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Sandia National Laboratories
Waste Isolation Pilot Plant

Updated Uncertainty Analysis of Actinide Solubilities For the Response to EPA Comment C-23-16, Rev. 1 (Supersedes ERMS 538219)

Work carried out under Task 3 of the Analysis Plan for the Calculation of Actinide Solubilities for the WIPP PABC, AP-120, Rev. 0

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1/50

TABLE OF CONTENTS

1 ABBREVIATIONS, ACRONYMS, AND INITIALISMS

Table 1 (see pp. 37-38 below) defines the abbreviations, acronyms, and initialisms used in this analysis report.

2 REVISION HISTORY

This is Revision (Rev.) 1 of this analysis report. Rev. 1 features the following changes from the original version of Xiong et al. (2004): (1) establishment of a revised uncertainty range and probability distribution for the solubilities of actinide (An) elements in the +IV oxidation state (An(IV) solubilities); (2) revision of the combined range and distribution for actinides in the +III, +IV, and +V oxidation states (An(III), An(IV), and An(V); (3) additions to and modifications of much of the text, especially Subsection 5.2.3; and (4) revised versions of Figures 3, 4, 7, and 8; and Tables 1, 3, and 5 from Xiong et al. (2004); and (5) seven entirely new figures (Figures 3-9) that were not in Xiong et al. (2004). This analysis was carried out under Task 3 of Brush and Xiong's (2005) analysis plan (AP) for the actinide-solubility calculations for the Performance Assessment Baseline Calculations (PABC) for the first Compliance Recertification Application (CRA-2004) for the U.S Department of Energy's (DOE's) Waste Isolation Pilot Plant (WIPP).

Xiong et al. (2004) carried out the original version of this analysis under the analysis plan (AP) for response activities related to the CRA-2004 (Kirkes and Wagner, 2004). Xiong et al. (2004) concluded that (1) the An(III) thermodynamic speciation and solubility model implemented in the speciation and solubility code Fracture-Matrix Transport (FMT) (Babb and Novak, 1997 and addenda; Wang, 1998) slightly overpredicted the measured An(III) solubilities; (2) the An(IV) model in FMT significantly underpredicted the measured An(IV) solubilities; (3) the An(V) model in FMT slightly overpredicted the measured An(V) solubilities; and (4) overall, the An(III), An(IV), and An(V) models in FMT together significantly underpredicted the measured An(III), An(IV), and An(V) solubilities. Xiong et al. (2004) used the thermodynamic database FMT_040628.CHEMDAT for their analysis.

The An(IV) model underpredicted the measured An(IV) solubilities to a significantly greater extent than the An(III) and An(V) models overpredicted the measured An(III) and An(V) solubilities, respectively. Therefore, Brush and Xiong (2005, Subsection 7.2) included Task 2 to identify and correct the cause of An(IV) underpredictions prior to starting the FMT calculations for the PABC (see Brush and Xiong, 2005, Subsection 7.2). Nowak (2005) identified the value of the dimensionless standard chemical potential (μ^0/RT) for $Th(OH)_4(aq)$ in FMT_040628.CHEMDAT, -622.4700, as the cause of this problem; and recommended that μ^0/RT for $Th(OH)_4(aq)$ be changed from -622.4700 to -626.5853. Xiong (2005) released the corrected version of the database, FMT_050405.CHEMDAT. Note that in the AP describing this work, Brush and Xiong (2005, Subsection. 7.2) tentatively concluded that μ^0/RT for $Th(OH)_4(aq)$ should be changed from -622.4700 to -626.8467. However, Nowak (2005) recommended revising this parameter to -626.5853 to avoid having to make changes in the parameters for other Th(IV) species in the FMT database. (The value tentatively recommended by Brush and Xiong (2005) was based on the formation constant for $Th(OH)_4(aq)$ in another database, in which the values of the parameters for several other Th(IV) species are slightly different from those in the FMT database.)

We used FMT_050405.CHEMDAT (Nowak, 2005; Xiong, 2005) to establish a revised uncertainty range and probability distribution for An(IV) solubility predictions (see Subsection 5.2.3 below) and a revised composite range and distribution for An(III), An(IV), and An(V) solubility predictions (Subsections 5.2.5). The ranges and distributions for An(III) and An(V) solubility predictions are unchanged from Xiong et al. (2004).

3 INTRODUCTION

The U.S. Environmental Protection Agency (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."

The original version of this analysis (Xiong et al., 2004) responded 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 used 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 derived a frequency distribution for differences between measured solubilities and those calculated (predicted) for the same conditions. The frequency distribution was used to represent the expected solubility uncertainty distribution.

For Rev. 1 of this analysis, we used methods identical to those of Xiong et al. (2004) to establish a revised uncertainty range and probability distribution for An(IV) solubility predictions (see Subsection 5.2.3 below) and a revised composite range and distribution for An(III), An(IV), and An(V) solubility predictions (Subsections 5.2.5). The ranges and distributions for An(III) and An(V) solubility predictions are unchanged from those of Xiong et al. (2004).

4 BACKGROUND

Novak et al. (1996a) used FMT to predict An(III), An(IV), and An(V) solubilities 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 then. 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. The FMT calculations of An(III), An(IV), and An(V) solubilities for the CRA-2004 PA is described in detail by U.S. DOE (2004, Appendix PA, Attachment SOTERM). After these calculations, Xiong (2004a) modified FMT_021120.CHEMDAT by correcting the molecular weight of oxalate in FMT_021120.CHEMDAT and adding solid calcium oxalate to this database; and Xiong (2004b) released FMT_040628.CHEMDAT. Subsequently, Xiong (2004c) modified μ^0/RT for $NpO_2Ac(aq)$ in FMT_040628.CHEMDAT, and Xiong (2004d) released FMT_041116.CHEMDAT. (Note that, in this report, the "Ac" in "$NpO_2Ac(aq)$" stands for the organic ligand acetate, not for the chemical element actinium.) Then Xiong (2004e) changed

μ^0/RT for $NpO_2Ac(aq)$ back to its original value, and Xiong (2004f) released FMT_041210.CHEMDAT.

Xiong et al. (2004) carried out the original version of the first uncertainty analysis of FMT predictions of actinide solubilities since that of Bynum (1996a, 1996b, 1996c). Xiong et al. (2004) used FMT_040628.CHEMDAT for their analysis. Note that the changes in μ^0/RT for $NpO_2Ac(aq)$ had no effect on the analysis of Xiong et al. (2004) because their analysis did not include any comparisons with solubilities measured with organic ligands such as acetate.)

Because the An(IV) model significantly underpredicted measured An(IV) solubilities, Nowak (2005) recommended that μ^0/RT for $Th(OH)_4(aq)$ be changed from -622.4700 to -626.5853 and Xiong (2005) released FMT_050405.CHEMDAT. Although Brush and Xiong (2005, Subsection. 7.2) tentatively concluded that μ^0/RT for $Th(OH)_4(aq)$ should be changed from -622.4700 to -626.8467, Nowak (2005) recommended revising this parameter to -626.5853. This avoided having to make changes in the parameters for other Th(IV) species in the FMT database. (The value tentatively recommended by Brush and Xiong (2005) was based on the formation constant for $Th(OH)_4(aq)$ in another database, in which the values of the parameters for several other Th(IV) species are slightly different from those in the FMT database.)

We used FMT_050405.CHEMDAT (Nowak, 2005; Xiong, 2005) to establish a revised uncertainty range and probability distribution for An(IV) solubility predictions (see Subsection 5.2.3 below) and a revised composite range and distribution for An(III), An(IV), and An(V) solubility predictions (Subsections 5.2.5). The ranges and distributions for An(III) and An(V) solubility predictions are unchanged from Xiong et al. (2004).

5 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, 2005). This 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 from each study included in this analysis (see Table 2 on pp. 39-42 below), 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 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 state. (It used estimates by Hobart and Moore (1996) for the CCA PA, the PAVT, and the CRA-2004 PA.)

5.1 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 2 (pp. 39-42) with values of major dissolved constituents, ionic strength, pH, solubility-controlling solid phases, and citations. Included are solubilities measured in synthetic WIPP brines such as ERDA-6, G-Seep, and SPC. 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.

5.2 Frequency Distribution of Differences between Measured and Predicted Solubilities

Frequency distributions of differences (D) between logarithms (base 10) of measured and predicted actinide solubilites (S_m and S_p, respectively) 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 S_m or S_p values (or logarithms thereof), frequency distributions, and histograms for the analysis in the original version of this analysis (Xiong et al., 2004). File "WIPP Solubility Uncertainty Values RevC_4-20-05.xls" contains additional plots based on the calculations in the original version of this analysis. File "New WIPP +IV Uncertainty Analysis 4-20-05.xls" contains spreadsheets with S_m or S_p values (or logarithms thereof), frequency distributions, and histograms used to establish a revised uncertainty range and probability distribution for An(IV) solubility predictions (see Subsection 5.2.3 below) and a revised composite range and distribution for An(III), An(IV), and An(V) solubility predictions (Subsection 5.2.5). 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 $D = Log_{10}(S_m) - Log_{10}(S_p)$.
>
> 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)

5.2.1 Updated FMT Database Used for This Revised Analysis

Xiong et al. (2004) used FMT_040628.CHEMDAT for the original version of this analysis. We used FMT_050405.CHEMDAT (Nowak, 2005; Xiong, 2005) to establish a revised uncertainty range and probability distribution for An(IV) solubility predictions (see Subsection 5.2.3 below) and a revised composite range and distribution for An(III), An(IV), and An(V) solubility predictions (Subsections 5.2.5). Section 4 of this analysis report provides a detailed revision history of the FMT database since its first use for compliance-related calculations in 1996 (see above).

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 was FMT_021120.CHEMDAT. The database used for the original version of this uncertainty analysis was "FMT_040628.CHEMDAT. The database used for Rev. 1 of this analysis was FMT_050405.CHEMDAT. The calculations used for this analysis are in the CMS library at LIB CRA1V_FMT in class CRA_RESP.

5.2.2 An(III) Frequency Distributions and CDF

A histogram of the frequency distribution of Bin N for all An(III) comparisons appears in Figure 1 (see p. 22 below). This is the same histogram established in the original version of this analysis (Xiong et al., 2004, Figure 1, p. 14). A total of 243 measured An(III) 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 3 (pp. 43-44 below) gives values of the corresponding CDF for Bin N for the An(III) comparisons. Figure 2 (p. 23) shows the plotted CDF. This is also the same CDF established by the original analysis (Xiong et al., 2004, Table 2 and Figure 2, pp. 15 and 27-28, respectively). Figure 2 shows that the median value of the An(III) comparison corresponds to a slight overprediction of the measured solubilities. Therefore, the An(III) thermodynamic speciation and solubility model implemented in the speciation and solubility code FMT slightly overpredicted the measured An(III) solubilities. Thus, we did not revise the An(III) comparison for the PABC, and we will use the uncertainty range and probability distribution in Table 3 in this report for the PABC. Brush et al. (2005) provided details on the PA implementation of this range and distribution.

5.2.3 An(IV) Frequency Distributions and CDF

The original histogram of the frequency distribution of Bin N for all An(IV) comparisons was provided by Xiong et al. (2004, Figure 3, p. 16); the original versions of the corresponding CDF for Bin N for An(IV) were tabulated and shown by Xiong et al. (2004, Table 3 and Figure 4, pp. 29-30 and 17, respectively). A total of 159 measured and predicted solubilities were compared in the original version of this analysis.

Because the An(IV) model in FMT significantly underpredicted the measured An(IV) solubilities (Xiong et al., 2004, Figures 3 and 4 and Table 3, pp. 16, 17, and 29-30, respectively), we: (1) identified the cause of these underpredictions, (2) corrected this problem, and (3) revised the comparison of measured and predicted An(IV) solubilities for the PABC. Brush and Xiong (2005, Subsection. 7.2) indicated that the cause of the underpredictions of An(IV) solubilities identified by Xiong et al. (2004, Subsection 3.2.3) resided in the FMT thermodynamic database used for the original version of this analysis, FMT_040628.CHEMDAT. In particular, Brush and Xiong (2005) identified the value of μ^0/RT for $Th(OH)_4(aq)$ in FMT_040628.CHEMDAT, -622.4700, as the problem, and tentatively concluded that μ^0/RT for $Th(OH)_4(aq)$ should be changed from -622.4700 to -626.8467.

However, Nowak (2005) recommended revising this parameter to -626.5853. This avoided having to make changes in the parameters for other Th(IV) species in the FMT database. (The value tentatively recommended by Brush and Xiong (2005) was based on the formation constant for $Th(OH)_4(aq)$ in another database, in which the values of the parameters for several other Th(IV) species are slightly different from those in the FMT database.) Nowak (2005) provided detailed explanations of the sources of both the original and the corrected values of μ^0/RT for $Th(OH)_4(aq)$ (-622.4700 and -626.5853, respectively). Xiong (2005) implemented Nowak's (2005) recommendation and released the corrected version of the database, FMT_050405.CHEMDAT. We used FMT_050405.CHEMDAT to revise Xiong et al.'s (2004) comparison of measured and predicted An(IV) solubilities. However, we had to exclude much of the measured solubility data used in the original comparison. Thus, the number of measured and predicted solubilities compared decreased from 159 in the original analysis to 45 in this revised analysis. The discussion below explains our reasons for excluding these data.

Figures 3, 4, and 5 (see pp. 24, 25, and 26 below) compare Th(IV) solubilities measured in 0.6, 1.2, and 3.0 M NaCl, respectively, by Felmy et al. (1991) to solubilities predicted by FMT using the uncorrected database (FMT_040628.CHEMDAT) with the uncorrected value of μ^0/RT for $Th(OH)_4(aq)$ (-622.4700). First, these plots demonstrate that the An(IV) model in FMT significantly underpredicted Th(IV) solubilities from a pH of ~5.6 to about ~10.2, the pH range in which $Th(OH)_4(aq)$ is the dominant dissolved Th(IV) species. Note that the deviation between the measured and predicted solubilities in this pH range appears to be greater for 3.0 M NaCl than for 0.6 or 1.2 M NaCl. Second, these plots demonstrate that FMT both overpredicted and underpredicted Th(IV) solubilities from a pH of about 3.6 to about 5.6, wherein Th^{4+} is the dominant dissolved Th(IV) species. Third, these plots show that, for pH $\leq$ ~3.6, FMT significantly overpredicted Th(IV) solubilites. Fourth, both the measured and predicted solubilities are independent of pH for pH $\leq$ ~3.6. This behavior is expected for ~5.6 $\leq$ pH = ~10.2, the range in which $Th(OH)_4(aq)$ is dominant. However, there is no reason based on aqueous Th(IV) speciation (or the speciation of any other dissolved metal known to us) to expect such behavior at pH $\leq$ ~3.6. Fortunately, this behavior can be explained by noting that in both the experiments at pH $\leq$ ~3.6 and in the FMT runs at pH $\leq$ ~3.4 (0.6 M NaCl) or pH $\leq$ ~3.6 (1.2 and 3.0 M NaCl), the Th(IV)-bearing solids were consumed before saturation was attained (see Felmy et al., 1991, p. 298). Therefore, the constant Th(IV) concentration of slightly less than 10^{-2} M in the experiments reflects the fact that the constant quantity of Th(IV) added at the start of these experiments was insufficient to saturate these solutions under these conditions, and the constant Th(IV) concentration of slightly less than 10^0 M predicted by FMT resulted from failure to add enough Th(IV) in the input files to achieve saturation in these simulations. Thus, the concentrations in the experiments at pH $\leq$ ~3.6, and in the FMT runs at pH $\leq$ ~3.4 (0.6 M NaCl) or pH $\leq$ ~3.6 (1.2 and 3.0 M NaCl) are not solubilities. Because the concentrations measured by Felmy et al. (1991) at pH $\leq$ ~3.6 are not solubilities, we excluded them from the revised An(IV) comparison.

Figures 6, 7, 8, and 9 (pp. 27, 28, 29, and 30 below) compare Th(IV) solubilities measured in 0.6, 1.2, and 3.0 M NaCl; and in 0.6 M KCl, respectively, by Felmy et al. (1991) to solubilities predicted by FMT using the corrected database (FMT_050405.CHEMDAT) with the new value of μ^0/RT for $Th(OH)_4(aq)$ (-626.5853). First and foremost, Figure 8 demonstrates that use of the corrected database results in good agreement between the measured and predicted

solubilities for ~5.6 ≤ pH = ~10.2 (i.e., the new FMT predictions plot close to the middle of the range of concentrations of the measured solubilities). (Note that this pH range includes the pH expected in the WIPP, ~9). However, for ~3.6 ≤ pH ≤ ~5.6, FMT still underpredicts An(IV) solubilities somewhat. Second, Figures, 6, 7, and 9 show that FMT significantly overpredicts Th(IV) solubilities in 0.6 and 1.2 M NaCl, and in 0.6 M KCl, respectively. Overprediction of Th(IV) solubilities in these intermediate-ionic-strength solutions is probably the result of carrying out too few experiments at these (and lower) ionic strengths when the Pitzer model was being parameterized for dissolved An species in high-ionic-strength solutions prior to the CCA. The objective of the WIPP Actinide Source Term Program then - as now - has been to develop thermodynamic models capable of predicting An(III), An(IV), and An(V) speciation and solubilities in high-ionic-strength WIPP brines.

Based on the results shown in Figures 6, 7, and 9 (caused by inadequate parameterization of the Pitzer model at low and intermediate ionic strengths), we excluded the solubilities measured using solutions with ionic strengths <3 M. This - along with the exclusion of data from Felmy et al. (1991) for pH ≤ 3.6 (see above) - decreased the number of An(IV) comparisons from 159 to 45. Specifically, the 114 measured solubilities excluded from the revised An(IV) comparison were: (1) all of the results of Baston et al. (1996), which were carried out in a standard NIREX solution with an ionic strength (I) of 0.02 M; (2) the results of Felmy et al. (1991) that were carried out in 0.6 or 1.2 M NaCl, or 0.6 M KCl (I = 0.6 or 1.2 M); (3) all of the results of Neck et al. (2002), which were conducted in 0.5 M NaCl (I = 0.5 M); and (4) all of the results of Osthlos et al. (1994), which were performed in 0.5 M $NaClO_4$ (I = 0.5 M. The revised version of Table 2 (see pp. 39-42 below) reflects these deletions. The 45 measured solubilities retained in this revised analysis are all from the experiments of Felmy et al. (1991) in 3.0 M NaCl (I = 3.0 M) and ~3.6 ≤ pH ≤ ~5.6. Neither of these exclusion criteria was applied to the measured solubilities in the An(III) and An(V) comparisons.

Exclusion of 114 solubilities measured in intermediate-ionic-strength solutions from the new An(IV) comparison is reasonable, given the intended use of FMT for predicting An(IV) solubilities in high-ionic-strength WIPP brines. Inclusion of these measured solubilities would probably result in unwarranted reductions of the solubilites predicted by FMT for high-ionic-strength brines in WIPP disposal rooms during sampling for use in PA. (The uncertainty range and probability distribution obtained from this comparison will correct for over- or underpredictions of solubilities by decreasing or increasing, respectively, the FMT predictions prior to use in PA; Brush et al. (2005) gave details of the PA implementation of the range and distribution.)

On the other hand, the range and distribution obtained from the revised An(IV) comparison would be inappropriate for solubilities predicted for low- and intermediate-strength Culebra ground waters. If FMT were used to predict An(IV) solubilites in such Culebra fluids, another comparison including solubilities measured in low- and/or intermediate-ionic-strength solutions would probably be required.

The fluids that could enter postclosure WIPP disposal rooms would have ionic strengths in excess of 3.0 M. However, we know of no An(IV) solubility experiments carried out in solutions with ionic strengths greater than that of the 3.0 M NaCl used by Felmy et al. (1991).

Therefore, we cannot extend the An(IV) comparison all the way up to the ionic strengths of the brines that could enter the repository.

The revised histogram of the frequency distribution of Bin N for all An(IV) comparisons appears in Figure 10 (p. 31). A total of 45 measured and predicted solubilities were compared in this new analysis. The distribution is relatively broad, and peaks at Bin -0.45 (N = -0.45) with a frequency of 9, and is skewed toward negative values of N.

Table 4 (p. 45) gives values of the corresponding cumulative distribution function (CDF) for Bin N for An(IV). We will use the uncertainty range and probability distribution in Table 4 for the PABC. Figure 11 (p. 32) shows the plotted CDF. The median value of the An(IV) comparison corresponds to a slight underprediction of the measured solubilities. Therefore, the corrected database (FMT_050405.CHEMDAT) with the corrected value of μ^0/RT for $Th(OH)_4(aq)$ (-626.5853) slightly underpredicted the measured solubilities. However, the extent of this underprediction is significantly less than that observed with the uncorrected database (FMT_040628.CHEMDAT) with the previous value of μ^0/RT for $Th(OH)_4(aq)$ (-622.4700).

5.2.4 An(V) Frequency Distributions and CDF

A histogram of the frequency distribution of Bin N for all An(V) comparisons is shown in Figure 12 (see p. 33 below). This is the same histogram established in the original version of this analysis (see Xiong et al., 2004, Figure 5, p. 18). 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 5 (p. 46 below) gives values of the corresponding CDF for Bin N for An(V). Figure 13 (p. 34) shows the plotted CDF. This is the same CDF established by the original analysis (Xiong et al., 2004, Table 4 and Figure 6, pp. 31 and 19, respectively). The median value of the An(V) comparison corresponds to a slight overprediction of the measured solubilities. Therefore, the An(V) model in FMT slightly overpredicted the measured An(V) solubilities slightly. However, the uncertainty range and probability distribution in Table 5 will not actually be used for the PABC because PA does not sample the solubility of Np(V), the only actinide that will speciate in the +V oxidation state. PA does not sample the solubility of Np(V) because Np is the least important of the five radioelements for which solubilities are calculated (see Brush and Garner, 2004).

5.2.5 Combined (An(III, IV, V)) Frequency Distribution and CDF

We revised the comparison of measured and predicted An(IV) solubilities carried out by Xiong et al. (2004, Subsection 3.2.3) (see Subsection 5.2.3 above). Therefore, we also had to revise the combined An(III, IV, V) comparison.

A histogram of the frequency distribution of Bin N for all combined An(III, IV, V)) comparisons is shown in Figure 14 (see p. 35 below). A total of 424 measured and predicted values were compared. The distribution is relatively broad and peaks at Bin 0 (N = 0) with a frequency of 69.

Table 6 (pp. 47-48 below) gives values of the corresponding CDF for Bin N for all An(III, IV, V) comparisons. Figure 15 (p. 36) shows the plotted CDF. The median value of the An(III, IV, V) comparison corresponds to a slight overprediction of the measured solubilities. Therefore, the corrected database (FMT_050405.CHEMDAT) with the corrected value of μ^0/RT for $Th(OH)_4(aq)$ (-626.5853) slightly overpredicted the measured An(III), An(IV), and An(V) solubilities. This is a significant improvement over the original analysis of Xiong et al. (2004), in which the uncorrected database (FMT_040628.CHEMDAT) with the previous value of μ^0/RT for $Th(OH)_4(aq)$ (-622.4700) significantly underpredicted the measured An(III), An(IV), and An(V) solubilities.

6 CONCLUSIONS

This actinide-solubility uncertainty analysis is Rev 1 of the first uncertainty analysis carried out to compare measured solubilities and predictions made with FMT (Xiong et al, 2004) since that of Bynum (1996a, 1996b, 1996c).

Rev. 1 of this 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, 2005). This analysis included 243 An(III) comparisons, 45 An(IV) comparisons, and 136 An(V) comparisons, for a total of 424 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 slightly overpredicted the measured An(III) solubilities, (2) the An(IV) model in FMT slightly underpredicted the measured An(IV) solubilities, (3) the An(V) model in FMT slightly overpredicted the measured An(V) solubilities, and (4) overall, the An(III), An(IV), and An(V) models in FMT together slightly overpredicted the measured An(III), An(IV), and An(V) solubilities.

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8 FIGURES

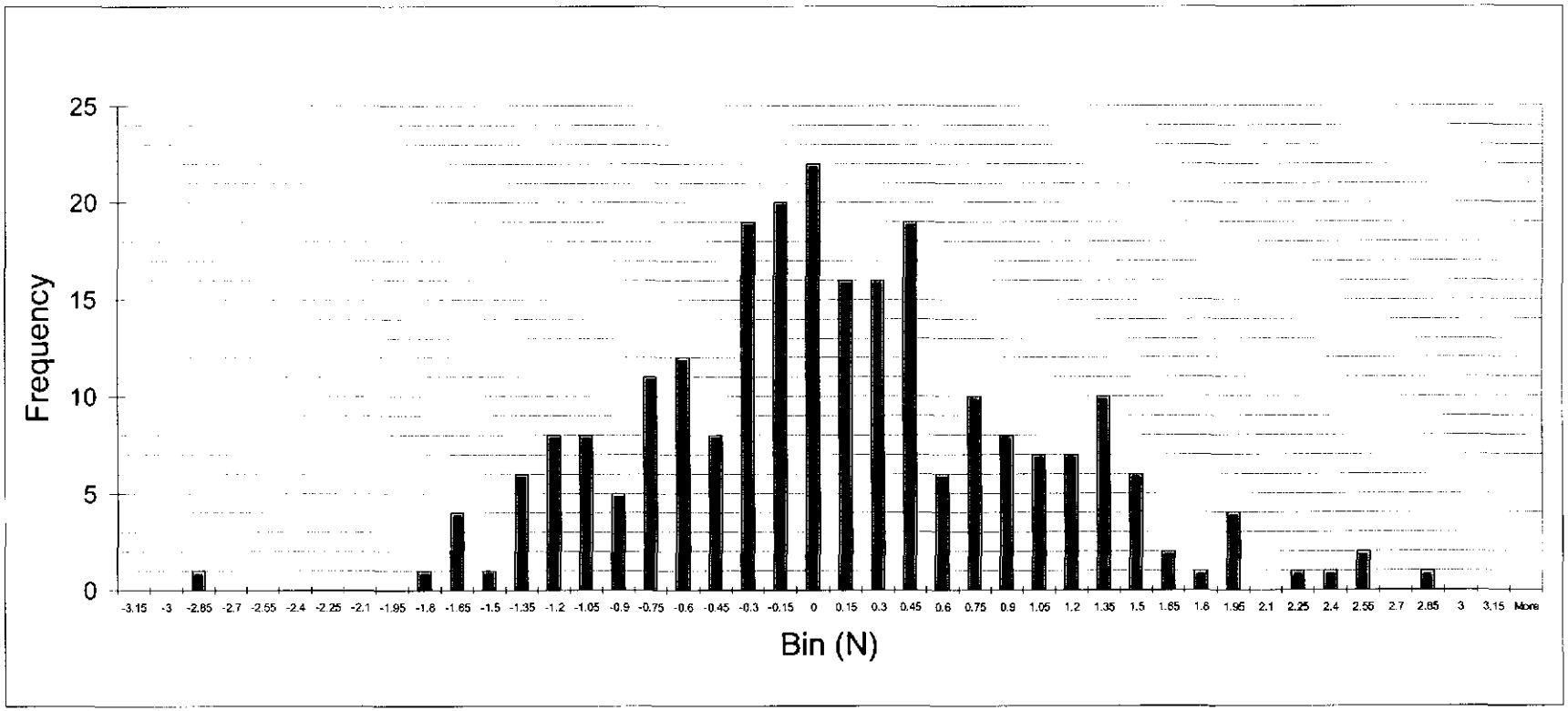

Figure 1. Histogram of the frequency distribution of Bin N for all An(III) comparisons. A total of 243 measured and predicted solubilities were compared.

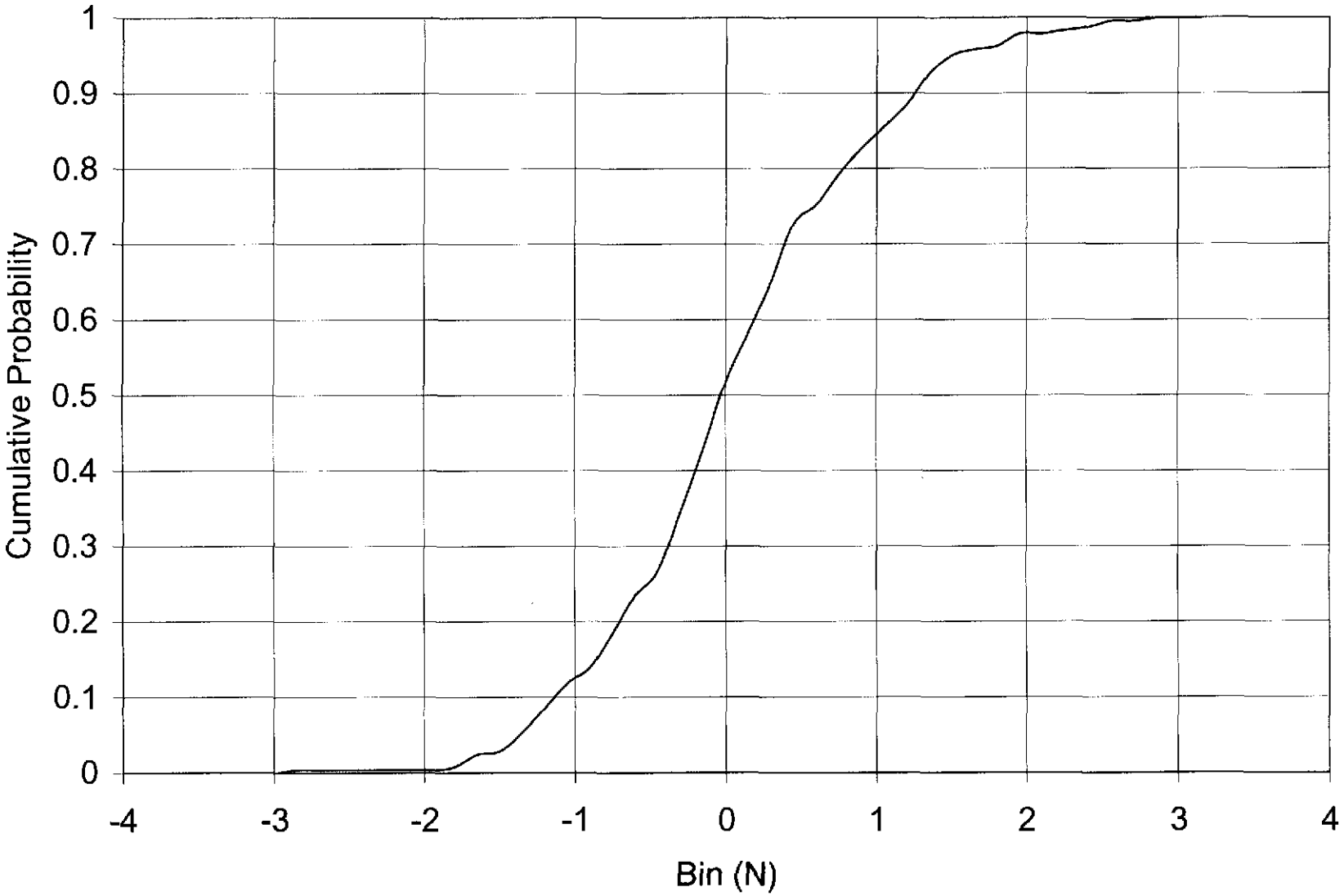

Figure 2. Plot of the CDF for Bin N for An(III).

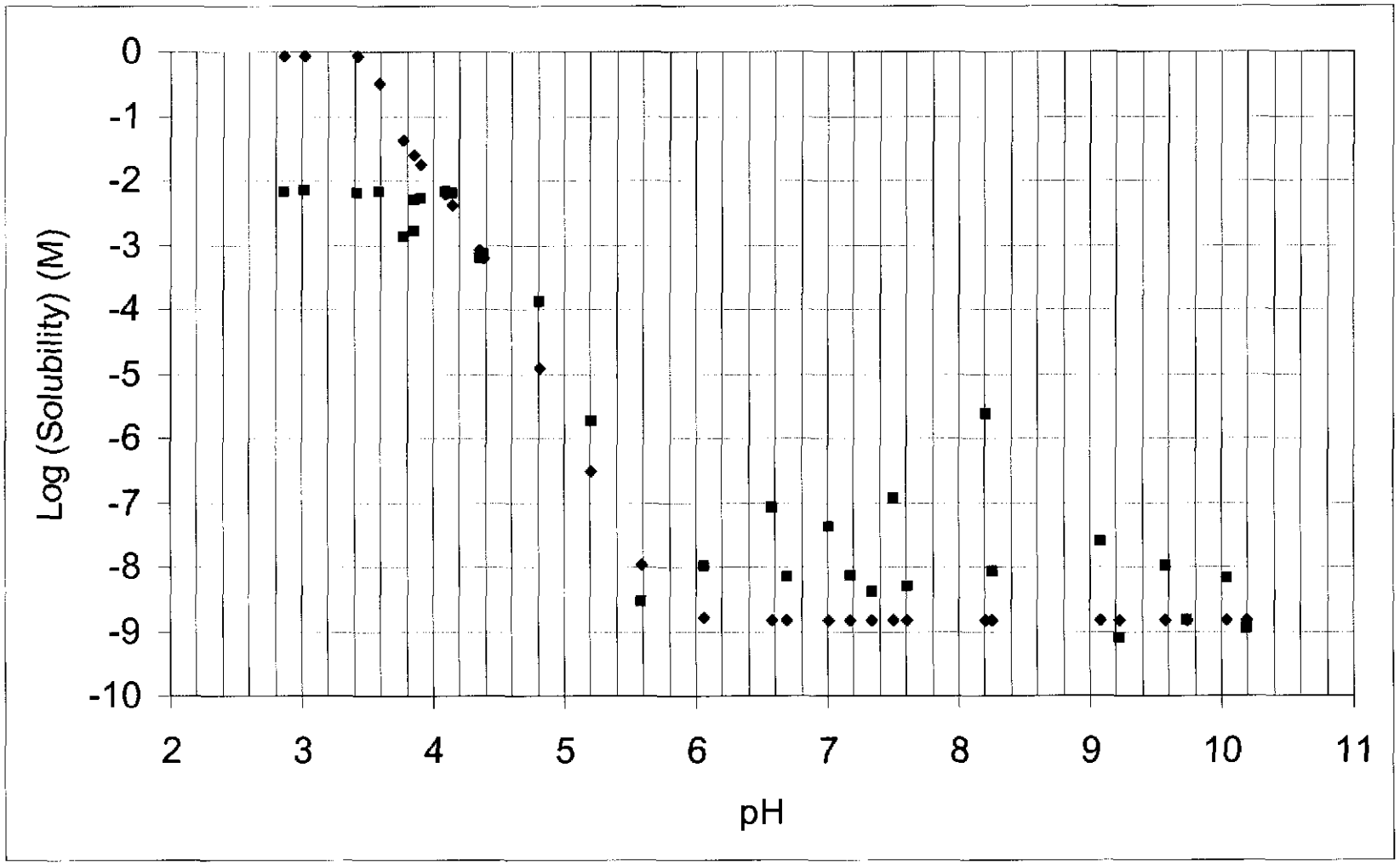

Figure 3. Comparison of Th(IV) solubilities measured in 0.6 M NaCl by Felmy et al. (1991) to solubilities predicted by FMT using the original database with the problematic value of μ^0/RT for $Th(OH)_4(aq)$ (see text for details). Red squares depict measured solubilities; blue diamonds show predicted.

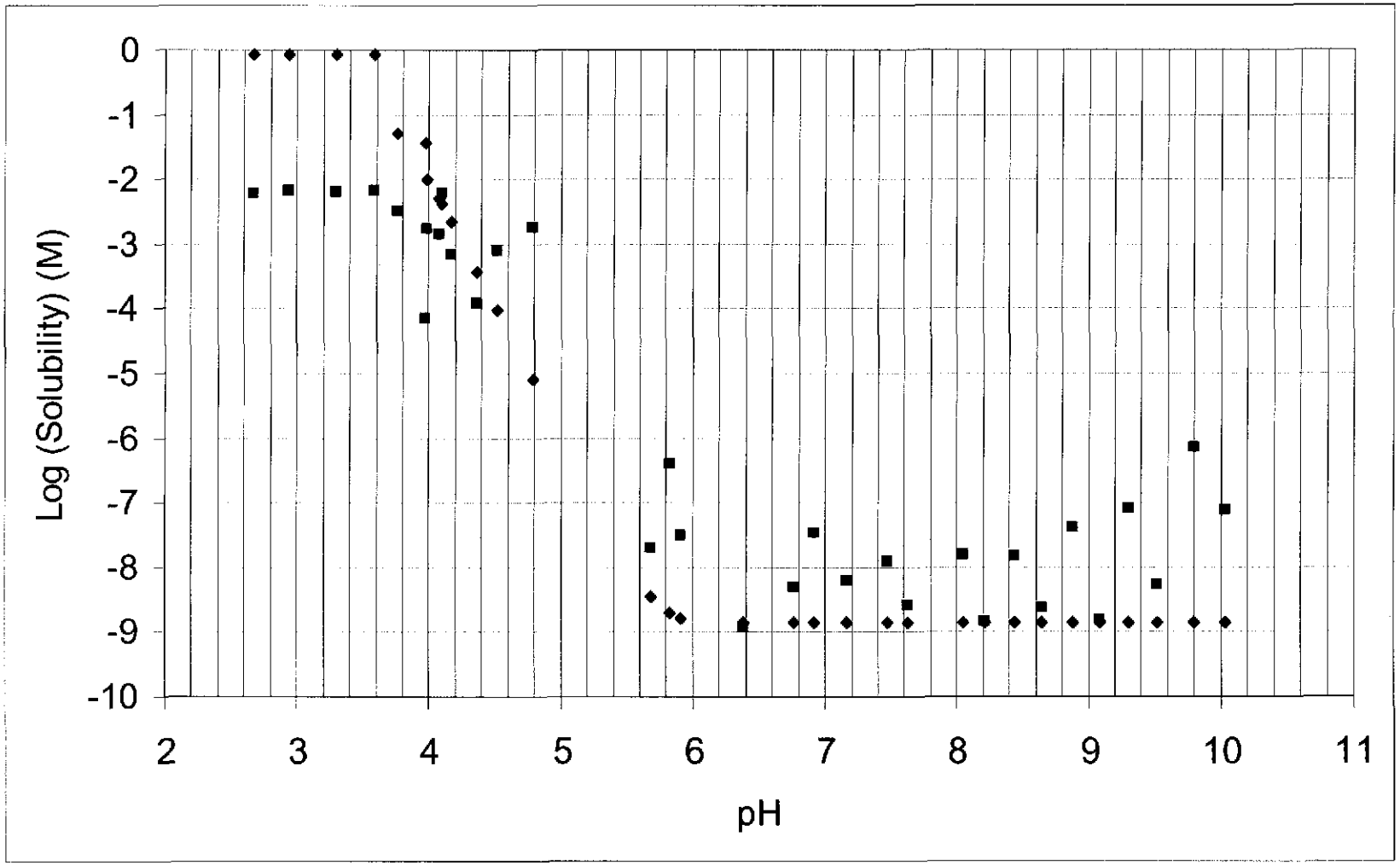

Figure 4. Comparison of Th(IV) solubilities measured in 1.2 M NaCl by Felmy et al. (1991) to solubilities predicted by FMT using the original database with the problematic value of μ^0/RT for $Th(OH)_4(aq)$ (see text for details). Red squares depict measured solubilities; blue diamonds show predicted.

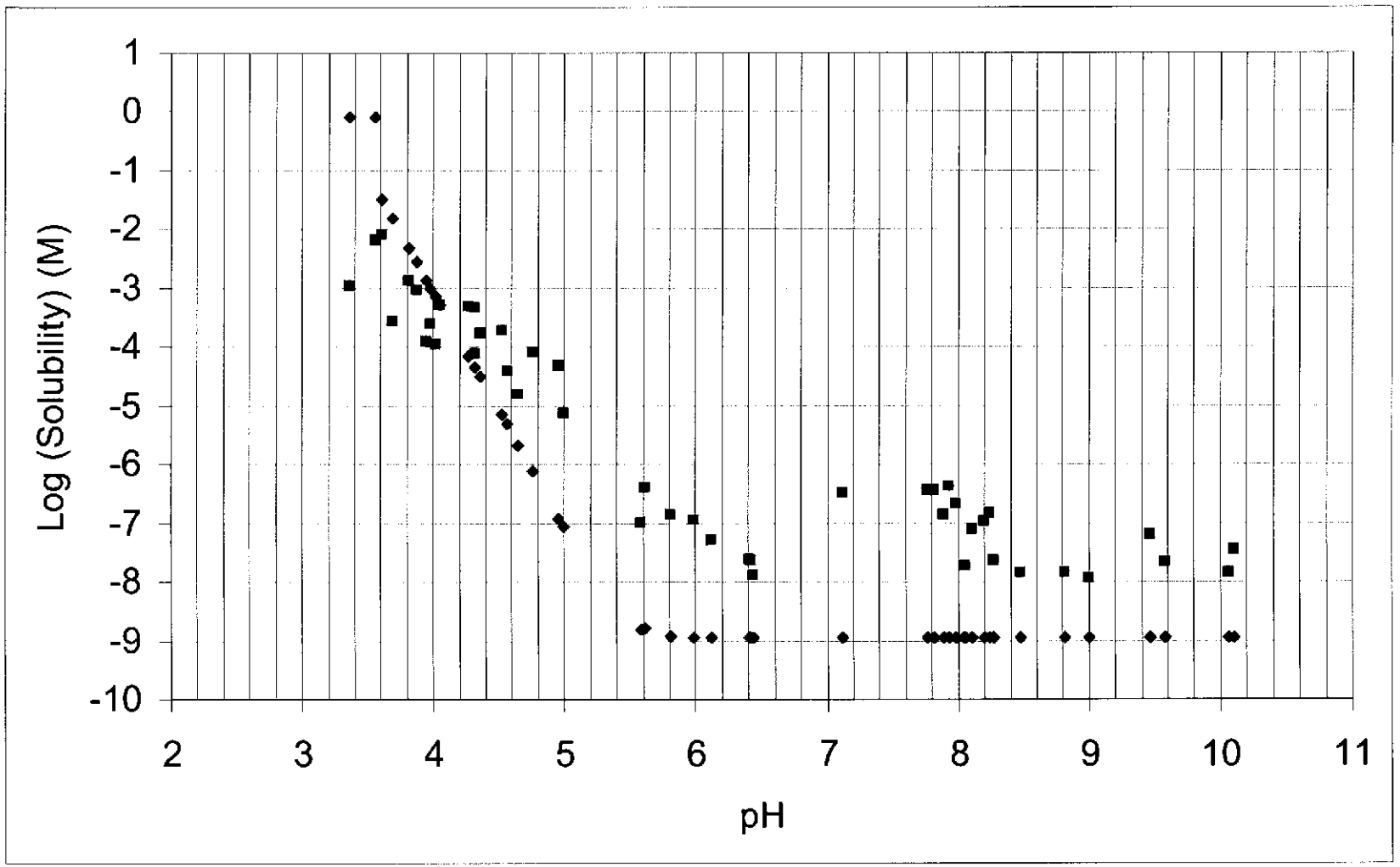

Figure 5. Comparison of Th(IV) solubilities measured in 3.0 M NaCl by Felmy et al. (1991) to solubilities predicted by FMT using the original database with the problematic value of μ^0/RT for $Th(OH)_4(aq)$ (see text for details). Red squares depict measured solubilities; blue diamonds show predicted.

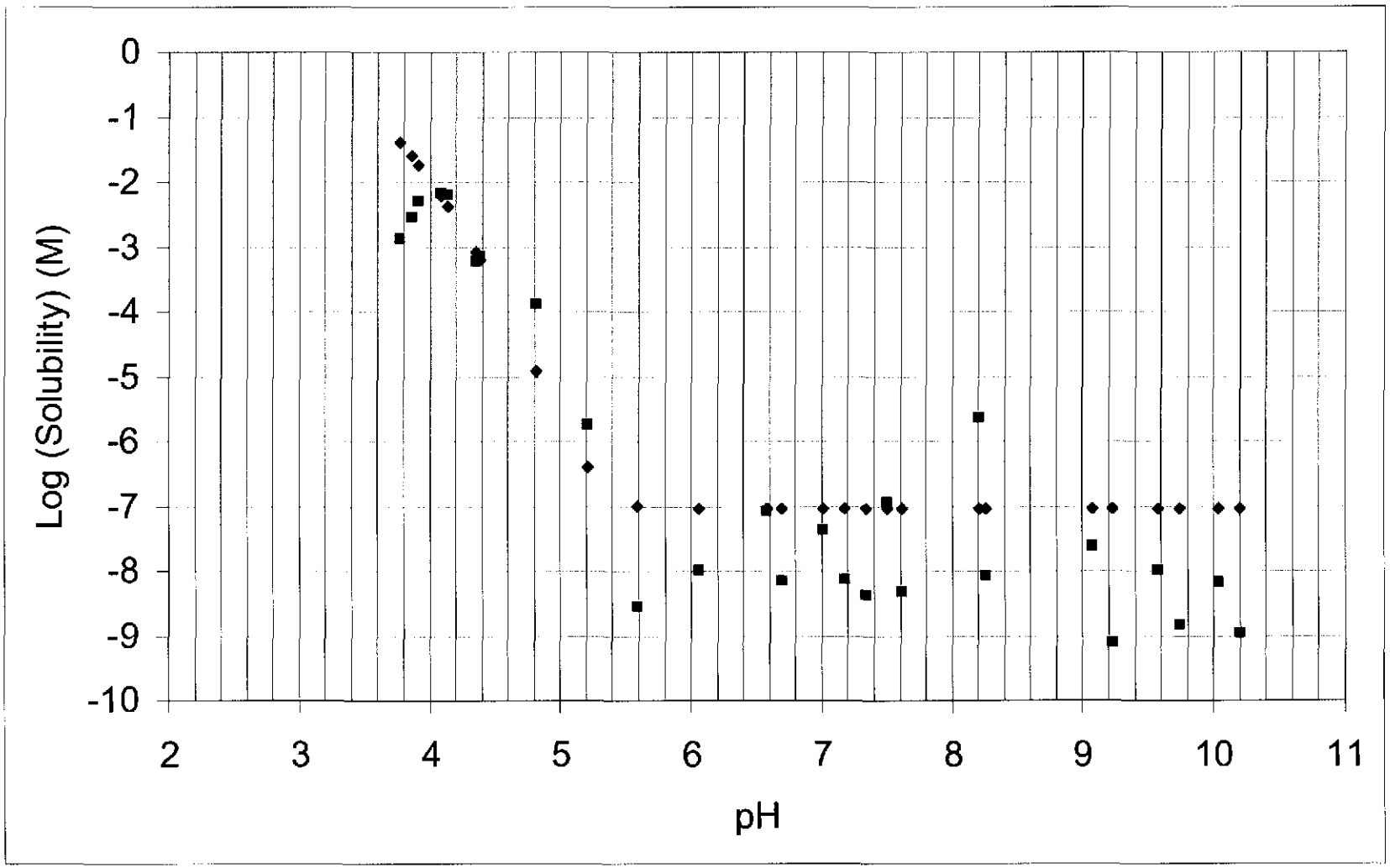

Figure 6. Comparison of Th(IV) solubilities measured in 0.6 M NaCl by Felmy et al. (1991) to solubilities predicted by FMT using the corrected database with the new value of μ^0/RT for $Th(OH)_4(aq)$ (see text for details). Red squares depict measured solubilities; blue diamonds show predicted.

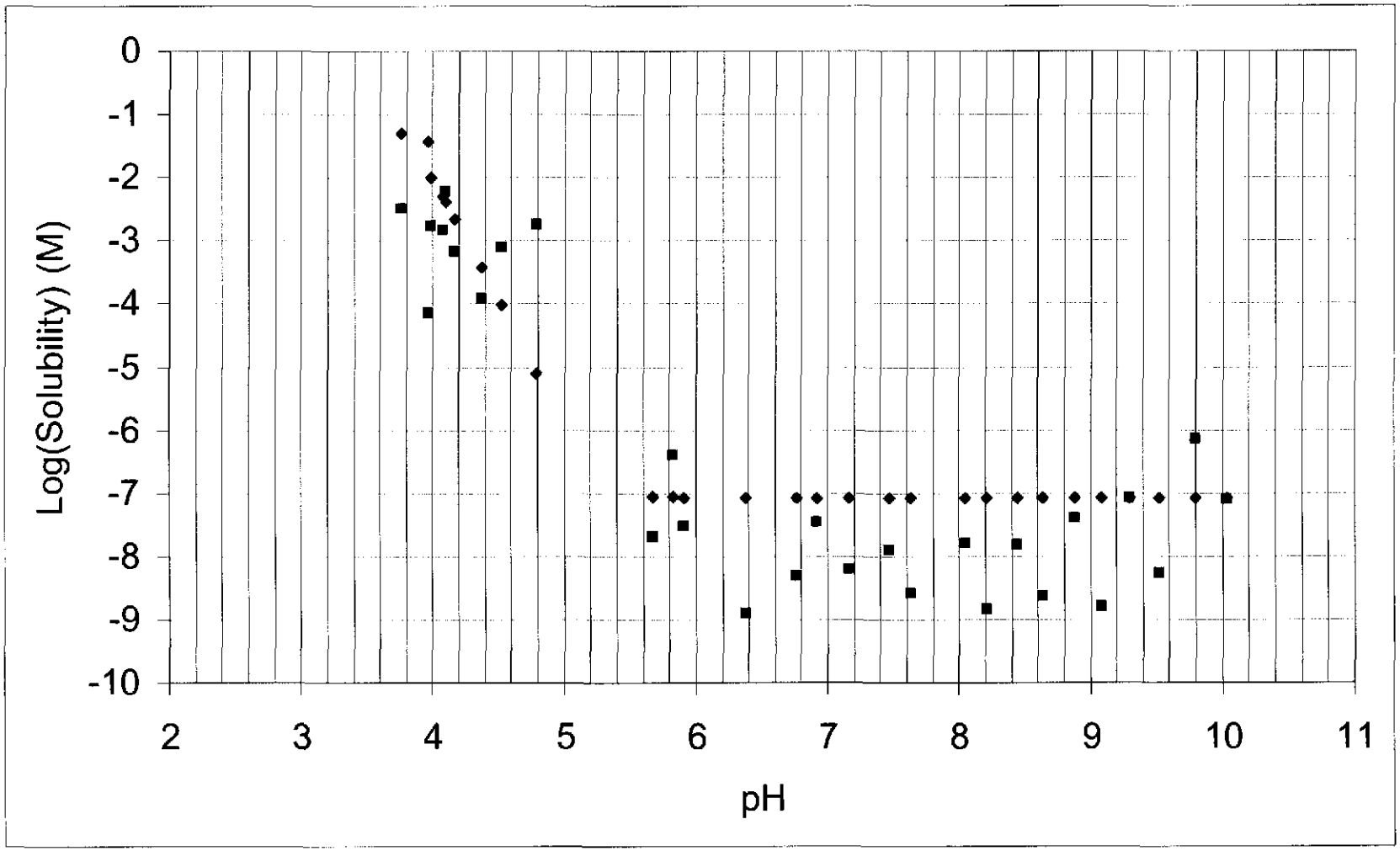

Figure 7. Comparison of Th(IV) solubilities measured in 1.2 M NaCl by Felmy et al. (1991) to solubilities predicted by FMT using the corrected database with the new value of μ^0/RT for $Th(OH)_4(aq)$ (see text for details). Red squares depict measured solubilities; blue diamonds show predicted.

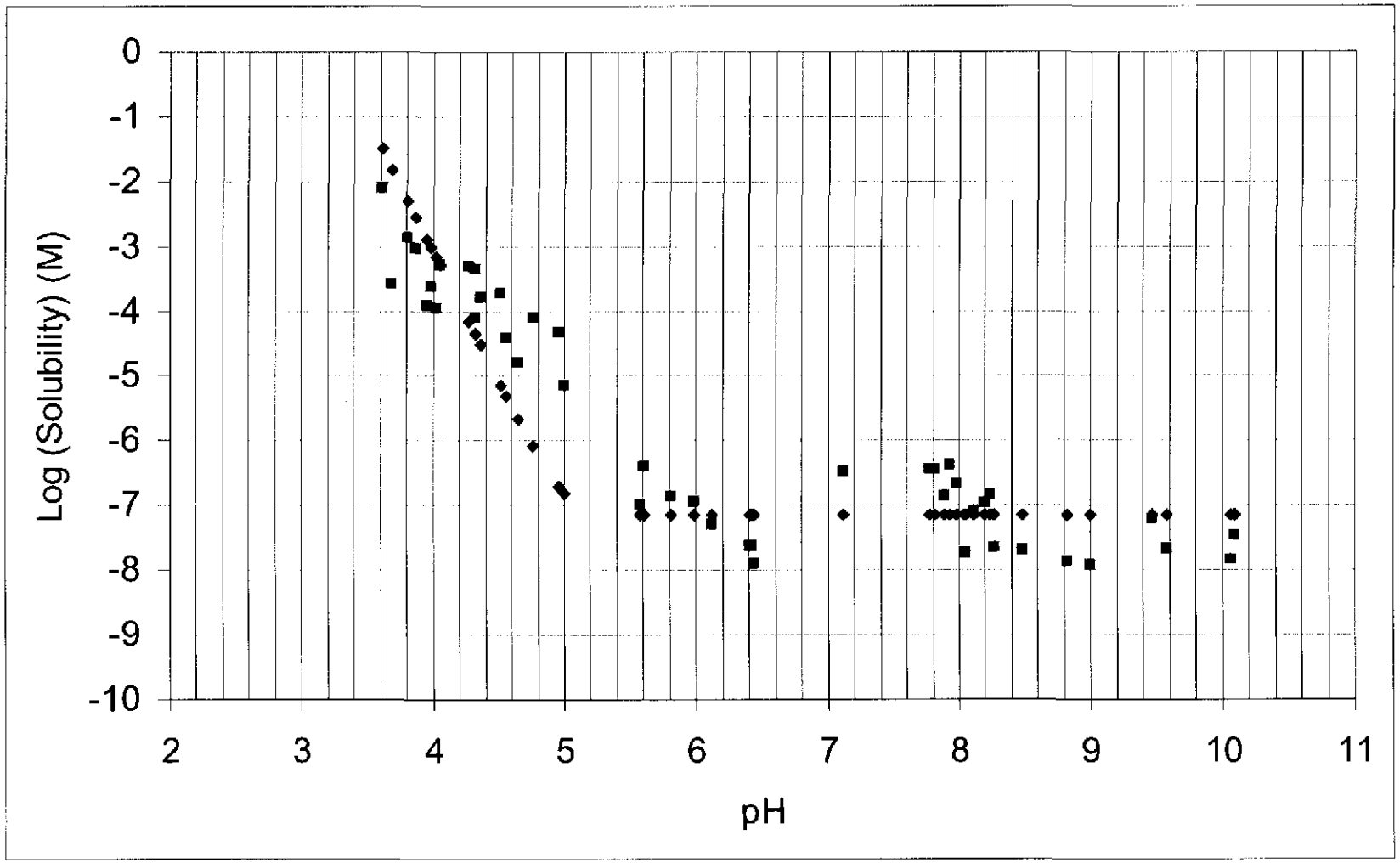

Figure 8. Comparison of Th(IV) solubilities measured in 3.0 M NaCl by Felmy et al. (1991) to solubilities predicted by FMT using the corrected database with the new value of μ^0/RT for $Th(OH)_4(aq)$ (see text for details). Red squares depict measured solubilities; blue diamonds show predicted.

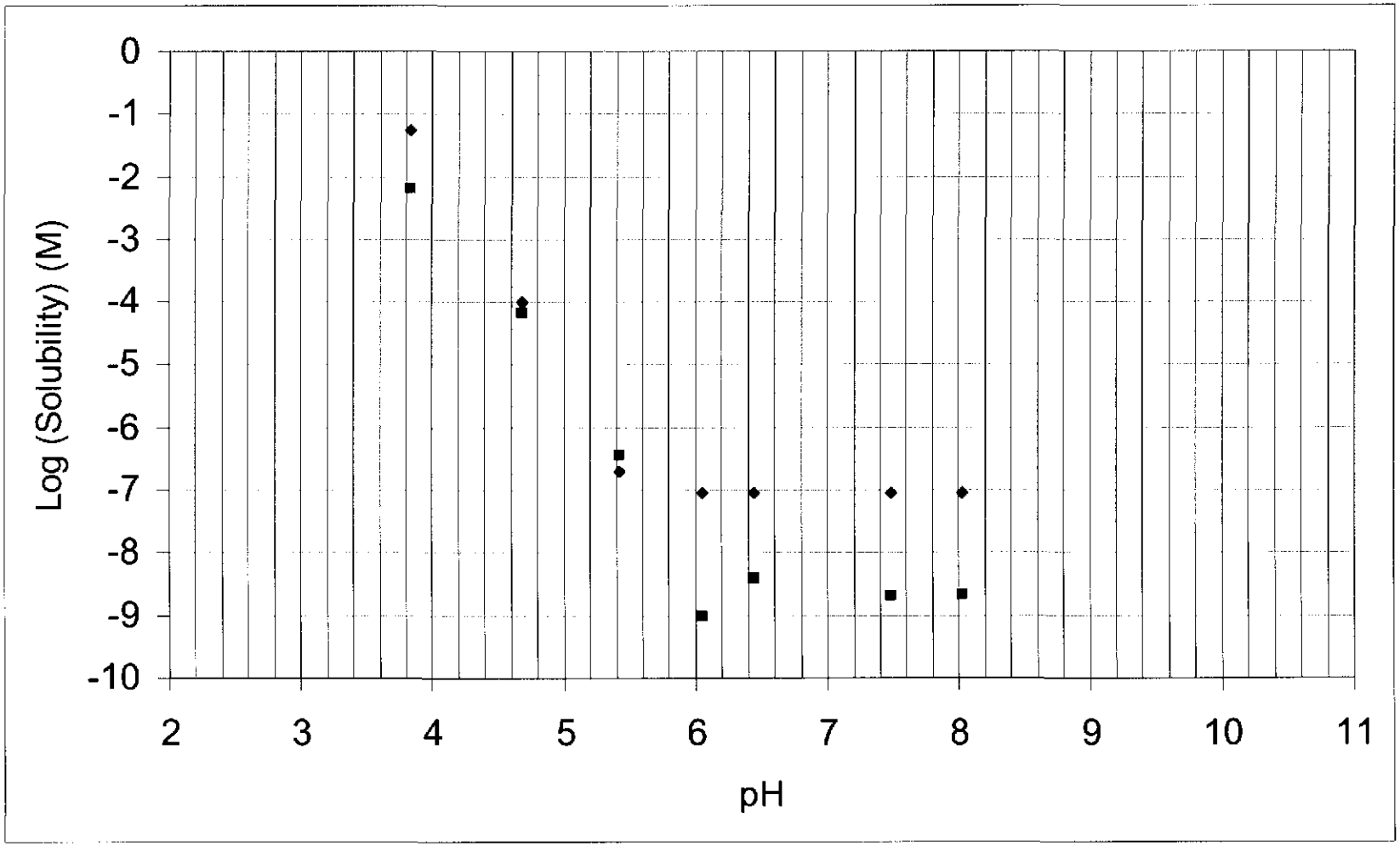

Figure 9. Comparison of Th(IV) solubilities measured in 0.6 M KCl by Felmy et al. (1991) to solubilities predicted by FMT using the corrected database with the new value of μ^0/RT for $Th(OH)_4(aq)$ (see text for details). Red squares depict measured solubilities; blue diamonds show predicted.

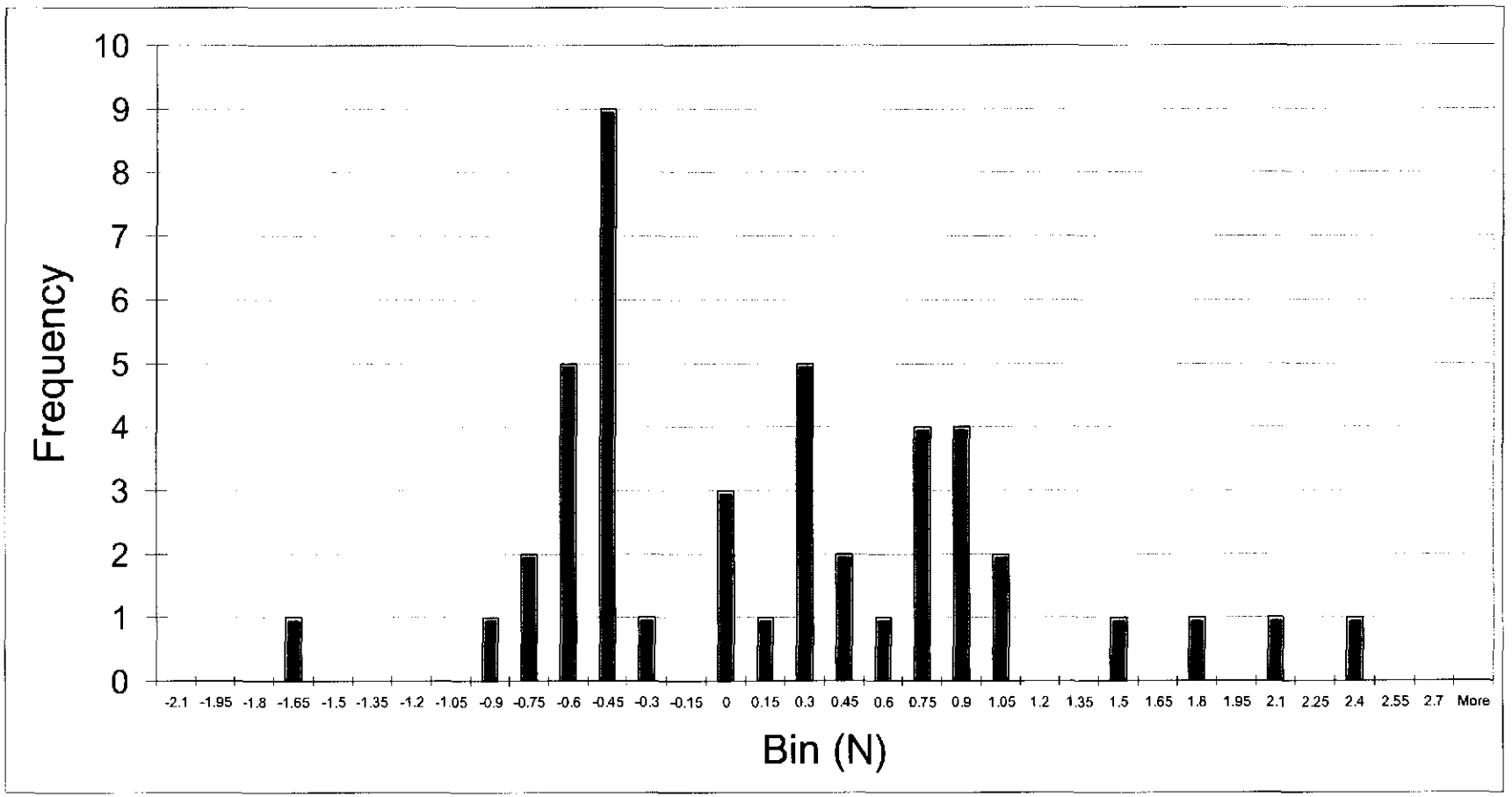

Figure 10. Histogram of the frequency distribution of Bin N for all An(IV) comparisons. A total of 45 measured and predicted solubilities were compared.

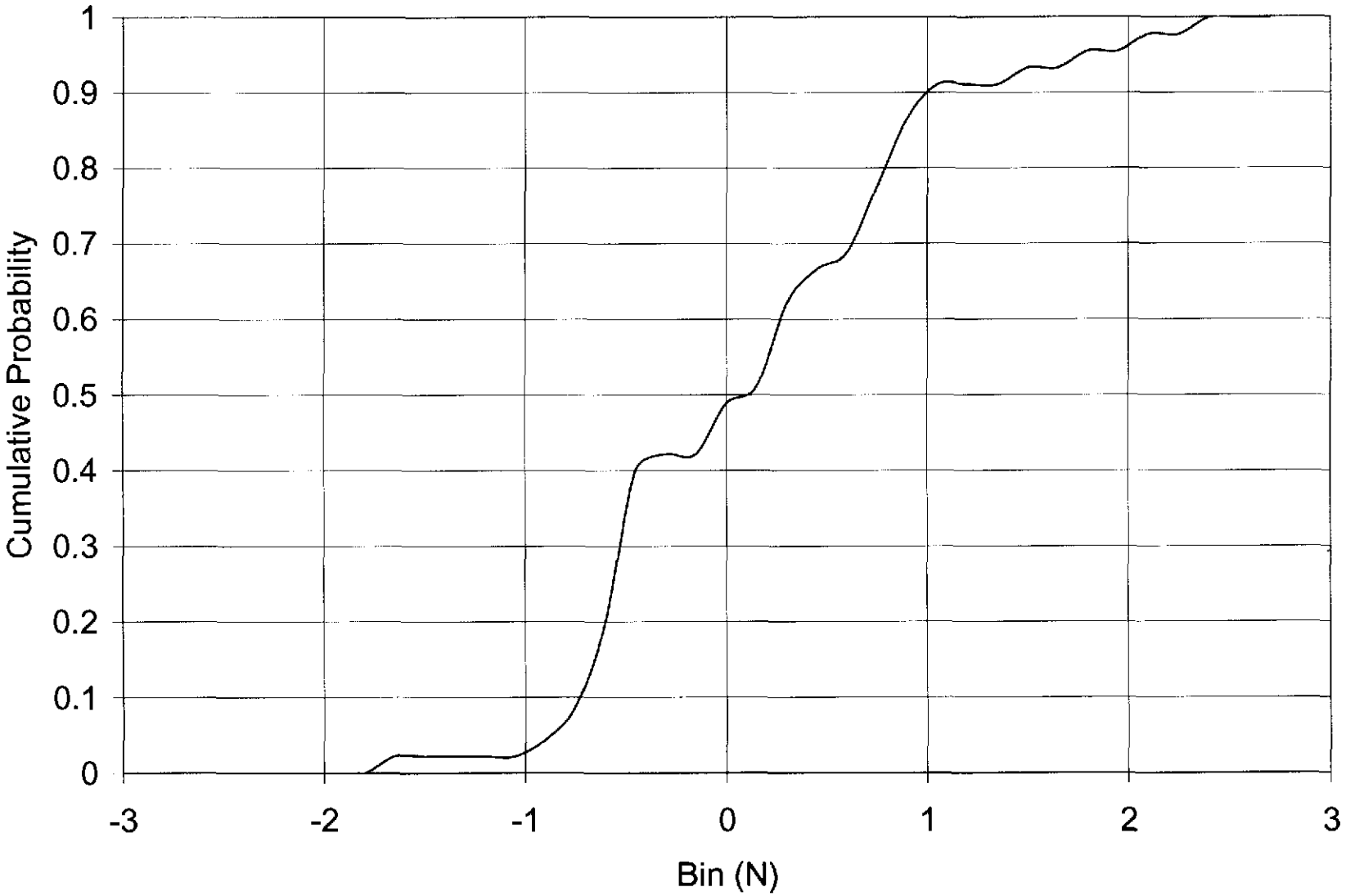

Figure 11. Plot of the CDF for Bin N for An(IV).

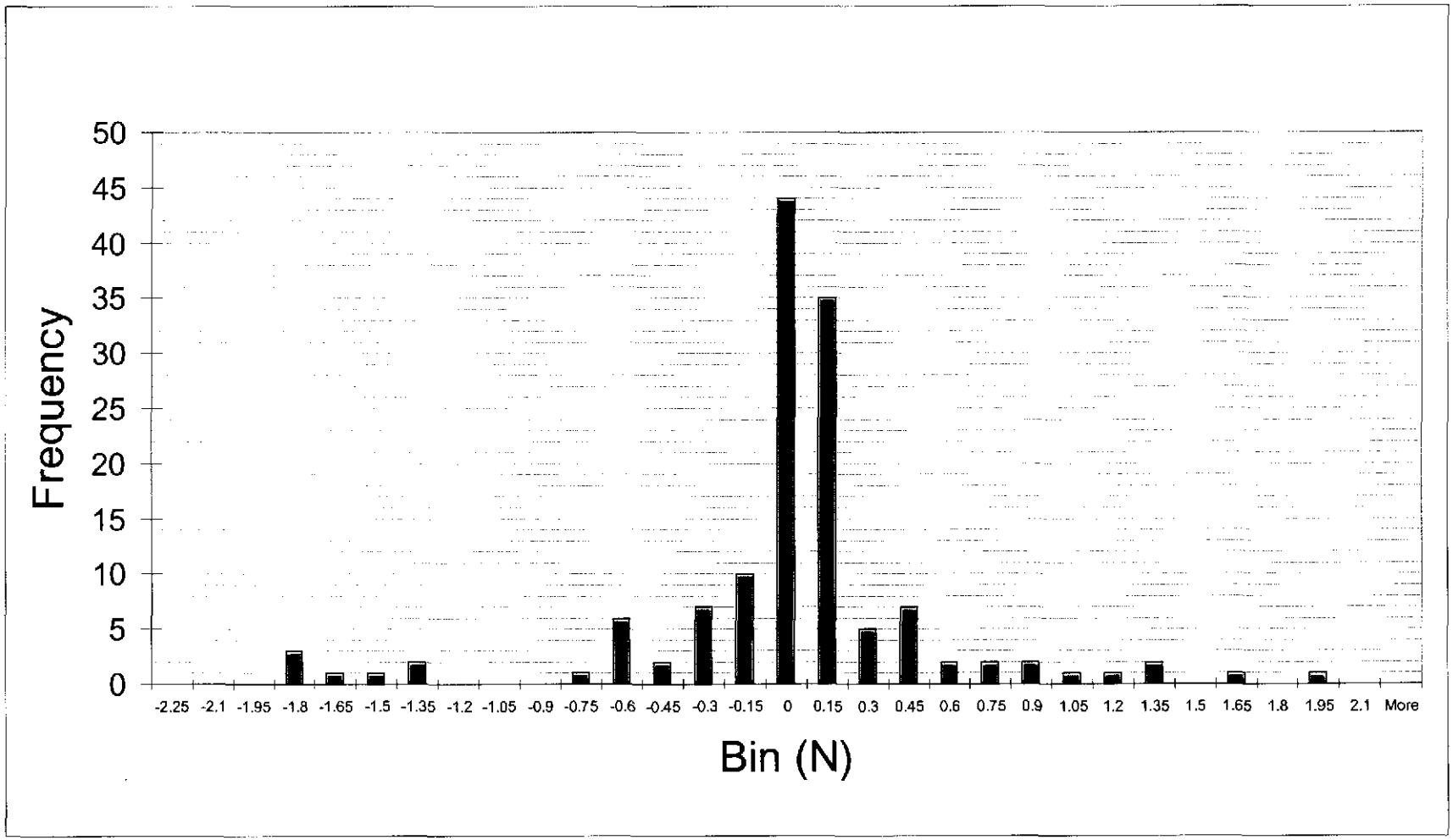

Figure 12. Histogram of the frequency distribution of Bin N for all An(V) comparisons. A total of 136 measured and predicted solubilities were compared.

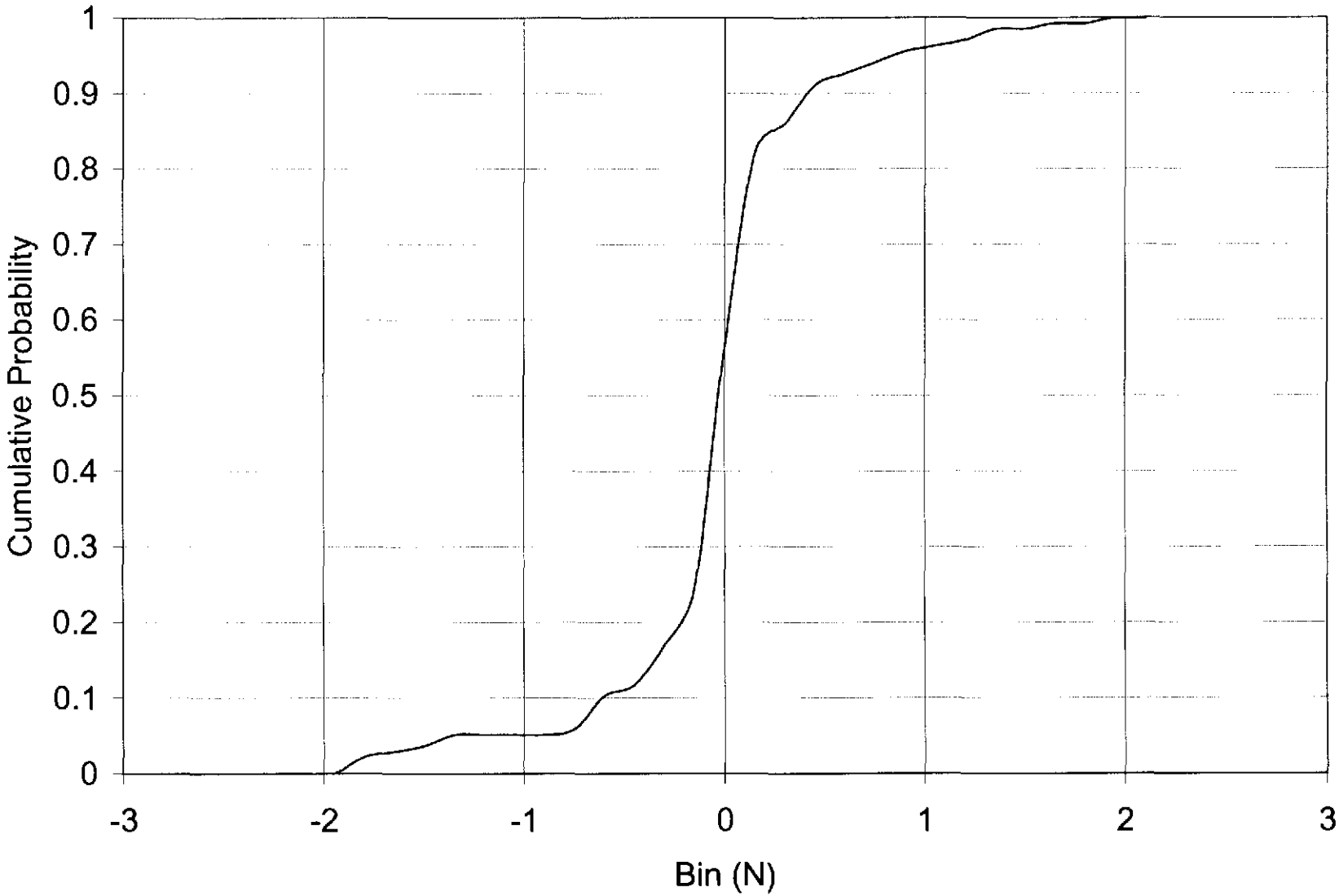

Figure 13. Plot of the CDF for Bin N for An(V).

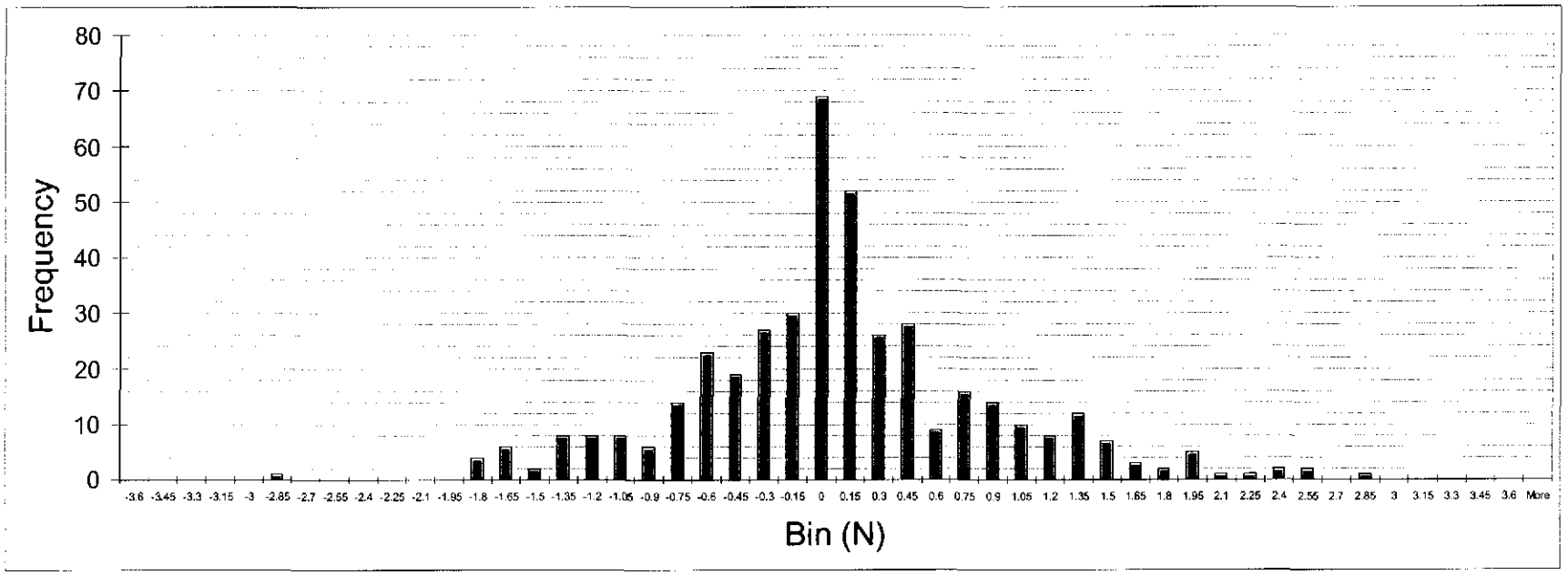

Figure 14. Histogram of the frequency distribution of Bin N for all combined (An(III, IV, V)) comparisons. A total of 424 measured and predicted solubilities were compared.

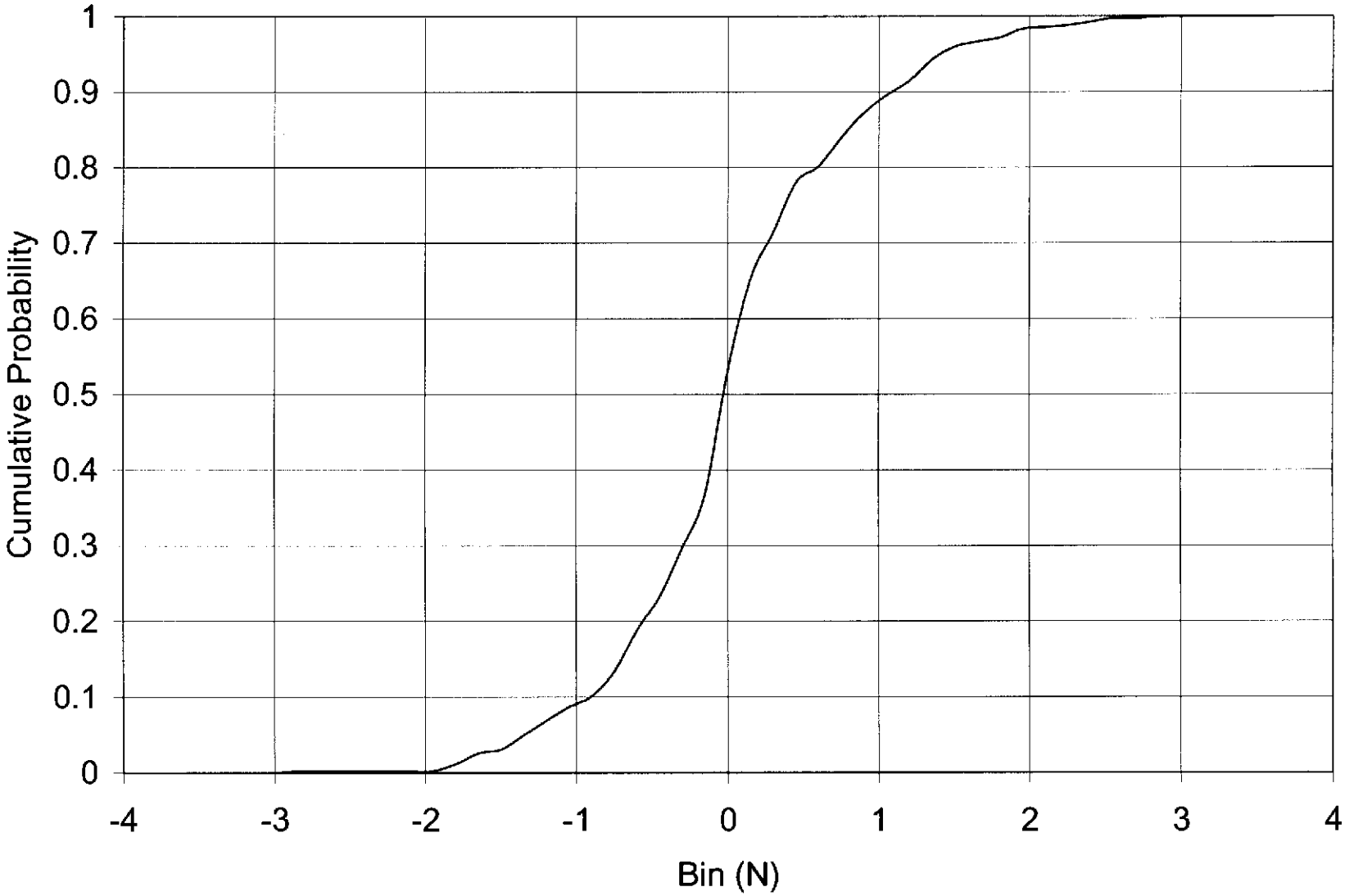

Figure 15. Plot of the CDF for Bin N for all three oxidation states (An(III, IV, V)).

9 TABLES

Table 1. Abbreviations, Acronyms, and Initialisms.

Abbreviation, Acronym, or Initialism	Definition
Ac	acetate ($CH_3CO_2^+$)
am	amorphous
An	actinide element
An(III)	actinide element(s) in the +III oxidation state
An(IV)	actinide(s) in the +IV oxidation state
An(V)	actinide(s) in the +V oxidation state
An(VI)	actinide(s) in the +VI oxidation state
AP	analysis plan
aq	aqueous (dissolved in an aqueous solution)
Brine A	a synthetic brine representative of intergranular Salado-Formation brines
CCA	(WIPP) Compliance Certification Application
CDF	cumulative distribution function
CMS	(Sandia/WIPP software) Configuration Management System
CRA-2004	(WIPP) Compliance Recertification Application, submitted to the EPA in March 2004
CRA-2004	the first (WIPP) Compliance Recertification Application
D	the difference(s) between logarithms (base 10) of measured and predicted actinide solubilites (S_m and S_p)
DOE	(U.S.) Department of Energy
EPA	(U.S.) Environmental Protection Agency
ERDA-6	Energy Research and Development Administration (WIPP Well) 6, a synthetic brine representative of fluids in Castile brine reservoirs
Fm.	Formation
FMT	Fracture-Matrix Transport, a geochemical speciation and solubility code
G-Seep	a naturally-occurring brine formerly collected from G Drift in the WIPP underground workings
H, H^+	hydrogen, hydrogen ion
KCl	potassium chloride
LIB	library
M	molar

Table 1. Abbreviations, Acronyms, and Initialisms.

Abbreviation, Acronym, or Initialism	Definition
N	bin number (see Subsection 5.2 for explanation)
NaCl	sodium chloride
NONLIN	a code used to fit parameters used in the FMT database to solubility, stability-constant, or other experimental data
Np	neptunium
O	oxygen
OH	hydroxide
PA	performance assessment
PABC	(WIPP) Performance Assessment Baseline Calculations, to be carried out in 2005
PAVT	(WIPP) Performance Assessment Verification Test, conducted in 1997
pH	negative logarithm (base 10) of the activity of H^+
R	gas constant
Rev.	Revision
S_m	measured solubility (Subsection 5.2)
S_p	predicted solubility (Subsection 5.2)
SOTERM	(Actinide) Source Term, an appendix in the CCA and an attachment to Appendix PA in the CRA-2004
SPC	Salado Primary Constituents, a synthetic brine representative of intergranular Salado brines
T	temperature
Th, Th(IV), ThO_2, $Th(OH)_4(aq)$	thorium, thorium in the +IV oxidation state, thorium dioxide, aqueous thorium hydroxide (a dissolved Th((IV) species)
WIPP	(U.S. DOE) Waste Isolation Pilot Plant
μ^0	standard chemical potential
μ^0/RT	dimensionless standard chemical potential

Table 2. 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 2. 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 (pcH)	$NaNd(CO_3)_2 \cdot 6H_2O$	*Rao et al. (1999)	Measured from published plots. Bynum (1996a, b) used 21 measured points.
+III	G-Seep	7.3	5.81-7.77 (pcH)	$NaNd(CO_3)_2 \cdot 6H_2O$	*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.

Table 2. Sources of Measured Actinide Solubilities (cont.).

Oxidation State	Solution	Ionic Strength, (M)	pH	Solubility-Controlling Solid	Reference, Type of Data Source	Remarks
+IV	3.0 M NaCl	3.0	3.6-10.2	$ThO_2(am)$	Felmy et al. (1991)	Taken from published tables
+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. (1996b)	Taken from published tables

Table 2. Sources of Measured Actinide Solubilities (cont.).

Oxidation State	Solution	Ionic Strength, (M)	pH	Solubility-Controlling Solid	Reference, Type of Data Source	Remarks
+V	WIPP H 17 brine	2.82	7.65	$KNpO_2CO_3$(cr)	Novak et al. (1996b)	Taken from published tables
+V	WIPP SPC brine	7.08	8.56	$KNpO_2CO_3$(cr)	Novak et al. (1996b)	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 3. 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

Note: Table 2 continued on next page.

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

Bin, N	CDF for All +III
2.40	0.987654321
2.55	0.995884774
2.70	0.995884774
2.85	1
3.00	1
3.15	1

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

Bin, N	CDF for All +IV
-2.10	0
-1.95	0
-1.80	0
-1.65	0.022222222
-1.50	0.022222222
-1.35	0.022222222
-1.20	0.022222222
-1.05	0.022222222
-0.90	0.044444444
-0.75	0.088888889
-0.60	0.2
-0.45	0.4
-0.30	0.422222222
-0.15	0.422222222
0.00	0.488888889
0.15	0.511111111
0.30	0.622222222
0.45	0.666666667
0.60	0.688888889
0.75	0.777777778
0.90	0.866666667
1.05	0.911111111
1.20	0.911111111
1.35	0.911111111
1.50	0.933333333
1.65	0.933333333
1.80	0.955555556
1.95	0.955555556
2.10	0.977777778
2.25	0.977777778
2.40	1
2.55	1
2.70	1

Table 5. 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 6. Values of the CDF for Combined An(III, IV, V) Solubility Predictions.

Bin, N	CDF for All Three Oxidation States (An(III, IV, V))
-3.60	0
-3.45	0
-3.30	0
-3.15	0
-3.00	0
-2.85	0.002358491
-2.70	0.002358491
-2.55	0.002358491
-2.40	0.002358491
-2.25	0.002358491
-2.10	0.002358491
-1.95	0.002358491
-1.80	0.011792453
-1.65	0.025943396
-1.50	0.030660377
-1.35	0.049528302
-1.20	0.068396226
-1.05	0.087264151
-0.90	0.101415094
-0.75	0.134433962
-0.60	0.188679245
-0.45	0.233490566
-0.30	0.297169811
-0.15	0.367924528
0.00	0.530660377
0.15	0.653301887
0.30	0.714622642
0.45	0.780660377
0.60	0.801886792
0.75	0.839622642
0.90	0.872641509
1.05	0.896226415
1.20	0.91509434
1.35	0.943396226
1.50	0.95990566

Note: Table 5 continued on next page.

Table 6. 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))
1.65	0.966981132
1.80	0.971698113
1.95	0.983490566
2.10	0.985849057
2.25	0.988207547
2.40	0.992924528
2.55	0.997641509
2.70	0.997641509
2.85	1
3.00	1
3.15	1

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