WPO 41374

Analysis Plan AP-024

Analysis to Estimate the Uncertainty for Predicted Actinide Solubilities

WBS 1.1.10.1.1
Rev. 0, effective date 9/3/96

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E. J. Nowak, 6831 — Date
Manager, Chemical and
Disposal Room Processes
Department

QA Reviewer: Deborah Coffey — 9/6/96
Deborah Coffey, 6811 — Date
Deputy Chief, Quality
Assurance Department

Analysis to Estimate the Uncertainty for Predicted Actinide Solubilities

Scope of the Analysis:

Objective
The objective of this work is to estimate the reasonable range and distribution of the uncertainty contained within the predictions of actinide solubility in model WIPP repository brines as required for WIPP Performance Assessment calculations. The predictions of actinide solubility were made via two routes. The justification for and the experiments planned to produce the chemical data required as input to the predictions are described in *SAND95-1895, The Waste Isolation Pilot Plant (WIPP) Actinide Source Term: Test Plan for the Conceptual Model and the Dissolved Concentration Submodel* (Novak, 1995). The actual predictions of dissolved concentrations are contained in *WPO#35835 Documentation Package for Solubility Parameters for Actinide Source Term Look-Up Tables* (Siegel, 1996).

Assumptions
This analysis will be performed using manual estimation of the difference between a model predicted curve and direct experimental observation (difference). These difference values will then be used to generate a histogram depicting the frequency of occurrence of a specific difference value and the range of difference values encountered. It is assumed that this method will give a reasonable representation of the model uncertainty within the range of conditions predicted for the repository. This analysis will not address the uncertainty in the repository conditions being modeled.

For the +3, +4, and +5 oxidation states, the Dissolved Concentration Submodel (DCS) was utilized to make the predictions. For the +6 oxidation state, sufficient Pitzer parameters were not available to allow for a reasonable prediction to be made via the DCS. Thus for the +6 oxidation state a combination of literature and direct empirical measurements were utilized to generate the solubility predictions.

Although the solubility predictions for the +6 oxidation state were made via a different method as described above, it was assumed that the error range which is determined for the other oxidation states will be applicable to the +6 oxidation state, and the distribution will be loguniform. During the performance of this analysis, the assumption of the distribution of the +6 oxidation state being log uniform was revised. Based on discussions with Martin Tierney and Mary-Alena Martell, it was determined that there was no justification for utilizing a different distribution for the +6 oxidation state. Therefore, the same uncertainty estimation and distribution was utilized for the +6 oxidation state.

Primary Task to Perform the Analysis

Step 1 Gather Data

To perform the analysis, suitable data must be identified. For the purposes of this uncertainty estimation, any data which includes a model prediction by FMT or a fitting of a curve utilizing NONLIN and the corresponding experimentally determined data points are suitable. The sources of these data are Pacific Northwest National Laboratory (under contract AF-3339 and "PNL-MA-70 Quality Assurance Plan No. EES-107, Revision O" dated 30 December 1993, approved on 11/29/93), Sandia National Laboratories (utilizing raw data from Lawrence Berkley Laboratory obtained under contract AH-5592 and "QA Program for WIPP Actinide Source Term Program (ASTP) R3", effective date March 28, approved 7/18/95, and the literature. The data sets gathered are included as Attachment A.

Step 2 Determine Applicability of Data

The oxidation state solubility models are in various degrees of completeness. Although the +3, +4, and +5 models are adequately complete in the chemical region of interest relevant to the repository, there are known problems in regions where extrapolation far beyond the conditions under which the model was parameterized. Including any data in these regions where there are known parameterization problems would lead to overestimation of the error. The data available for the +3 and +5 models were determined to be adequate for this analysis in all regions and were thus utilized in their entirety. The data available for the +4 model were found to have significant problems in the extrapolated regions and were thus determined to be inadequate for use in this analysis. To utilize any of the data for the +4 model in this analysis, a subjective judgment would have to be made as to which values to include and which to exclude. In order to avoid any bias in this selection, the entire data set was not utilized.

Step 3 Assess Difference Of Predicted Values Versus Observed Values

The difference is defined as the difference between an experimentally measured point and the model prediction or fitted curve at the conditions as close as possible to the conditions under which the experimental data were collected. The difference values were determined by direct measurement utilizing a ruler from the graphs provided by the researchers. These values are included as Attachment B.

Step 4 Develop Error Range and Distribution

Each of the difference values were entered into Microsoft Excel 5.0c utilizing a Microsoft DOS platform run on a Gateway 2000 486/33 computer. The built in Histogram function was then utilized to generate a frequency distribution and to sort the data into ranges. The

histogram is presented as attachment C. The frequency associated with each bin in the histogram reflects the number of occurrences of values between the previous bin number and the subject bin number. For example, the bin labeled '-1.95' contains all of the values between -1.95 and the previous value of -2.1. Investigator judgment with assistance from a PA staff member was used to determine the data bins appropriate for use by the Performance Assessment. Once these bins were determined, the histogram function was again run to generate the cumulative distribution function (CDF) which was transmitted to Performance Assessment as shown in Attachment D.

Step 5 Transmittal of Results to PA and SNL WIPP Central Files (SWCF)

Results of the analysis were documented in a series of memoranda to the Performance Assessment Department, Attachment D. The memoranda were prepared and reviewed according to applicable requirements of QAP 6-3 for Conducting and Documenting Reviews of Documents. This analysis documentation was prepared according to the requirements of QAP 9-1 and is being submitted to the SWCF.

Individuals Who Performed the Analysis

The primary individual who performed the analysis was Vann Bynum, an SAIC contractor to Sandia National Laboratories (SNL) Department 6831. Assistance was obtained from Christine Stockman, SNL Department 6849 with respect to determining the data bins appropriate for use by the Performance Assessment.

Deliverables

The specified deliverable, the memoranda presented in Appendix D, satisfied the deliverable requirements specified in AP-024.

Controlling Documents

The controlling documents for this analysis are:

AP-024 Estimation of Uncertainty for Predicted Actinide Solubilities
QAP6-3 Conducting and Documenting Reviews of Documents
QAP9-1 Quality Assurance Requirements for Conducting Analyses
QAP17-1 Quality Assurance Records Source Requirements
QAP19-1 Rev 2 WIPP Computer Software Requirements

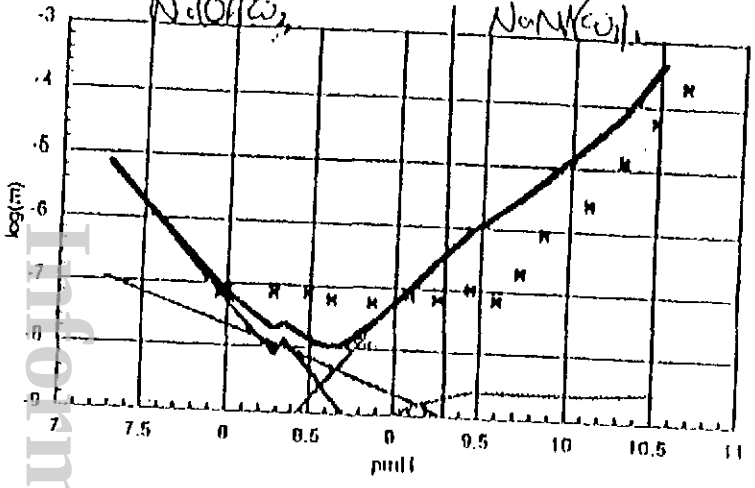
References

Novak, C. F. 1995 *The Waste Isolation Pilot Plant (WIPP) Actinide Source Term: Test Plan for the Conceptual Model and the Dissolved Concentration Submodel, SAND95-1895,* Sandia National Laboratories, Albuquerque, NM.

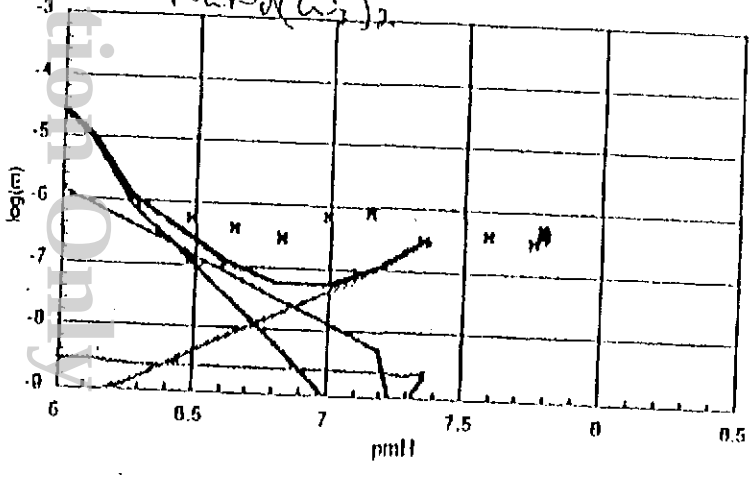
Siegel, M. D. 1996 *Documentation Package for Solubility Parameters for Actinide Source Term Look-Up Tables WPO#35835* Sandia National Laboratories, Albuquerque, NM.

PREDICTIONS

ERDA6, PCO2(g) = 10-3.5

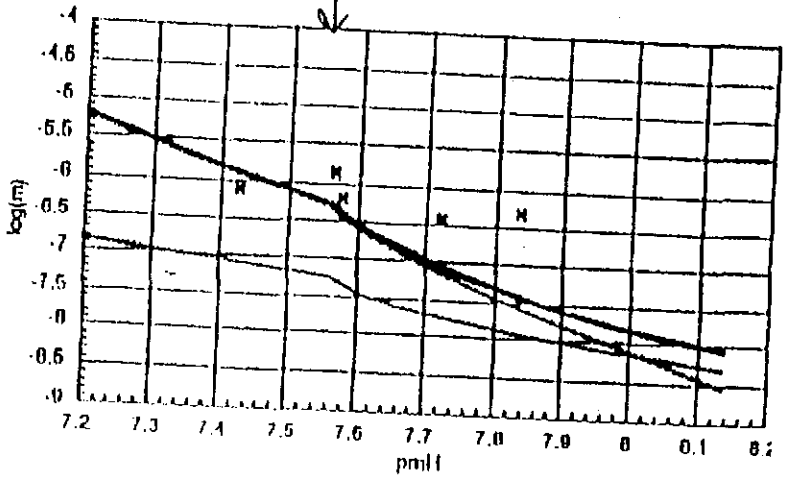


ERDA6, PCO2(g) = 1

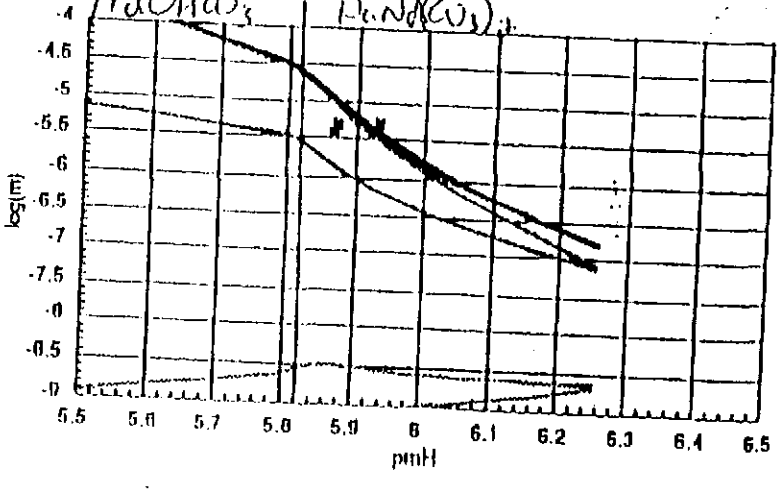


PRELIMINARY

GSeep, PCO2(g) = 10-3.5



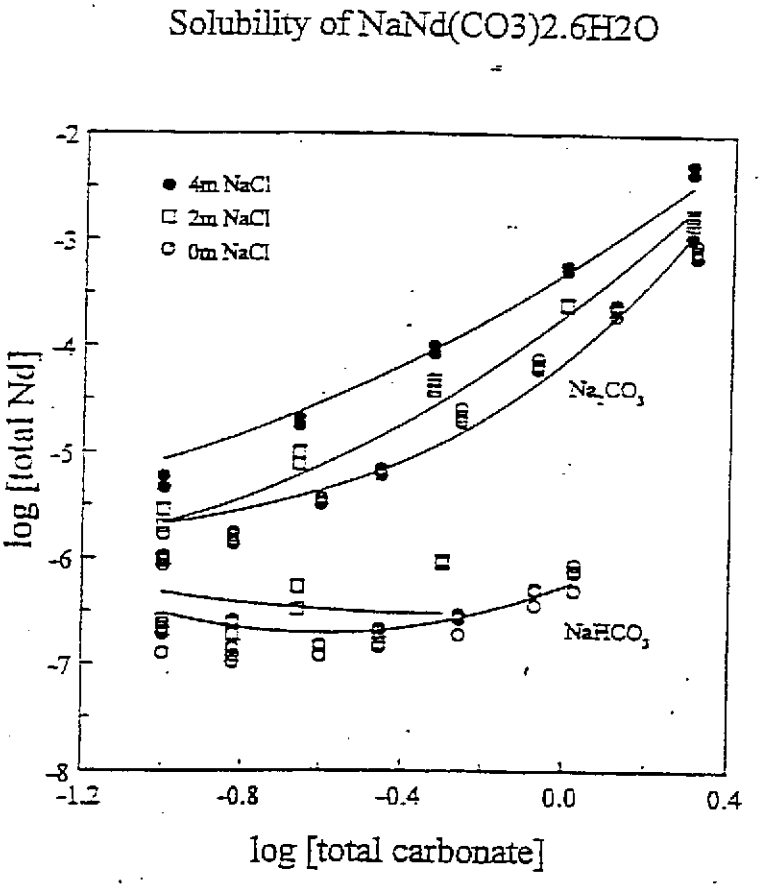
GSeep, PCO2(g) = 1



PRELIMINARY

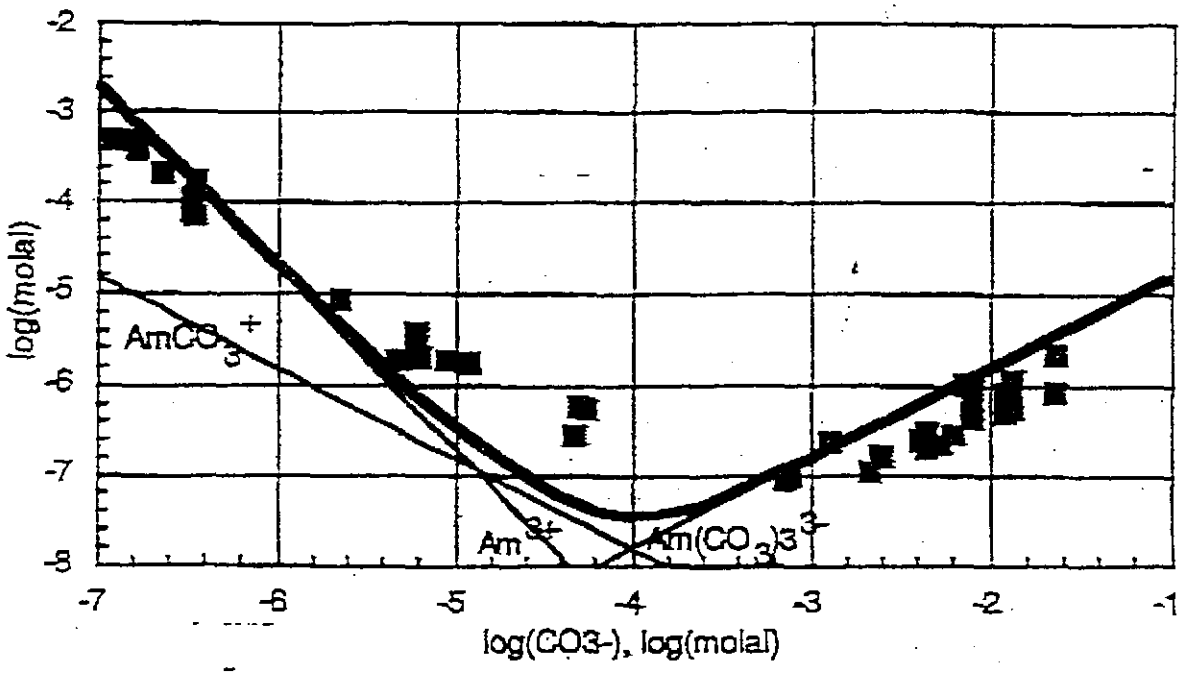
Data from C. F. Novak based on raw data from PNNL

Attachment A2



Data from Dhan Rai, PNNL

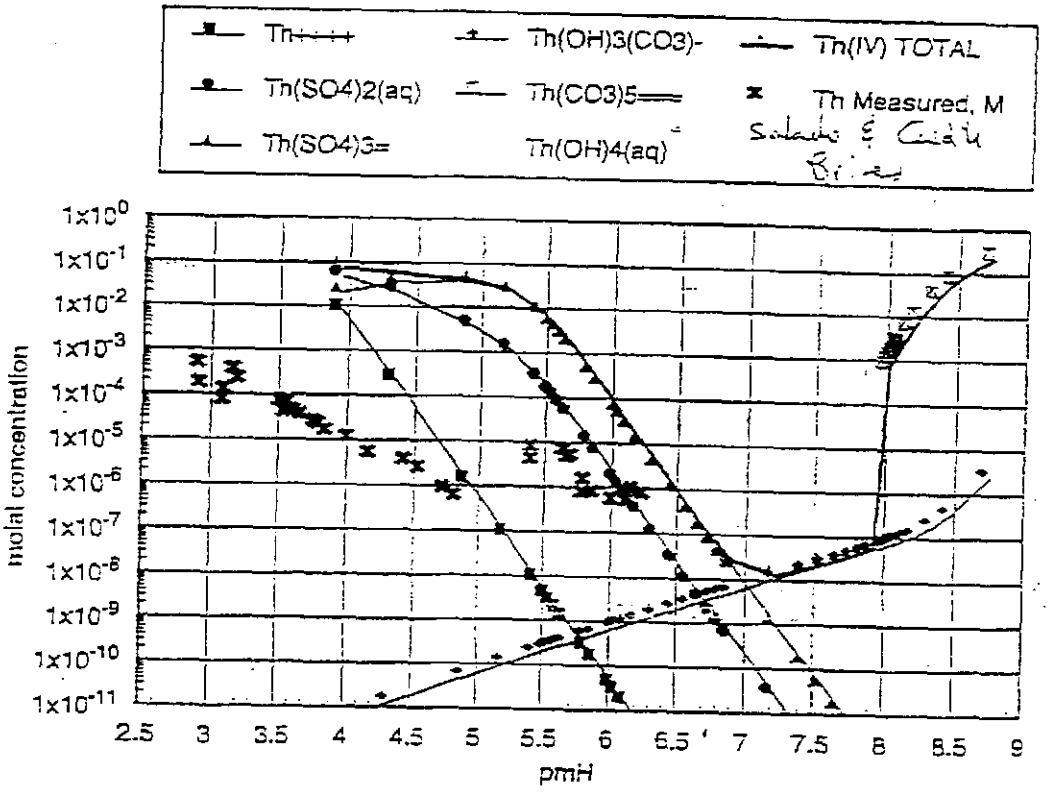
Attachment A3



Comparison of the current +III actinide solubitliy model with measured solubility of $^{241}Am(III)$ in 5.6m NaCl with $pCO_2(g)$=0.01 data from Runde and Kim (1995). The bold line gives total Am(III) dissolved concentration as calculated with the WIPP model and data base.

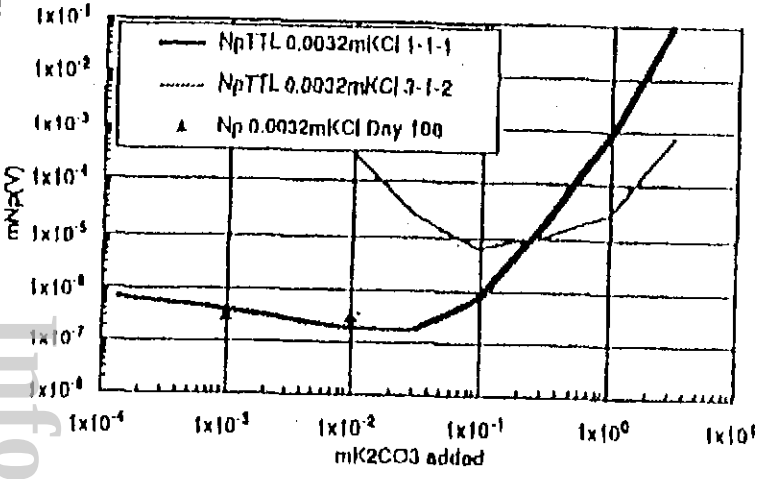
Comparison by C. F. Novak with the data contained in Runde, W. and J. I. Kim 1994 "Untersuchungen der Ubertragbarkeit von Labordaten naturliche Verhaltnisse. Chemisches Verhalten von drei- und funf wertigen Americium in salinen NaCl Losungen" Report RCM-01094, Institut fur Radiochemie, Technische Universitat Munchen.

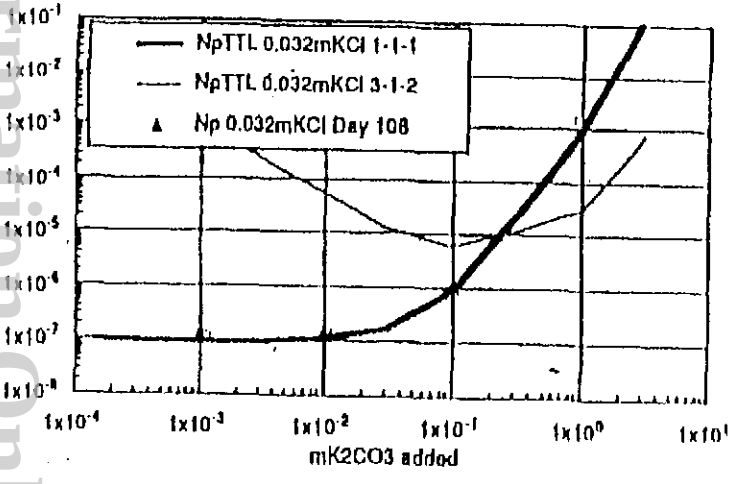
Attachment A4



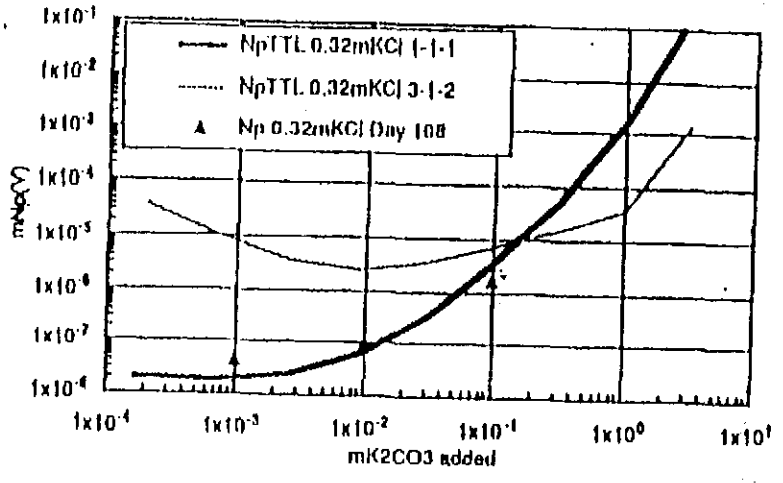
Data from Dhan Rai, PNNL

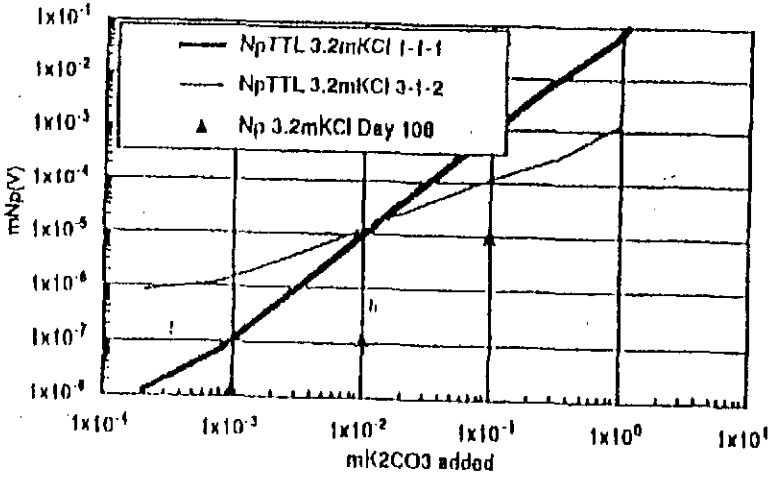
PREDICTIONS





OLD (NOV 95) MODEL VS DATA





OLD (NOV 95) MODEL VS DATA

Data from C. F. Novak, based on raw data from LBL

+3	ERDA6	-3.5
(log molality Nd)		
0		
0.7		
0.9		
0.8		
0.2		
0		
-0.7		
-1.1		
-1.4		
-1.2		
-0.8		
-1		
-0.7		
-0.8		
+3	ERDA6	1
(log molality Nd)		
0		
0.4		
0.6		
0.8		
1.1		
1		
0		
+3	GSeep	-3.5
(log molality Nd)		
-0.2		
0.6		
0.2		
0.8		
1.4		
+3	GSeep	1
(log molality Nd)		
-0.4		
-0.5		
0.2		

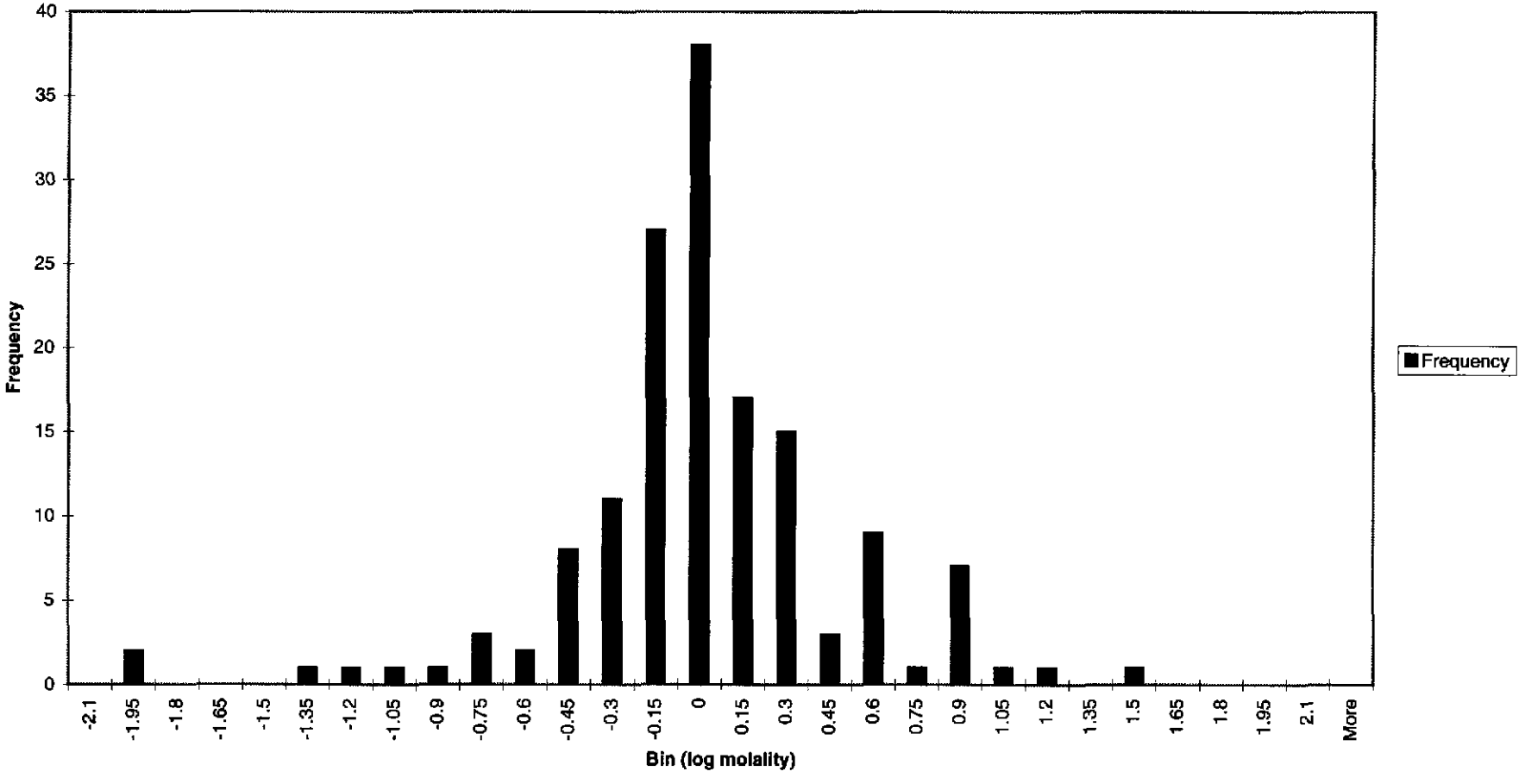
4m NaCl	Na2CO3		2m NaCl	NaHCO3
(log molality Nd)			(log molality Nd)	
-0.25			-0.3	
-0.15			-0.25	
-0.1			-0.2	
-0.2			-0.3	
-0.05			0.2	
0.05			0	
0.05			0.5	
0			0.5	
0.2				
0.15			0m NaCl	NaHCO3
-0.5			(log molality Nd)	
			-0.1	
2m NaCl	Na2CO3		-0.2	
(log molality Nd)			-0.4	
0			0.1	
0.1			-0.25	
0.1			-0.3	
0.2			-0.2	
0.1			-0.1	
0.2			0	
0.3			0	
0.1			-0.1	
0			0	
-0.1			0	
-0.15			-0.15	
			0.1	
0m NaCl	Na2CO3		0.1	
(log molality Nd)			-0.1	
-0.1			0.1	
-0.4			0.15	
-0.4			-0.1	
-0.5				
-0.25				
-0.25				
-0.2				
-0.2				
-0.1				
-0.1				
0				
0				
0				
0.25				
0.2				
0.15				
0.2				
0.2				
0.1				
0.1				
0.1				
0				
-0.1				
-0.15				
-0.2				

log molality Am	
-0.5	
-0.2	
-0.2	
0	
-0.2	
-0.4	
0.2	
0.2	
0.4	
0.6	
0.8	
0.5	
0.6	
0.6	
0.5	
0.8	
0.8	
-0.2	
-0.2	
0	
-0.5	
-0.4	
-0.4	
-0.3	
-0.2	
-0.4	
-0.4	
0	
-0.5	
-0.4	
-0.6	
-0.4	
-0.3	
-0.2	
-0.5	

The data for the +4 model were excluded from the analysis as described in Step 2 of this document.

+5	KCl/K2CO3	0.0032m KCl
(log molality Np)		
-0.2		
0.2		
+5	KCl/K2CO3	0.032m KCl
(log molality Np)		
0		
0		
0		
+5	KCl/K2CO3	0.32m KCl
(log molality Np)		
0.4		
0.1		
-0.2		
+5	KCl/K2CO3	3.2m KCl
(log molality Np)		
-0.9		
-2		
-2		

Attachment C
Model Uncertainty



Attachment D1

Date: May 22, 1996

To: Martin S. Tierney, (Dept. 6741), MS-1328
Christine Stockman, (Dept. 6749), MS-1328

From: R. V. Bynum

Subject: Update of Uncertainty Range and Distribution for Actinide Solubility to be Used in CCA Nuts Calculations

A more detailed analysis of the expected uncertainty range and distribution for the actinide solubilities resulting from the FMT modeling simulations has been performed. This analysis was performed as described in the Analysis Plan *AP-024, Estimation of Uncertainty for Predicted Actinide Solubilities, Rev. 0.*

The log values reported should be added to the log value of the actinide concentrations.

CDF	log value
0	-2
4E-2	-1
0.13	-0.5
0.27	-0.25
0.63	0
0.84	0.25
0.89	0.5
0.99	1
1	2

The mean value is 0.24 and the median value is -0.09.

cc:
J. T. Holmes
M. D. Siegel
E. J. Nowak
C. C. Crafts
R. F. Weiner
C. F. Novak
R. Moore
M. Chu
SWCF-A:WBS1.1.10.1.1:AST;WBS1.2.07.1:PDD:QA:GENERAL:WPO#35835

Attachment D2

Date: May 23, 1996

To: Martin S. Tierney, (Dept. 6741), MS-1328
Christine Stockman, (Dept. 6749), MS-1328

From: R. V. Bynum

Subject: Revised Update of Uncertainty Range and Distribution for Actinide Solubility to be Used in CCA Nuts Calculations

Additional review of the analysis of the expected uncertainty range and distribution for the actinide solubilities resulting from the FMT modeling simulations has raised questions concerning the forced symmetry of the uncertainty range previously transmitted. The range of actual values is from -2 to +1.4, but the distribution was made symmetrical resulting in a range of -2 to +2. This symmetrical range cannot be supported by the existing data. Therefore, this range is being revised to reflect the actual range of the data. This analysis was performed as described in the Analysis Plan *AP-024, Estimation of Uncertainty for Predicted Actinide Solubilities, Rev. 0.*

The revised log values reported below should be added to the log value of the actinide concentrations.

CDF	log value
0	-2
4E-2	-1
0.13	-0.5
0.27	-0.25
0.63	0
0.84	0.25
0.89	0.5
0.99	1
1	-1.4

cc:

J. T. Holmes
M. D. Siegel
E. J. Nowak
C. C. Crafts
R. F. Weiner
C. F. Novak
R. Moore
M. Chu
SWCF-A:WBS1.1.10.1.1:AST;WBS1.2.07.1:PDD:QA:GENERAL:WPO#35835

Attachment D3

Date: June 20, 1996

To: Martin S. Tierney, (Dept. 6741), MS-1328
Christine Stockman, (Dept. 6749), MS-1328

From: R. V. Bynum

Subject: Revision to Memo "Revised Update of Uncertainty Range and Distribution for Actinide Solubility to be Used in CCA Nuts Calculations" Dated May 23, 1996

In the subject memo, I made a typographical error in the last entry in the table. The last entry should read

CDF	log value
1	1.4

The log value was previously reported as -1.4 in error. I have reviewed the applicable 464 forms and the value is correctly represented in each case. During that review however I noticed that the headings on the attached calculation sheet indicated that the range was -2 to +2 instead of -2 to +1.4. My review of the calculations indicate that they were performed with the correct range of -2 to +1.4.

cc:
J. T. Holmes
M. D. Siegel
E. J. Nowak
C. C. Crafts
R. F. Weiner
C. F. Novak
R. Moore
M. Chu
SWCF-A:WBS1.1.10.1.1:AST;WBS1.2.07.1:PDD:QA:GENERAL:WPO#35835