538219

Sandia National Laboratories
Waste Isolation Pilot Plant

SUPERSEDED
BY 539595 5/2/05 RH

Updated Uncertainty Analysis of Actinide Solubilities For the Response to EPA Comment C-23-16

Work carried out under the Analysis Plan for CRA Response Activities, AP-112, Rev. 0

Author:	Yongliang Xiong, 6822	12/17/04 Date
Author:	L. H. Brush for E. J. Nowak E. James Nowak, 6822	12/17/04 Date
Author:	L. H. Brush Laurence H. (Larry) Brush, 6822	12/17/04 Date
Technical Reviewer:	Donald E. Wall Donald E. Wall, 6822	12/17/04 Date
QA Reviewer:	Mario Chavez Mario J. Chavez, 6820	12/17/04 Date
Management Reviewer:	Mark Rigali Mark J. Rigali, 6822	12/17/04 Date

WIPP:1.2.7:PA:QA-L:PKG 533999

TABLE OF CONTENTS

1 INTRODUCTION 3

2 BACKGROUND 4

3 RESPONSE 5

3.1 Measured Actinide Solubilities 5
3.2 Frequency Distribution of Differences between Measured and Predicted Solubilities 6

3.2.1 Updated FMT Database Used for This Analysis 6
3.2.2 An(III) Frequency Distributions and CDF 7
3.2.3 An(IV) Frequency Distributions and CDF 7
3.2.4 An(V) Frequency Distributions and CDF 7
3.2.5 Combined (An(III, IV, V)) Frequency Distributions and CDF 8

4 CONCLUSIONS 9

5 REFERENCES 10

6 FIGURES 14

7 TABLES 22

1 INTRODUCTION

The U.S. EPA provided the following request (Cotsworth, 2004, Enclosure 1, Comment C-23-16):

> "DOE used the differences between modeled and measured actinide solubilities to estimate the uncertainties associated with actinide solubilities for the PA. Based on the figure presented in the CRA[-2004] [Compliance Recertification Application] (Figure SOTERM-1), it appears DOE used the solubilities calculated for the CCA rather than for the CRA. However, DOE indicates that solubilities calculated for the CRA[-2004] were different than the CCA (Table SOTERM-2)."
>
> "DOE must re-evaluate the uncertainties associated with actinide solubilities using solubilities calculated for the CRA, and use this information in the CRA[-2004] PA."

This analysis responds to the EPA's request that the "DOE must re-evaluate the uncertainties associated with actinide solubilities using solubilities calculated for the CRA[-2004], and use this information in the CRA[-2004] PA." It uses both previous (pre-CCA) and new (post-CCA) measurements of actinide solubilities and uses the latest (post-CCA) actinide-solubility data developed for the WIPP Project, as well as for non-WIPP-related applications. Then it derives a frequency distribution for differences between measured solubilities and those calculated (predicted) for the same conditions. The frequency distribution is used to represent the expected solubility uncertainty distribution.

This work was carried out under the Analysis Plan for CRA Response Activities (Kirkes and Wagner, 2004).

2 BACKGROUND

Novak et al. (1996) used Fracture Matrix Transport (FMT) to predict the solubilities of the +III, +IV, and +V actinides (An(III), An(IV), and An(V)) in the WIPP brines SPC and Energy Research and Development Administration [Well]-6 (ERDA-6) for the 1996 WIPP Compliance Certification Application (CCA) (U.S. DOE, 1996). FMT is a thermodynamic speciation and solubility code developed for the WIPP Project by Babb and Novak (1995). SPC is a synthetic brine similar to Brine A, which was used to simulate intergranular (grain-boundary) fluids from the Salado Formation (Fm.) at or near the stratigraphic horizon of the repository (Molecke, 1983). ERDA-6 is a synthetic brine typical of brines in the Castile Fm. (Popielak, et al., 1983). The An(III), An(IV), and An(V) thermodynamic speciation and solubility models implemented in FMT at the time of the CCA PA are described in detail by U.S. DOE (1996, Appendix SOTERM).

The DOE did not develop a thermodynamic speciation-and-solubility model for U(VI) in brines for the CCA PA, and has not developed one since the CCA. Instead, it used estimates by Hobart and Moore (1996) for the CCA PA, the 1997 PAVT, and the CRA-2004 PA.

Bynum (1996a, 1996b, 1996c) carried out an analysis to estimate the uncertainties in the An(III), An(IV), and An(V) solubility models implemented in FMT at the time of the CCA PA. These uncertainties were estimated mainly by comparing solubilities measured to develop these models and curves fitted to the data by the code NONLIN (Babb, 1996) to parameterize the Pitzer database. A few comparisons were also made between solubilities reported in the literature and FMT predictions for the conditions used in the experiments. A frequency distribution was generated for the differences between logarithms (base 10) of measured solubilities and logarithms of the values predicted for comparable conditions. Those differences quantified the ratios of measured to predicted values. The distribution was used to represent the expected uncertainty distribution for the solubilities predicted by FMT for the CCA PA, the 1997 PAVT, and the CRA-2004 PA.

Since the CCA, Babb and Novak (1997 and addenda) and Wang (1998) modified FMT. The FMT thermodynamic database has also been modified. Novak (1997) revised the database used for the CCA PA; his new database was used for the EPA's 1997 Performance Assessment Verification Test (PAVT). Giambalvo (2002a, 2002b, 2002c, 2002d, 2002e, 2003) modified the PAVT database and issued FMT_021120.CHEMDAT, the database used for the CRA-2004 PA. After the FMT calculations for the CRA-2004 PA (U.S. DOE, Appendix PA, Attachment SOTERM), Xiong (2004a) modified FMT_021120.CHEMDAT and Xiong (2004b) released FMT_040628.CHEMDAT. Xiong (2004a) corrected the molecular weight of oxalate in FMT_021120.CHEMDAT, and added solid calcium oxalate to this database.

This is the first uncertainty analysis of FMT predictions of actinide solubilities since that of Bynum (1996a, 1996b, 1996c).

3 RESPONSE

This section describes the methods used to address EPA Comment C-23-16. It discusses an updated actinide solubility uncertainty analysis comparing both previous (pre-CCA) measurements of actinide solubilities - including data used by Bynum (1996a, 1996b, 1996c) in the analysis for the CCA PA - and new (post-CCA) measurements of actinide solubilities, and predictions made with the latest (post-CCA) version of FMT (Babb and Novak, 1997 and addenda; Wang, 1998) and the current FMT thermodynamic database (Xiong, 2004a, 2004b). The analysis produced a probability distribution for FMT solubility predictions in the form of a distribution of differences between logarithms (base 10) of measured and predicted solubilities.

Separate comparisons were made between measured solubilities by the author or coauthors of each study included in this analysis, and FMT predictions for the actinide oxidation state (An(III), An(IV), or An(V)) and the conditions used in that study. The results were then combined for each oxidation state. Finally, the results for each oxidation state were combined to produce an overall comparison for all three oxidation states.

This analysis included the first comparisons for An(IV), because Bynum (1996a, 1996b, 1996c) did not include any comparisons for this oxidation state.

None of the experiments that produced the measured solubilities compared in this analysis included any organic ligands. Therefore, the results obtained from this analysis apply only to the inorganic components of the An(III), An(IV), and An(V) thermodynamic speciation and solubility models implemented in the FMT.

Finally, this analysis does not include any comparisons for An(VI), because the DOE has not developed a thermodynamic speciation and solubility model for this oxidation sate. (It used estimates by Hobart and Moore (1996) for the CCA PA, the 1997 PAVT, and the CRA-2004 PA.)

3.1 Measured Actinide Solubilities

Measured actinide solubilities were taken from documented sources that include both previous (pre-CCA) and new (post-CCA) studies. These studies are summarized in Table 1 with values of major dissolved constituents, ionic strength, pH, solubility-controlling solid phases, and citations. Included are solubilities measured in complex WIPP-related brine solutions such as ERDA-6, G-Seep, and SPC brines. Citations of studies that were included in the uncertainty analysis for the CCA PA (Bynum, 1996a; 1996b; 1996c) are identified with asterisks in the table.

3.2 Frequency Distribution of Differences between Measured and Predicted Solubilities

Frequency distributions of differences (D) between logarithms (base 10) of measured (Sm) and predicted (Sp) actinide solubilites were generated and displayed in tabular and histogram forms in Microsoft Excel spreadsheets using the "histogram" data analysis tool in this commercial spreadsheet software. Negative values of D indicate that the thermodynamic speciation and solubility model implemented in FMT predicted a solubility greater than the corresponding measured value (overprediction of solubility by the model). File "WIPP Solubility Uncertainty Values Rev1_12-1-04.xls" contains spreadsheets with measured and predicted solubility values (or logarithms thereof) frequency distributions, and histograms. Bin numbers (N) in the histograms were defined as follows:

> Bin N contains the count of values of D from greater than (N - 0.15) up to and including N, where D = LOG10(Sm) – LOG10(Sp).
>
> Examples of the definitions of bins follow:
>
> Bin -0.3 contains the count of values of D from > -0.45 to and including -0.3;
> Bin -0.15 contains the count of values of D from > -0.30 to and including -0.15;
> Bin 0 contains the count of values of D from > -0.15 to and including 0; and
> Bin 0.15 contains the count of values of D from > 0 to and including 0.15.

Results of this analysis are presented in the following subsections for each actinide oxidation state separately (An(III), An(IV), and An(V)) and for all three oxidation states combined. The results are presented as histograms of the frequency distribution and as cumulative distribution functions (CDFs)

3.2.1 Updated FMT Database Used for This Analysis

The FMT thermodynamic database used for this analysis is FMT_040628.CHEMDAT. Xiong (2004a, 2004b) modified FMT_021120.CHEMDAT, the database developed by Giambalvo (2002a, 2002b, 2992c, 29002d, 2002e, 2003) for use in the CRA-2004 PA, by correcting the molecular weight of oxalate in FMT_021120.CHEMDAT and adding solid calcium oxalate to this database. Because this analysis did not include any comparisons of measured and predicted solubilities for systems with organic ligands, use of FMT_040628.CHEMDAT gave results identical to those that would have been obtained with FMT_021120.CHEMDAT.

The FMT code and databases are stored on the server named "CCR." Typing "libfmt" accesses the FMT library. The code and databases are stored at the address "PACMS: [CMS_WIPP_NONPA.FMT]." The database used for the CRA-2004 PA calculations is "FMT_021120.CHEMDAT." The updated database used for this uncertainty analysis is "FMT_040628.CHEMDAT." The calculations used for this analysis are in the CMS library at "LIB CRA1V_FMT" in class "CRA_RESP."

3.2.2 An(III) Frequency Distributions and CDF

A histogram of the frequency distribution of Bin N for all An(III) comparisons is shown in Figure 1. A total of 243 measured solubilities were compared with the corresponding predictions. The distribution is relatively broad and peaks at Bin 0 (N = 0) with a frequency of 22.

Table 2 gives values of the corresponding CDF for Bin N for the An(III) comparisons. Figure 2 shows the plotted CDF.

These results show that the An(III) thermodynamic speciation and solubility model implemented in the speciation and solubility code FMT overpredicted the measured An(III) solubilities slightly.

3.2.3 An(IV) Frequency Distributions and CDF

A histogram of the frequency distribution of Bin N for all An(IV) comparisons is shown in Figure 3. A total of 159 measured and predicted solubilities were compared. The distribution is relatively broad and peaks at Bin 1.05 (N = 1.05) with a frequency of 15. There is one occurrence of D in Bin N = -4.95 that was erroneous and should be neglected. (The log of the measured value of this solubility was -8.15 and the log of the predicted value was -8.80, so the difference is 0.65. Therefore, this comparison should have appeared in Bin 0.65, not Bin -4.95.)

Table 3 gives values of the corresponding cumulative distribution function (CDF) for Bin N for AnIV). Figure 4 shows the plotted CDF.

These results show that the An(IV) model in FMT underpredicted the measured An(IV) solubilities. Figure 4 shows that the median value of all An(IV) comparisons is underprediction of the measured solubilities by about 0.5 to 1 log unit.

3.2.4 An(V) Frequency Distributions and CDF

A histogram of the frequency distribution of Bin N for all An(V) comparisons is shown in Figure 5. A total of 136 measured and predicted values were compared. The distribution is relatively narrow and peaks at Bin 0 (N = 0) with a frequency of 44.

Table 4 gives values of the corresponding cumulative distribution function (CDF) for Bin N for An(V). Figure 6 shows the plotted CDF.

These results show that the An(V) model in FMT overpredicted the measured An(III) solubilities slightly.

3.2.5 Combined (An(III, IV, V)) Frequency Distributions and CDF

A histogram of the frequency distribution of Bin N for all combined (An(III, IV, V)) comparisons is shown in Figure 7. A total of 538 measured and predicted values were compared. The distribution is relatively broad and peaks at Bin 0 (N = 0) with a frequency of 71.

Table 5 gives values of the corresponding CDF for Bin N for all An(III, IV, V) comparisons. Figure 8 shows the plotted CDF.

These composite results show that the An(III), An(IV), and An(V) models in FMT underpredicted the measured An(III), An(IV), and An(V) solubilities.

4 CONCLUSIONS

This actinide solubility uncertainty analysis is the first uncertainty analysis carried out to compare measured solubilities and predictions made with FMT since that of Bynum (1996a, 1996b, 1996c).

This updated analysis used both previous (pre-CCA) measurements of actinide solubilities - including values used by Bynum (1996a, 1996b, 1996c) in the analysis for the CCA PA - and new (post-CCA) measurements of actinide solubilities, and predictions made with the latest (post-CCA) version of FMT (Babb and Novak, 1997 and addenda; Wang, 1998) and the most recent FMT thermodynamic database (Xiong, 2004a, 2004b). This analysis included 243 An(III) comparisons, 159 An (IV) comparisons, and 136 An(V) comparisons, for a total of 538 comparisons for all three oxidation states. This analysis provided individual probability distributions for An(III), An(IV), and An(V), and combined results for all three oxidation states.

This analysis included the first comparisons for An(IV), but did not include any comparisons with organic ligands or any An(VI) comparisons.

The results of this analysis are: (1) the An(III) thermodynamic speciation and solubility model implemented in the speciation and solubility code FMT overpredicted the measured An(III) solubilities slightly, (2) the An(IV) model in FMT underpredicted the measured An(IV) solubilities, (3) the An(V) model in FMT overpredicted the measured An(V) solubilities slightly, and (4) overall, the An(III), An(IV), and An(V) models in FMT underpredicted the measured An(III), An(IV), and An(V) solubilities.

5 REFERENCES

Al Mahamid, I., C.F. Novak, K.A. Becraft, S.A. Carpenter, and N. Hakem. 1998. "Solubility of Np(V) in K-Cl-CO_3 and Na-K-Cl-CO_3 Solutions to High Concentrations: Measurements and Thermodynamic Model Predictions," *Radiochimica Acta*. Vol. 81, 93–101.

Babb, S.C. 1996. "NONLIN, Ver. 2.00, User's Manual." January 31, 1996. Albuquerque, NM: Sandia National Laboratories. ERMS 230740.

Babb, S.C., and C.F. Novak. 1995. "WIPP PA User's Manual for FMT, Version 2.0." Albuquerque, NM: Sandia National Laboratories. ERMS 228119.

Babb, S.C., and C.F. Novak. 1997 and addenda. "User's Manual for FMT Version 2.3: A Computer Code Employing the Pitzer Activity Coefficient Formalism for Calculating Thermodynamic Equilibrium in Geochemical Systems to High Electrolyte Concentrations." Albuquerque, NM: Sandia National Laboratories. ERMS 243037.

Baston, G.M.N., M. Brownsword, A.J. Smith, and J.L. Smith-Briggs. 1996. *Further Near-Field Solubility Studies*. NSS/R357. Harwell, UK: AEA Technology plc.

Bynum, R.V. 1996a. "Estimation of Uncertainty for Predicted Actinide Uncertainties." Analysis plan, AP-024, Rev. 0, May 22, 2004. Albuquerque, NM: Sandia National Laboratories. ERMS 410354.

Bynum, R.V. 1996b. "Update of Uncertainty Range and Distribution for Actinide Solubilities to Be Used in CCA NUTS Calculation." Memorandum to M.S. Tierney and C.T. Stockman, May 22, 1996. Albuquerque, NM: Sandia National Laboratories. ERMS 238268.

Bynum, R.V. 1996b. "Analysis to Estimate the Uncertainty for Predicted Actinide Solubilities." Analysis report, Rev. 0, September 6, 1996. Albuquerque, NM: Sandia National Laboratories. ERMS 241374.

Cotsworth, E. 2004c. Untitled letter with enclosure to R.P. Detwiler with third set of CRA comments and requests for additional information, September 2, 2004. Washington, DC: U.S. Environmental Protection Agency Office of Air and Radiation. ERMS 536771.

Felmy, A.R., D. Rai, J.A. Schramke, and J.L. Ryan. 1989. "The Solubility of Plutonium Hydroxide in Dilute Solution and in High-Ionic-Strength Chloride Brines," *Radiochimica Acta*. Vol. 48, 29-35.

Giambalvo, E.R. 2002a. "Recommended Parameter Values for Modeling An(III) Solubility in WIPP Brines." Memorandum to L.H. Brush, July 25, 2002. Carlsbad, NM: Sandia National Laboratories. ERMS 522982.

Giambalvo, E.R. 2002b. "Recommended Parameter Values for Modeling Organic Ligands in WIPP Brines." Memorandum to L.H. Brush, July 25, 2002. Carlsbad, NM: Sandia National Laboratories. ERMS 522981.

Giambalvo, E.R. 2002c. "Recommended Parameter Values for Modeling An(IV) Solubility in WIPP Brines." Memorandum to L.H. Brush, July 26, 2002. Carlsbad, NM: Sandia National Laboratories. ERMS 522986.

Giambalvo, E.R. 2002d. "Recommended Parameter Values for Modeling An(V) Solubility in WIPP Brines." Memorandum to L.H. Brush, July 26, 2002. Carlsbad, NM: Sandia National Laboratories. ERMS 522990.

Giambalvo, E.R. 2002e. "Recommended μ^0/RT Values for Modeling the Solubility of Oxalate Solids in WIPP Brines." Memorandum to L.H. Brush, July 31, 2002. Carlsbad, NM: Sandia National Laboratories. ERMS 523057.

Giambalvo, E.R. 2003. "Release of FMT Database FMT_021120.CHEMDAT." Memorandum to L.H. Brush, March 10, 2003. Carlsbad, NM: Sandia National Laboratories. ERMS 526372.

Hobart, D.E., and R.C. Moore. 1996. "Analysis of Uranium(VI) Solubility Data for WIPP Performance Assessment: Implementation of Analysis Plan AP-028." Analysis report, August 12, 1996. Albuquerque, NM: Sandia National Laboratories. ERMS 236488.

Kirkes, G.R., and S.W. Wagner. 2004. "Analysis Plan for CRA Response Activities." AP 112, Rev. 0, April 26, 2004. Carlsbad, NM: Sandia National Laboratories. ERMS 534690.

Molecke, M.A. 1983. *A Comparison of Brines Relevant to Nuclear Waste Experimentation.* SAND83-0516. Albuquerque, NM: Sandia National Laboratories.

Neck, V., R. Muller, M. Bouby, M. Altmaier, J. Rothe, M.A. Denecke, and J.I. Kim. 2002. "Solubility of Amorphous Thorium (IV) Hydroxide – Application of LIBD to Determine the Solubility Product and EXAFS for Aqueous Speciation," *Radiochimica Acta.* Vol. 90, 485-494.

Novak, C.F. 1997. "Calculation of Actinide Solubilities in WIPP SPC and ERDA-6 Brines under MgO Backfill Scenarios Containing either Nesquehonite or Hydromagnesite as the Mg-CO_3 Solubility-Limiting Phase." Memorandum to R.V. Bynum, April 21, 1997. Albuquerque, NM: Sandia National Laboratories. ERMS 246124.

Novak, C.F., I. Al Mahamid, K.A. Becraft, S.A. Carpenter, N. Hakem, and T. Prussin. 1997. "Measurement and Thermodynamic Modeling of Np(V) Solubility in Aqueous K_2CO_3 Solutions to High Concentrations," *Journal of Solution Chemistry*. Vol. 26, 681-697.

Novak, C.F., R.C. Moore, and R.V. Bynum. 1996. "Prediction of Dissolved Actinide Concentrations in Concentrated Electrolyte Solutions: A Conceptual Model and Model Results for the Waste Isolation Pilot Plant (WIPP)." Unpublished presentation at the 1996 International Conference on Deep Geological Disposal of Radioactive Waste, September 16-19, 1996, Winnipeg, Manitoba, Canada. ERMS 418098, SAND96-2695C.

Novak, C.F., H. Nitsche, H.B. Silber, K. Roberts, P.C. Torretto, T. Prussin, K. Becraft, S.A. Carpenter, D.E. Hobart, and I. Al Mahamid. 1996 "Neptunium(V) and Neptunium(VI) Solubilities in Synthetic Brines of Interest to the Waste Isolation Pilot Plant (WIPP)," *Radiochimica Acta*. Vol. 74, 31–36.

Osthlos, E., J. Bruno, and I. Grenthe. 1994. "On the Influence of Carbonate on Mineral Dissolution. III. The Solubility of Microcrystalline ThO_2 in CO_2-H_2O Media," *Geochimica et Cosmochimica Acta*. Vol. 58, 613-623.

Popielak, R.S., R.L. Beauheim, S.R. Black, W.E. Coons, C.T. Ellingson and R.L. Olsen. 1983. *Brine Reservoirs in the Castile Formation, Waste Isolation Pilot Plant Project, Southeastern New Mexico*. TME 3153. Carlsbad, NM: U.S. Department of Energy WIPP Project Office.

Rao, L., D. Rai, A.R. Felmy, and C.F. Novak. "Solubility of $NaNd(CO_3)_2 \cdot 6H_2O(c)$ in Mixed Electrolyte (Na-Cl-CO_3-HCO_3) and Synthetic Brine Solutions," *Actinide Speciation in High Ionic Strength Media.* Eds. D.T. Reed, S.B. Clark, and L. Rao. New York, NY: Kluwer Academic/Plenum Publishers. 153-169.

Runde, W., and J.I. Kim. 1995. *Untersuchungen der Übertragbarkeit von Labordaten natürliche Verhältnisse: Chemisches Verhalten von drei- und fünfwertigem Americium in salinen NaCl-Lösungen (Study of the Extrapolability of Laboratory Data to Natural Conditions: Chemical Behavior of Trivalent and Pentavalent Americium in Saline NaCl Solutions)*. RCM-01094. Munich, FRG: Institute for Radiochemistry, Technical University of Munich. ERMS 241862.

Silva, R.J. 1982. *The Solubilities of Crystalline Neodymium and Americium Trihydroxides*. LBL-15055. Berkeley, CA: Lawrence Berkeley National Laboratory.

U.S. DOE. 1996b. *Title 40 CFR Part 191 Compliance Certification Application for the Waste Isolation Pilot Plant, Vol. 1-21.* DOE/CAO-1994-2184. Carlsbad, NM: U.S. Department of Energy Carlsbad Area Office.

U.S. DOE. 2004. *Title 40 CFR Part 191 Compliance Recertification Application for the Waste Isolation Pilot Plant, Vol. 1-8.* DOE/WIPP 2004-3231. Carlsbad, NM: U.S. Department of Energy Carlsbad Field Office.

Wang, Y. 1998. "WIPP PA Validation Document for FMT (Version 2.4), Document Version 2.4." Carlsbad, NM: Sandia National Laboratories. ERMS 251587.

Xiong, Y.-L. 2004a. "A Correction of the Molecular Weight of Oxalate in FMT_021120.CHEMDAT, and Incorporation of Calcium Oxalate Monohydrate (Whewellite) into CHEMDAT with Its Recommended Dimensionless Standard Chemical Potential (μ^0/RT) Value." Memorandum to L.H. Brush, June 8, 2004. Carlsbad, NM: Sandia National Laboratories. ERMS 535813.

Xiong, Y.-L. 2004b. Release of FMT_040628.CHEMDAT. E-mail to J.J. Long, June 28, 2004. Carlsbad, NM: Sandia National Laboratories. ERMS 536022.

6 FIGURES

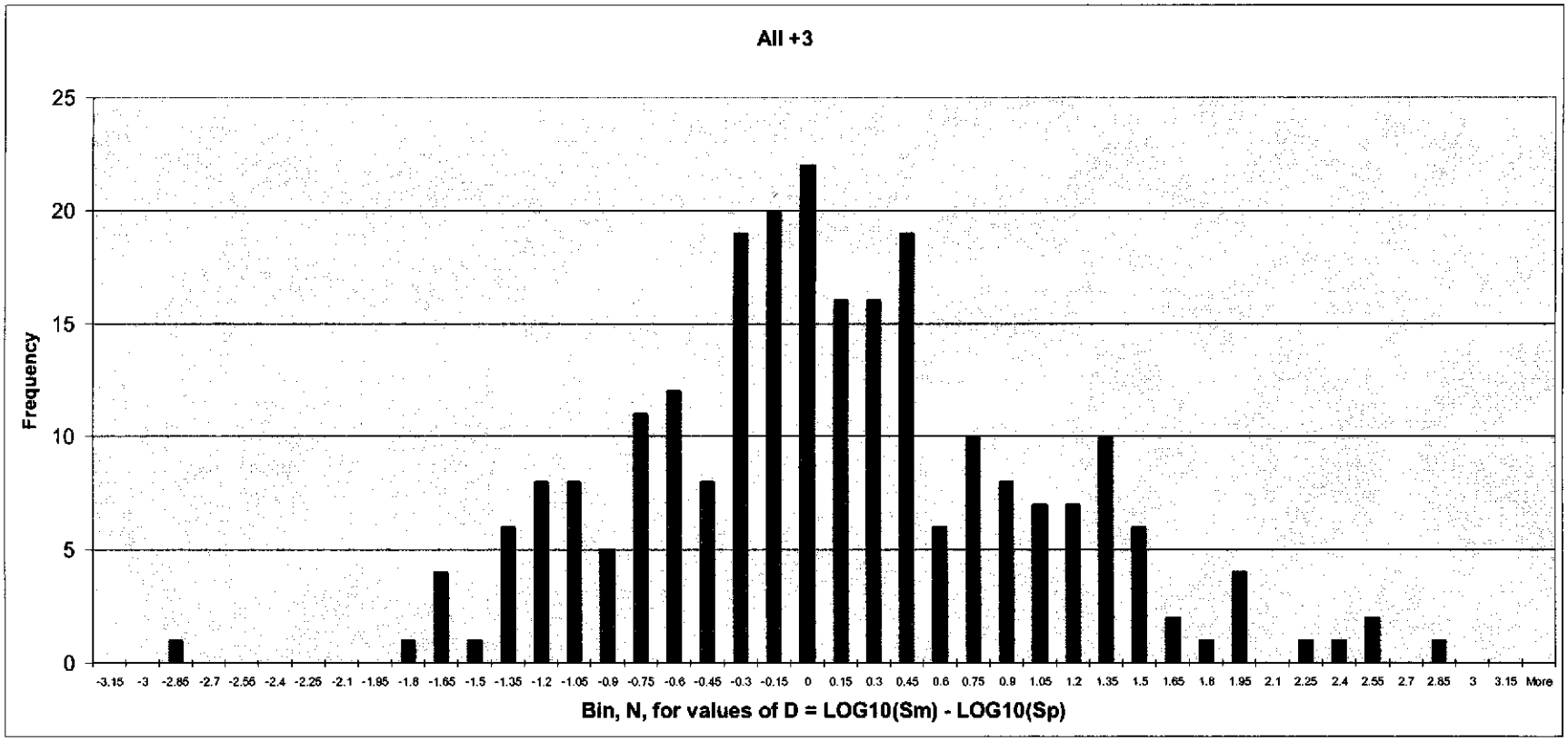

Figure 1. Histogram of frequency distribution of Bin N for all An(III) comparisons. A total of 243 measured and predicted solubilities were compared.

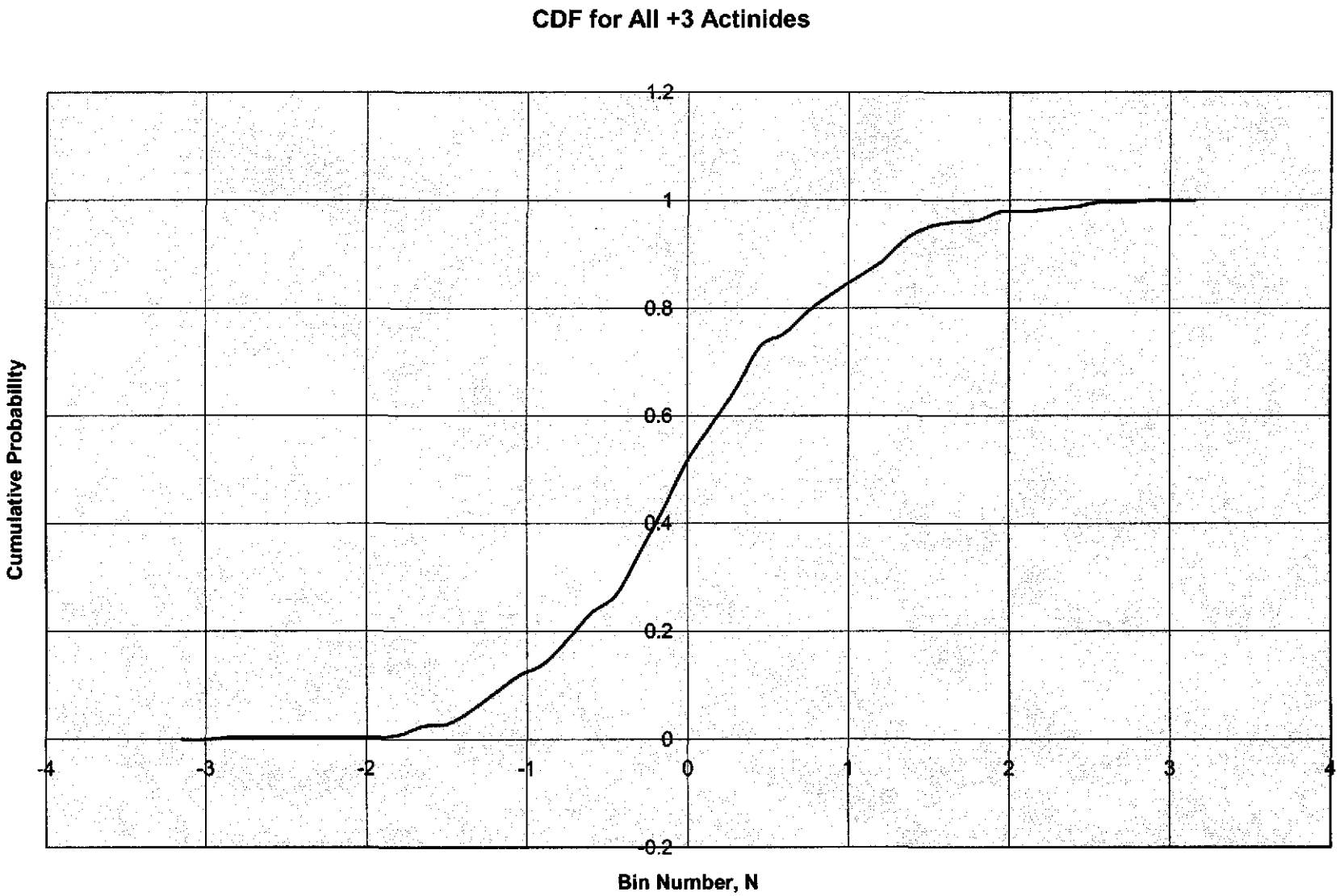

Figure 2. Plot of the CDF for Bin N for An(III).

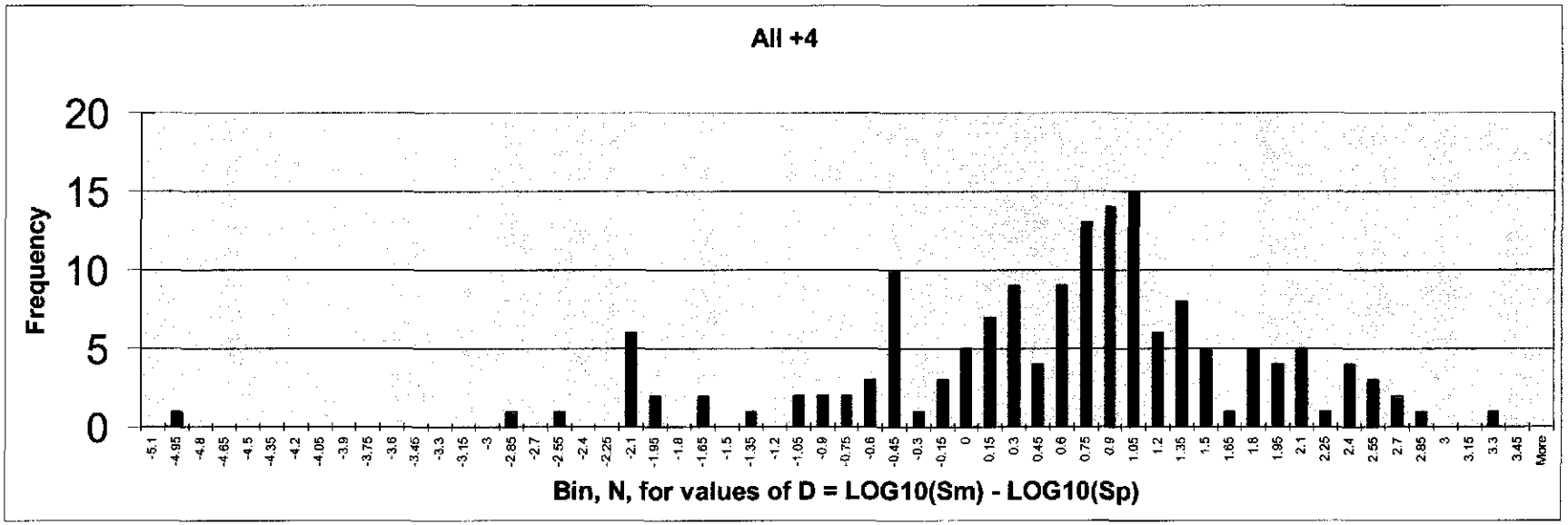

Figure 3. Histogram of Frequency Distribution of Bin N for all An(IV) Comparisons. A total of 159 measured and predicted solubilities were compared.

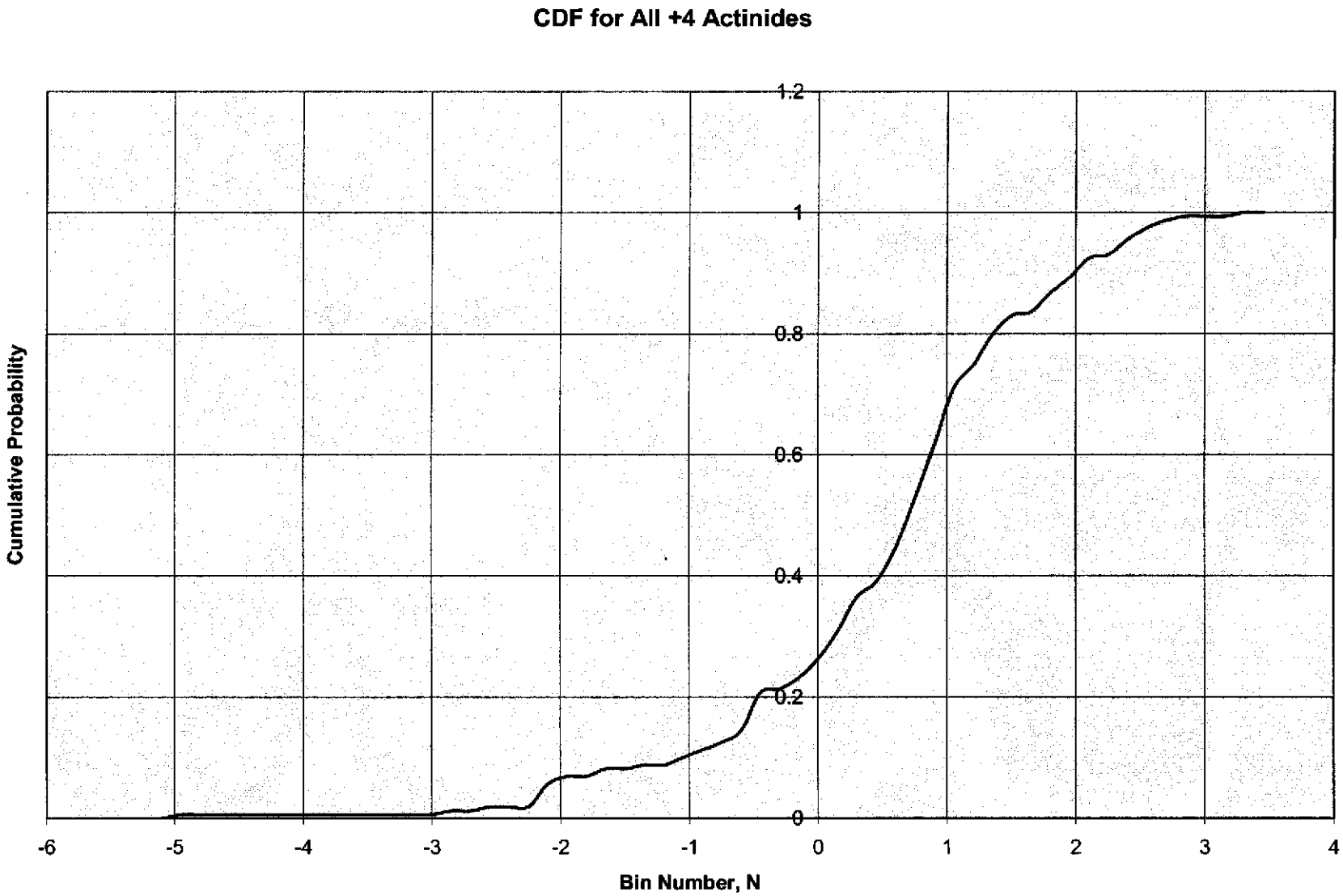

Figure 4. Plot of the CDF for Bin N for An(IV).

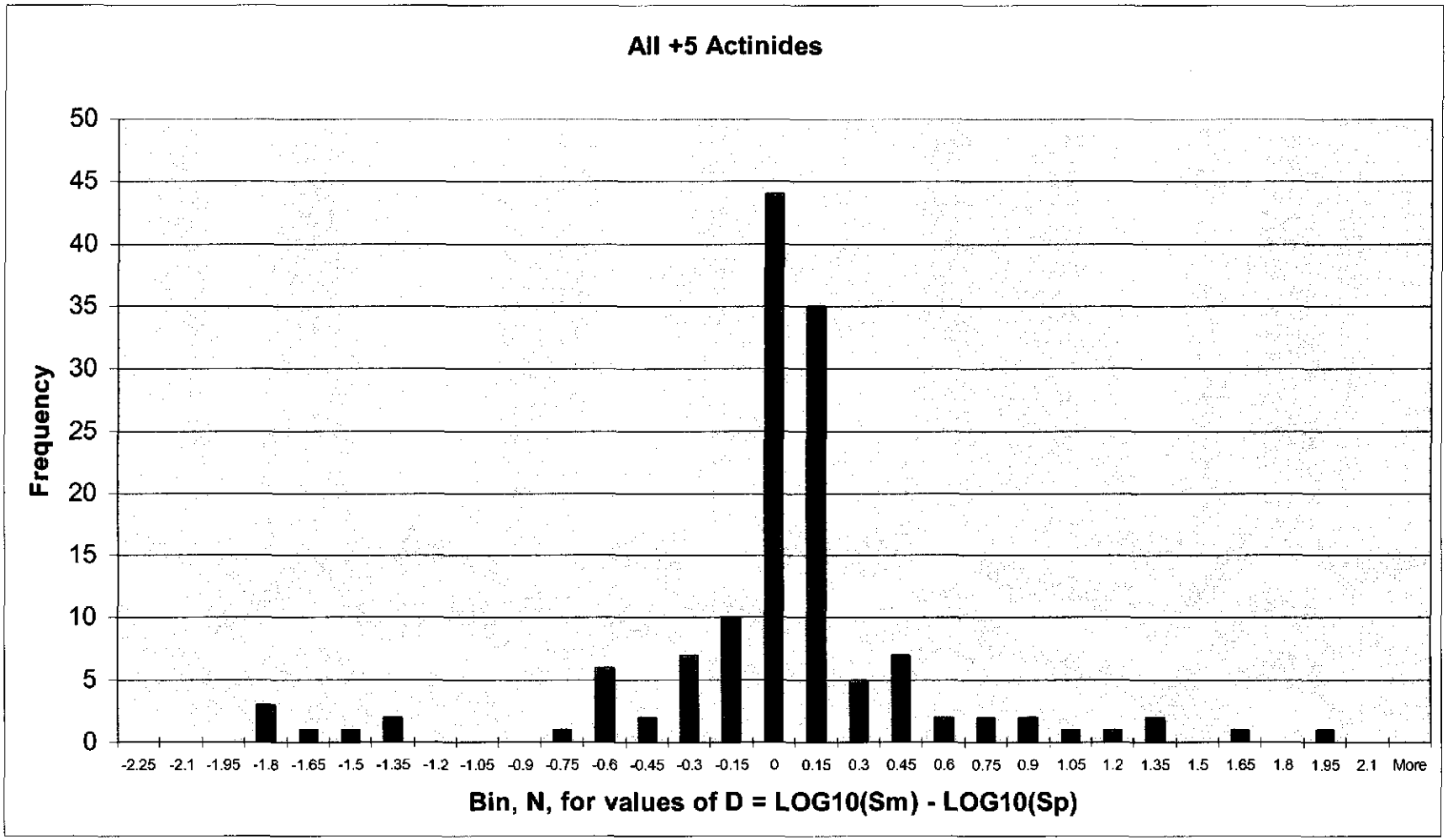

Figure 5. Histogram of Frequency Distribution of Bin N for all An(V) Comparisons. A total of 136 measured and predicted solubilities were compared.

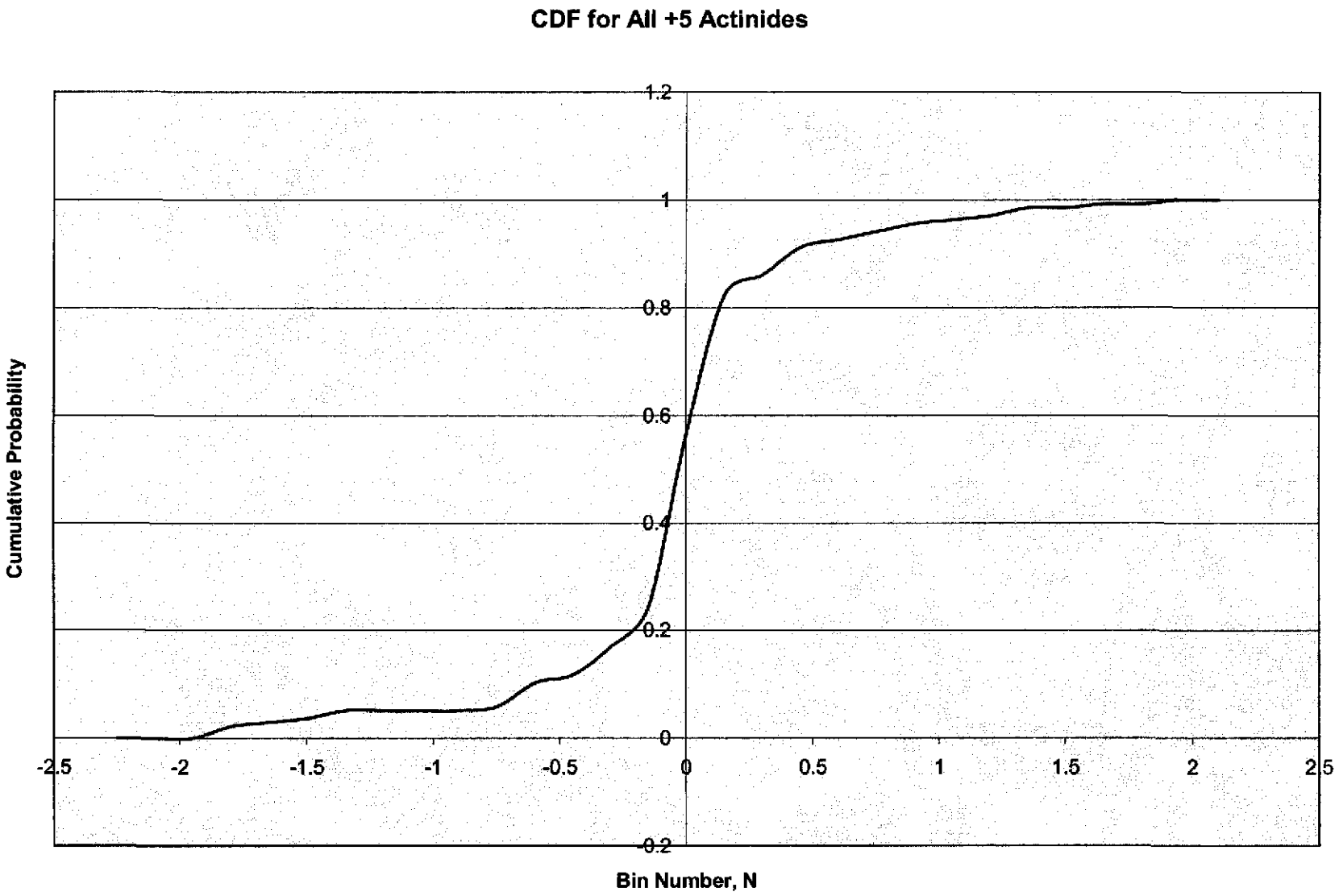

Figure 6. Plot of the CDF for Bin N for An(V).

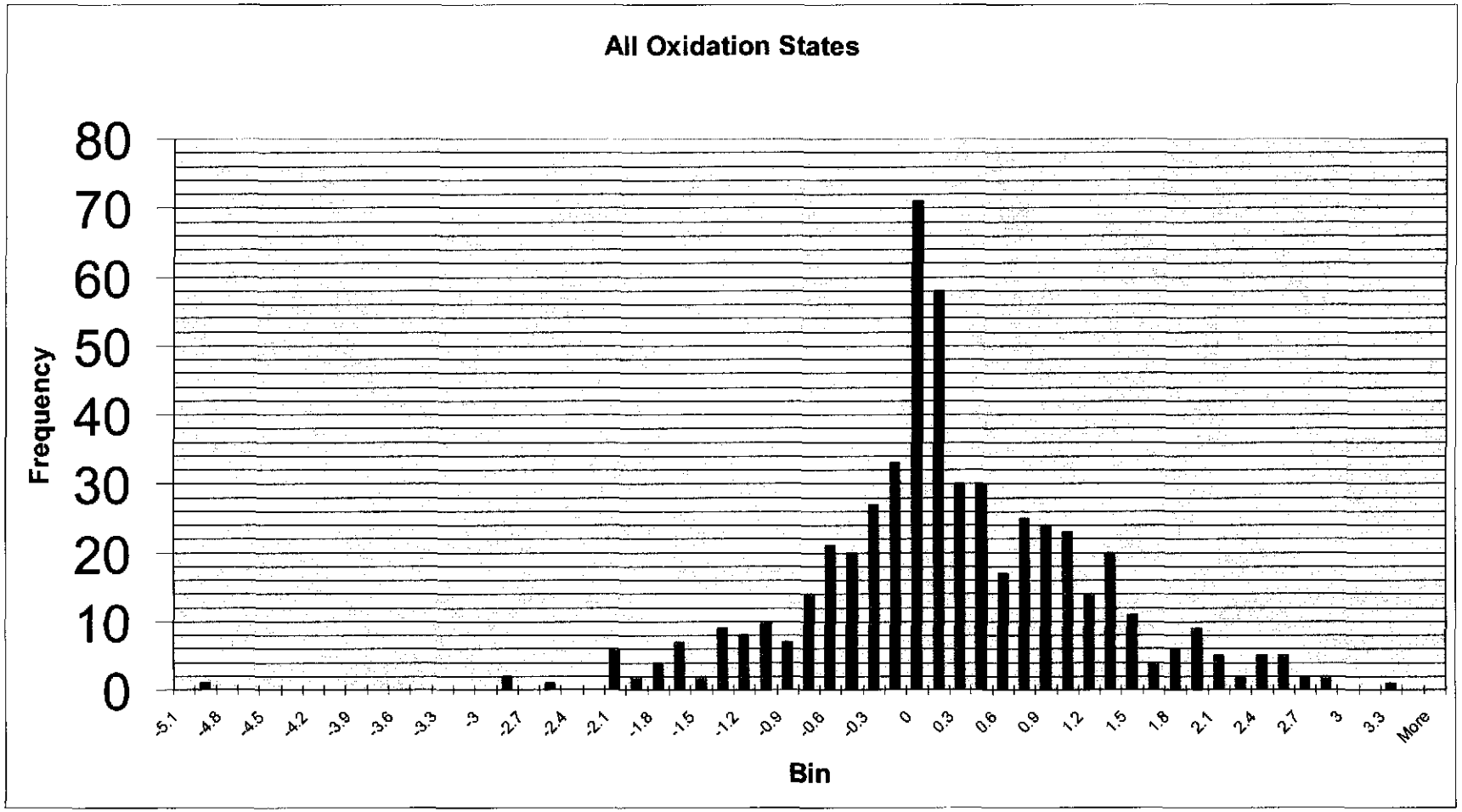

Figure 7. Histogram of Frequency Distribution of Bin N for all combined (An(III, IV, V)) Comparisons. A total of 538 measured and predicted solubilities were compared.

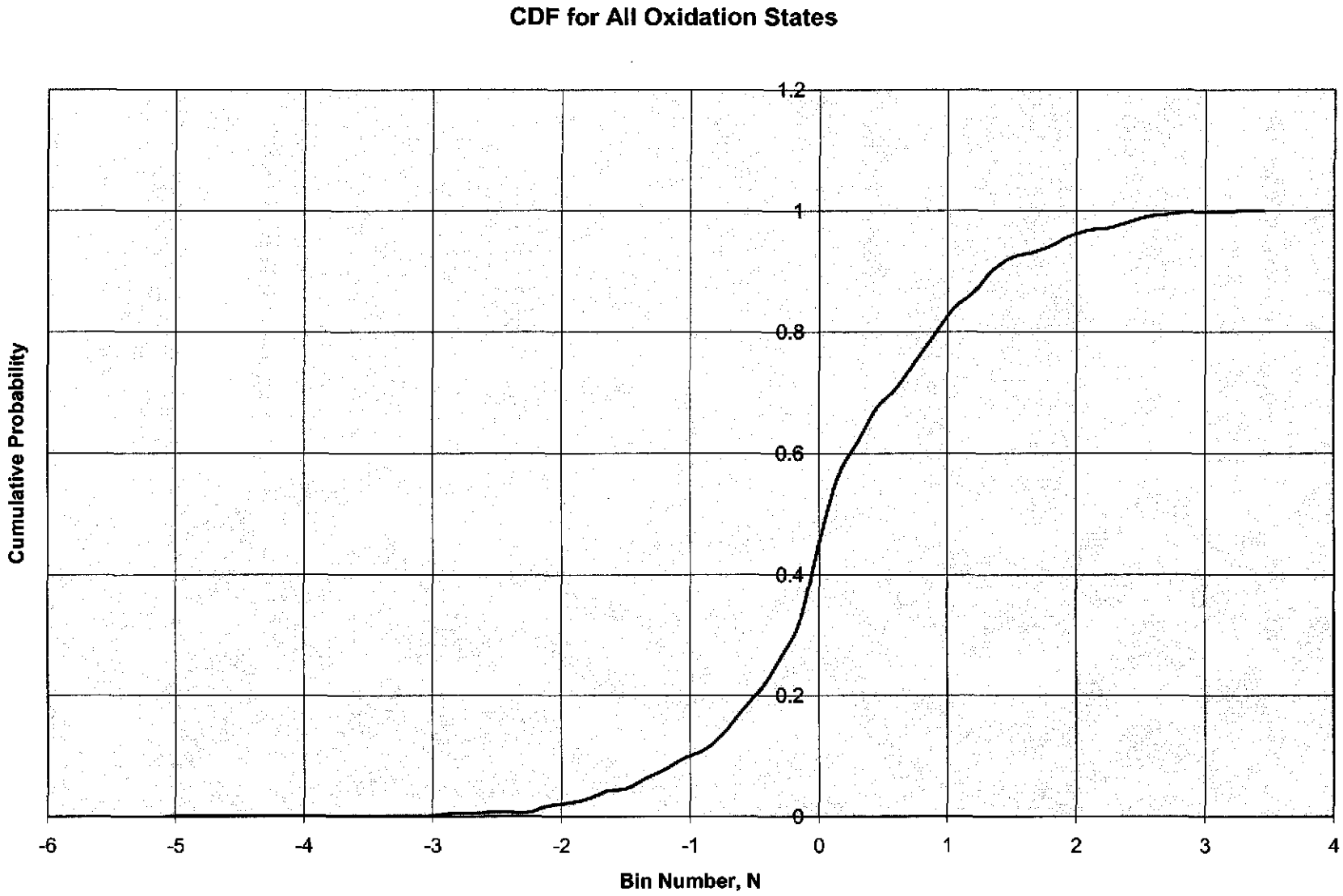

Figure 8. Plot of the CDF for Bin N for all three oxidation states (An(III, IV, V)).

7 TABLES

Table 1. Sources of Measured Actinide Solubilities.

Oxidation State	Solution	Ionic Strength, (M)	pH	Solubility-Controlling Solid	Reference, Type of Data Source	Remarks
+III	NaCl-KCl and $MgCl_2$ brine (similar to Brine A)	7.8	6.4-8.4[A]	$Nd(OH)_3(am)$	Khalili et al. (1994)	Measured from published plots
+III	0 M NaCl, 0.1-1.1 m $NaHCO_3$, or 0.1-2 m Na_2CO_3	-	-	$NaNd(CO_3)_2 \cdot 6H_2O$	*Rao et al. (1999)	Measured from published plots. *Bynum (1996a&b) used 25 meas. points(Na_2CO_3, and 20 meas. points ($NaHCO_3$).*
+III	2 M NaCl, 0.1-0.5 m $NaHCO_3$, or 0.1-2 m Na_2CO_3	-	-	$NaNd(CO_3)_2 \cdot 6H_2O$	*Rao et al. (1999)	Measured from published plots. Bynum (1996a&b) used 11 meas. points (Na2CO3, and 8 meas. points (NaHCO3)

A. pcH.

Table 1. Sources of Measured Actinide Solubilities (cont.).

Oxidation State	Solution	Ionic Strength, (M)	pH	Solubility-Controlling Solid	Reference, Type of Data Source	Remarks
+III	4 M NaCl, 0.1–2 m Na_2CO_3	-	-	$NaNd(CO_3)_2 \cdot 6H_2O$	*Rao et al. (1999)	Measured from published plots. (Bynum (1996a, b) used 11 meas. points.)
+III	ERDA-6	6.7	6.38-10.62[A]	$NaNd(CO_3)_2 \cdot 6H_2O$	*Rao et al. (1999)	Measured from published plots. (Bynum (1996a, b) used 21 measured points.)
+III	G-Seep	7.3	5.81-7.77[A]	$NaNd(CO_3)_2 \cdot 6H_2O$	*Rao et al. (1999)	Measured from published plots. (Bynum (1996a, b) used 7 measured points.)
+III	5.0 M NaCl or 5.0 m NaCl under 10^{-2} atm CO_2	5.0	6.57-12.47[A]	$Am(OH)_3$(cr) or $NaAm(CO_3)_3 \cdot xH_2O$(cr)	*Runde and Kim (1995)	Measured from published plots. *Bynum (1996a&b) used 35 meas. points*
+III	0.1 M $NaClO_4$	0.1	7.05-9.43	$Am(OH)_3$(cr)	Silva (1982)	Taken from published tables.
+III	0.1 M $NaClO_4$	0.1	5.67-9.52	$Nd(OH)_3$(cr)	Silva (1982)	Taken from published tables.

A. pcH.

Table 1. Sources of Measured Actinide Solubilities (cont.).

Oxidation State	Solution	Ionic Strength, (M)	pH	Solubility-Controlling Solid	Reference, Type of Data Source	Remarks
+IV	Nirex	0.02	10-12	ThO_2(am)	Baston et al. (1996)	Taken from published tables
+IV	0.6 M NaCl, 1.2 M NaCl, 3.0 M NaCl, or 0.6 M KCl	-		ThO_2	Felmy et al. (1991)	Taken from published tables
+IV	0.5 M NaCl	0.5	6.06-11.73	$Th(OH)_4$(am)	Neck et al. (2002)	Taken from published tables
+IV	0.5 M $NaClO_4$	0.5	8.21-10.45	ThO_2(micro cr)	Osthlos et al. (1994)	Taken from published tables

Table 1. Sources of Measured Actinide Solubilities (cont.).

Oxidation State	Solution	Ionic Strength, (M)	pH	Solubility-Controlling Solid	Reference, Type of Data Source	Remarks
+V	$KCl + K_2CO_3$	0.0064-3.19	10.50-10.86	$KNpO_2CO_3(cr)$	*Al Mahamid et al. (1998)	Taken from published tables. (Bynum (1996a, b) used 2 points @ 0.0032 M KCL, 3 points @ 0.032 M KCl, 3 points @ 0.32 M KCl, and 3 points @ 3.2 M KCl.)
+V	$KCl + K_2CO_3$	3.71-3.94	11.77-11.80	$K_3NpO_2(CO_3)_2$	Al Mahamid et al. (1998)	Taken from published tables
+V	$NaCl + KCl + Na_2CO_3$	4.58-4.60	9.79-10.36	$KNpO_2CO_3(cr)$	Al Mahamid et al. (1998)	Taken from published tables
+V	$NaCl + KCl + Na_2CO_3$	4.60-7.08	10.47-11.61	$Na_3NpO_2(CO_3)_2(cr)$	Al Mahamid et al. (1998)	Taken from published tables
+V	WIPP AISinR brine	0.85	7.67	$KNpO_2CO_3(cr)$	Novak et al. (1996)	Taken from published tables
+V	WIPP H 17 brine	2.82	7.65	$KNpO_2CO_3(cr)$	Novak et al. (1996)	Taken from published tables

Table 1. Sources of Measured Actinide Solubilities (cont.).

Oxidation State	Solution	Ionic Strength, (M)	pH	Solubility-Controlling Solid	Reference, Type of Data Source	Remarks
+V	WIPP SPC brine	7.08	8.56	$KNpO_2CO_3$(cr)	Novak et al. (1996)	Taken from published tables
+V	0.011 M to 0.401 M K_2CO_3	0.033-0.69	11.02-11.48	$KNpO_2CO_3$(cr)	Novak et al. (1997)	Taken from published tables
+V	0.249 M to 4.83 M K_2CO_3	0.747-14.43	11.50-13.26	$K_3NpO_2(CO_3)_2$	Novak et al. (1997)	Taken from published tables
+V	1 M NaCl under 10^{-2} atm CO_2, or 5 M NaCl under 10^{-2} atm CO_2	1.0-5.0	6.19-8.78 5.65-8.51	$NaNpO_2CO_3 \cdot xH_2O$(cr)	Runde and Kim (1995)	Taken from published tables

Table 2. Values of the CDF for All An(III) Solubility Predictions.

Bin, N	CDF for All +III
-3.15	0
-3.00	0
-2.85	0.004115226
-2.70	0.004115226
-2.55	0.004115226
-2.40	0.004115226
-2.25	0.004115226
-2.10	0.004115226
-1.95	0.004115226
-1.80	0.008230453
-1.65	0.024691358
-1.50	0.028806584
-1.35	0.053497942
-1.20	0.086419753
-1.05	0.119341564
-0.90	0.139917695
-0.75	0.185185185
-0.60	0.234567901
-0.45	0.267489712
-0.30	0.345679012
-0.15	0.427983539
0.00	0.518518519
0.15	0.58436214
0.30	0.650205761
0.45	0.728395062
0.60	0.75308642
0.75	0.794238683
0.90	0.827160494
1.05	0.855967078
1.20	0.884773663
1.35	0.925925926
1.50	0.950617284
1.65	0.958847737
1.80	0.962962963
1.95	0.979423868
2.10	0.979423868
2.25	0.983539095
2.40	0.987654321

Note: Table 2 continued on next page.

Table 2. Values of the CDF for All An(III) Solubility Predictions (cont.).

Bin, N	CDF for All +III
2.55	0.995884774
2.70	0.995884774
2.85	1
3.00	1
3.15	1

Table 3. Values of the CDF for All An(IV) Solubility Predictions.

Bin, N	CDF for All +IV
-5.10	0
-4.95	0.006289
-4.80	0.006289
-4.65	0.006289
-4.50	0.006289
-4.35	0.006289
-4.20	0.006289
-4.05	0.006289
-3.90	0.006289
-3.75	0.006289
-3.60	0.006289
-3.45	0.006289
-3.30	0.006289
-3.15	0.006289
-3.00	0.006289
-2.85	0.012579
-2.70	0.012579
-2.55	0.018868
-2.40	0.018868
-2.25	0.018868
-2.10	0.056604
-1.95	0.069182
-1.80	0.069182
-1.65	0.081761
-1.50	0.081761
-1.35	0.08805
-1.20	0.08805
-1.05	0.100629
-0.90	0.113208
-0.75	0.125786
-0.60	0.144654
-0.45	0.207547
-0.30	0.213836
-0.15	0.232704
0.00	0.264151
0.15	0.308176
0.3	0.36478
0.45	0.389937

Note: Table 3 continued on next page.

Table 3. Values of the CDF for All An(IV) Solubility Predictions (cont.).

Bin, N	CDF for All +IV
0.6	0.446541
0.75	0.528302
0.9	0.616352
1.05	0.710692
1.2	0.748428
1.35	0.798742
1.5	0.830189
1.65	0.836478
1.8	0.867925
1.95	0.893082
2.1	0.924528
2.25	0.930818
2.4	0.955975
2.55	0.974843
2.7	0.987421
2.85	0.993711
3	0.993711
3.15	0.993711
3.3	1
3.45	1

Table 4. Values of the CDF for All An(V) Solubility Predictions.

Bin, N	CDF for all +V
-2.25	0
-2.10	0
-1.95	0
-1.80	0.022059
-1.65	0.029412
-1.50	0.036765
-1.35	0.051471
-1.20	0.051471
-1.05	0.051471
-0.90	0.051471
-0.75	0.058824
-0.60	0.102941
-0.45	0.117647
-0.30	0.169118
-0.15	0.242647
0.00	0.566176
0.15	0.823529
0.30	0.860294
0.45	0.911765
0.60	0.926471
0.75	0.941176
0.90	0.955882
1.05	0.963235
1.20	0.970588
1.35	0.985294
1.50	0.985294
1.65	0.992647
1.80	0.992647
1.95	1
2.10	1

Table 5. Values of the CDF for Combined An(III, IV, V) Solubility Predictions.

Bin, N	CDF for all three oxidation states (An(III, IV, V))
-5.10	0
-4.95	0.001859
-4.80	0.001859
-4.65	0.001859
-4.50	0.001859
-4.35	0.001859
-4.20	0.001859
-4.05	0.001859
-3.90	0.001859
-3.75	0.001859
-3.60	0.001859
-3.45	0.001859
-3.30	0.001859
-3.15	0.001859
-3.00	0.001859
-2.85	0.005576
-2.70	0.005576
-2.55	0.007435
-2.40	0.007435
-2.25	0.007435
-2.10	0.018587
-1.95	0.022305
-1.80	0.02974
-1.65	0.042751
-1.50	0.046468
-1.35	0.063197
-1.20	0.078067
-1.05	0.096654
-0.90	0.109665
-0.75	0.135688
-0.60	0.174721
-0.45	0.211896
-0.30	0.262082
-0.15	0.32342
0.00	0.45539
0.15	0.563197
0.30	0.618959

Note: Table 5 continued on next page.

Table 5. Values of the CDF for Combined An(III, IV, V) Solubility Predictions cont.).

Bin, N	CDF for all three oxidation states (An(III, IV, V))
0.45	0.674721
0.60	0.70632
0.75	0.752788
0.90	0.797398
1.05	0.840149
1.20	0.866171
1.35	0.903346
1.50	0.923792
1.65	0.931227
1.80	0.942379
1.95	0.959108
2.10	0.968401
2.25	0.972119
2.40	0.981413
2.55	0.990706
2.70	0.994424
2.85	0.998141
3.00	0.998141
3.15	0.998141
3.30	1
3.45	1

Brush, Laurence H L. H. Brush

From: jimnowak [jimnowak@ixpn.com]
Sent: Thursday, December 16, 2004 6:33 PM
To: Brush, Laurence H

Larry,

Below is a statement authorizing you to sign for me on the report:
****

I authorize Laurence H. (Larry) Brush, one of my co-authors, to sign for me as co-author on "Updated Uncertainty Analysis of Actinide Solubilities for the Response to EPA Comment C-23-16."

Edwin J. Nowak

****
Have fun at the final signing.

We're getting very enjoyable rest here.

Jim