation coefficients. Care must be taken in estimating  $dS/dC$ . Currently, the data is too sparse to allow a reasonable finite difference estimate and attempts to fit various isotherms to the data have been done poorly and are unreliable. In general, the constant  $K_d$  approach is applicable only in dilute solutions and therefore should not be used near the waste form.

As a final comment, it is emphasized that retardation through sorption reactions will delay the time at which the radionuclides first reach a given point, however, behind this front, once steady-state conditions arise there will be no net retardation because it is assumed that the reactions are in equilibrium.

## REFERENCES

1. Smith, M. J., et al., 1980. Engineered Barrier Development for a Nuclear Waste Repository in Basalt: An Integration of Current Knowledge, RHO-BWI-ST-7, Rockwell Hanford Operations, Richland, Washington 99352.
2. Salter, P. F., and G. K. Jacobs, 1982. "Evaluation of Radionuclide Transport: Effect of Radionuclide Sorption and Solubility," Scientific Basis for High-Level Waste Management V, Vol. 11, Elsevier, N.Y., pp. 801-810.
3. Salter, P. F., and G. K. Jacobs, 1983. BWIP Data Package for Reference Solubility and Kd Values, SD-BWI-DP-001, Rockwell Hanford Operations, Richland, WA.
4. Salter, P.F., L. L. Ames, and J. E. McGarran, 1981. The Sorption Behavior of Selected Radionuclides on Columbia River Basalts, RHO-BWI-LD-48, Rockwell Hanford Operations, Richland, WA.

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Department of Nuclear Energy

July 26, 1984

Mr. Everett A. Wick  
High Level Waste Licensing Management Branch  
Division of Waste Management  
Mail Stop 965 SS  
U. S. Nuclear Regulatory Commission  
Washington, DC 20555

Dear Mr. Wick:

Comparison of SRL Glass Leaching Data to Recent MCC Long-Term Results

At your request, a comparison has been made of glass leaching data from SRL and recent information reported by PNL on SRL-131 glass.<sup>1</sup> All tests were carried out on "composite" borosilicate glass devised by SRL to simulate a composition that is an "intermediate" between the Stage 1 and Stage 2 waste form compositions to be produced at the Defense Waste Processing Facility.

In order to compare data from the two studies, SRL results on composite glass, given in reference 2, have been used. Data from this report are, however, not comprehensive, and are for tests lasting up to 28 days. Therefore, only short-term-test data from the MCC tests were used to serve as a basis for comparison. The test conditions common to both sets of data include the following:

Test temperature	90°C
Test solutions	Deionized water (DIW), MCC brine, MCC silicate water
Test times	7d, 28d
Glass surface area to solution volume ratio (SA/V)	0.1 cm <sup>-1</sup>
Elements leached	Si, B, Na, Cs, U.

1. "MCC-D2 One-Year Leach Test Data for SRL-131 Glass," Materials Characterization Center, May 1, 1984.
2. M. J. Plodinec and others, "An Assessment of Savannah River Borosilicate Glass in the Repository Environment," DP-1629, 1982.

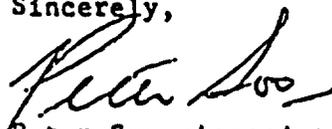
Mr. Everett A. Wick  
July 26, 1984  
Page 2.

Tables 1 and 2 summarize the SRL and MCC data, respectively. The reproducibility of the MCC data is excellent. This is to be expected since they were obtained under carefully controlled conditions for one series of tests. The SRL results were probably obtained over a period of time in separate experiments.

Table 3 shows a comparison of leaching results for Si, B, Na, Cs and U. Usually, there is good agreement between the two data bases for DIW and MCC silicate water. For brine, however, the leach rates obtained by SRL are much higher. Also, the U release rates are several times higher in the SRL studies for all three tests solutions. Reasons for these discrepancies are not clear at this time.

In summary, the MCC and SRL leaching data for tests lasting up to 28 days are basically in agreement. Exceptions are found for the SRL brine leachant studies and for U releases in all solutions in which cases the MCC leach rates are much lower.

Sincerely,



Peter Soo, Associate Division Head  
Nuclear Waste Management Division

PS:gfs

Enclosure

cc: M. S. Davis  
W. Y. Kato  
H. J. C. Kouts  
D. G. Schweitzer  
HLW Staff  
M. Bell, NRC  
T. Johnson, M&C  
Docket Control Center, NRC

Table 1. Summary of composite SRL glass leaching data from Report Number DP-1629.

Temp. (°C)	Test Time (d)	Solution	SA/V (cm <sup>-1</sup> )	Leachability (g/m <sup>2</sup> -d)					DP-1629 Table Number
				Si	B	Na	Cs	U	
90	28	DIW	0.1	0.61	0.86	0.94			15
90	7	DIW	0.1	1.52					23
90	7	MCC Brine	0.1	0.34					23
90	7	MCC Silicate	0.1	0.75					23
90	28	DIW	?	0.55	0.79		1.39		24
90	28	MCC Brine	?	0.21	0.31		0.24		24
90	28	MCC Silicate	?	0.21	0.38		0.83		24
90	28	DIW	0.1	0.96			0.58	0.13	25
90	28	MCC Brine	0.1	0.32			0.35	<0.019	25
90	28	MCC Silicate	0.1	0.56			0.49	0.44	25

Table 2. Summary of composite SRL glass leaching data from MCC-D2 tests.

Temp. (°C)	Test Time (d)	Solution	SA/V (cm <sup>-1</sup> )	Leachability (g/m <sup>2</sup> -d)					MCC-D2 Table Number
				Si	B	Na	Cs	U	
90	7	DIW	0.1	2.30	2.89	2.73	3.43	0.15	4.1
90	28	DIW	0.1	0.88	1.19	1.10	1.29	0.03	4.1
90	28	DIW	0.1	0.87	1.17	1.09	1.27	0.06	4.1
90	28	DIW	0.1	0.81	1.11	1.03	1.21	0.06	4.1
90	28	MCC Brine	0.1	0.05	0.03	=0	=0	0.003	4.7
90	28	MCC Brine	0.1	0.06	0.01	=0	=0	0.004	4.7
90	28	MCC Brine	0.1	0.05	0.03	=0	=0	0.002	4.7
90	28	MCC Silicate	0.1	0.44	0.70	0.84	0.75	0.17	4.4
90	28	MCC Silicate	0.1	0.44	0.70	0.88	0.74	0.19	4.4
90	28	MCC Silicate	0.1	0.45	0.74	0.87	0.77	0.15	4.4

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Table 3. Comparison of MCC-D2 and SRL leaching data for composite glass for 7- and 28-day tests.

Temp. (°C)	Test Time (d)	Solution	SA/V (cm <sup>-1</sup> )	Element	Leachability Range (g/m <sup>2</sup> -d)	
					SRL	MCC-D2
90	7	DIW	0.1	Si	1.52	2.30
90	28	DIW	0.1	Si	0.55-0.96	0.81-0.88
90	28	DIW	0.1	B	0.79-0.86	1.11-1.19
90	28	DIW	0.1	Na	0.94	1.03-1.10
90	28	DIW	0.1	Ca	0.58-1.39	=0
90	28	DIW	0.1	U	0.13	0.03-0.06
90	28	MCC Brine	0.1	Si	0.21-0.32	0.05-0.06
90	28	MCC Brine	0.1	B	0.31	0.01-0.03
90	28	MCC Brine	0.1	Ca	0.24-0.35	=0
90	28	MCC Brine	0.1	U	<0.019	0.002-0.004
90	28	MCC Silicate	0.1	Si	0.21-0.56	0.44-0.45
90	28	MCC Silicate	0.1	B	0.38	0.70-0.74
90	28	MCC Silicate	0.1	Ca	0.49-0.83	0.74-0.77
90	28	MCC Silicate	0.1	U	0.44	0.15-0.19