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Density Calculations of Actinide Solutions using the Pitzer Method

Charles Weber
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OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

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Nuclear Nonproliferation Division

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CONTENTS

LIST OF FIGURES

LIST OF TABLES

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ABSTRACT

This report summarizes an exhaustive literature search for density data of actinide solutions in support of the Nuclear Criticality Safety Program (NCSP). This work comprises an extension of work originally begun in 2003 that implemented an advanced density calculation scheme in the SCALE material input processor. This method demonstrated improved criticality calculations for aqueous systems of $UO_2(NO_3)_2$ and $Pu(NO_3)_4$ in excess acid over a considerable range of concentrations and temperatures. It also provided for UO_2F_2 in acid at room temperature, although with higher uncertainty. The current study was intended to search all available sources for additional data on uranium and plutonium solutions, including the open literature, reports with limited distribution, and unpublished sources such as laboratory notebooks. Data for some of the systems has been regressed to obtain model parameters for the Pitzer formalism of modeling electrolyte solutions.

1. INTRODUCTION

To assess the criticality conditions of actinide solutions, an accurate calculation of the solution density is essential. In 2003, the input processor of the SCALE code system [1] was updated to use the ion-interaction method developed by Pitzer [2], [3] for such calculations. This approach was immediately applicable to systems of uranyl nitrate, $UO_2(NO_3)_2$, in acid because of the plethora of data that was available over a wide range of temperatures [4]. The revised density calculations produced immediate improvements in nuclear criticality calculations [5].

Additional systems were also modeled, including $Pu(NO_3)_4$, $Th(NO_3)_4$, and UO_2F_2, although considerably less data was available for these systems. For $Pu(NO_3)_4$, there was noticeable scatter in some of the data, raising questions that could only be answered by additional high-fidelity experiments. In the case of fluoride systems, data were quite sparse and somewhat questionable because of scatter in available data.

Rather than immediately fund additional density experiments, the NCSP decided to conduct a thorough and up-to-date examination of available data and to assess its integrity and usefulness in further developing the Pitzer method for use in criticality calculations. This report represents an evaluation of some additional data, and an assessment of gaps where additional data would be necessary. In Section 2, we review an exhaustive search through published and unpublished sources for additional density measurements of actinide solutions. In Section 3, we describe the regression of these data to obtain the parameters needed in the Pitzer method. Finally, in Section 4 we describe additional data and analyses that would be needed to provide a comprehensive and robust model of densities in a variety of actinide solutions.

2. DENSITY DATA

A number of sources for density data have been located through painstaking searches of unpublished sources and follow-up of references listed in existing sources. Many of the unpublished sources are old laboratory notebooks for criticality experiments at ORNL made during the 1950s, 1960s, and 1970s. Some of the published sources are found in obscure references that are old, brief, and quite possibly in languages other than English. Thus, although these sources represent a potential treasure of additional data, they must be used soberly, recognizing that the integrity of the final result hinges on the consistency of the data used. The data described herein involve solutions of uranyl fluoride and uranyl sulfate, each with excess acid.

2.1 DATA FOR UO_2F_2

A number of individual data points are available at temperatures 17°C–30°C and are listed in Appendix A. A few of the references are published in open literature, but most of these measurements represent a single record in a laboratory notebook. In general, the data appear to be fairly consistent with each other, as shown in Figure 1 for UO_2F_2 data at 25°C, although there is a little scatter at very high concentrations near 4.3 m for the logbook data. Unpublished measurements are quite consistent with the smoothed values of Söhnel and Novotny [6] and Johnson and Kraus [7].

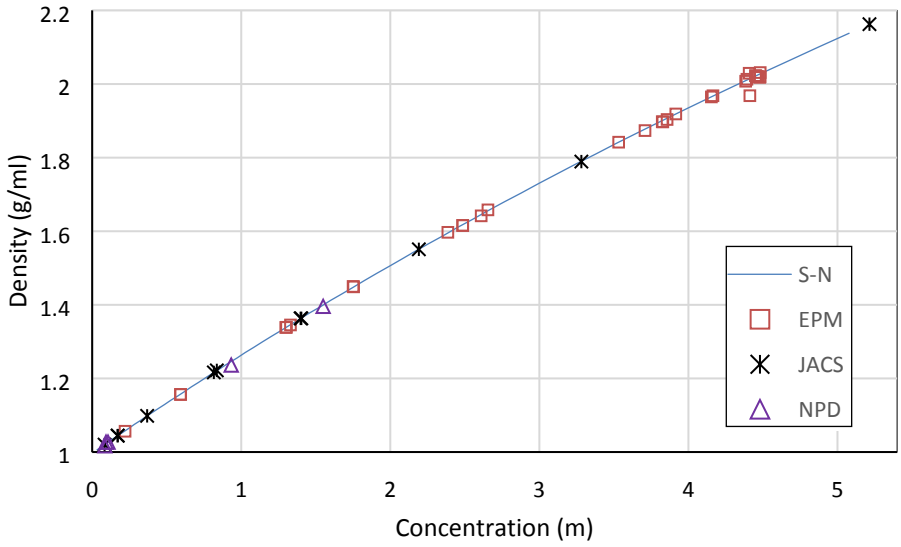

Figure 1. Density of UO_2F_2 Solutions at 25°C. Data from Ref. [6] (S-N), Ref. [7] (JACS), and two unpublished sources from ORNL: Ref. [8] (EPM) and Ref. [9] (NPD).

When multiple temperatures are plotted together (Figure 2), it becomes apparent that there is very little change due to temperature. In fact, any discernable temperature effect is probably within the margin of error for the measurements themselves. Consequently, it is likely that we will not be able to obtain temperature coefficients for the parameter regressions.

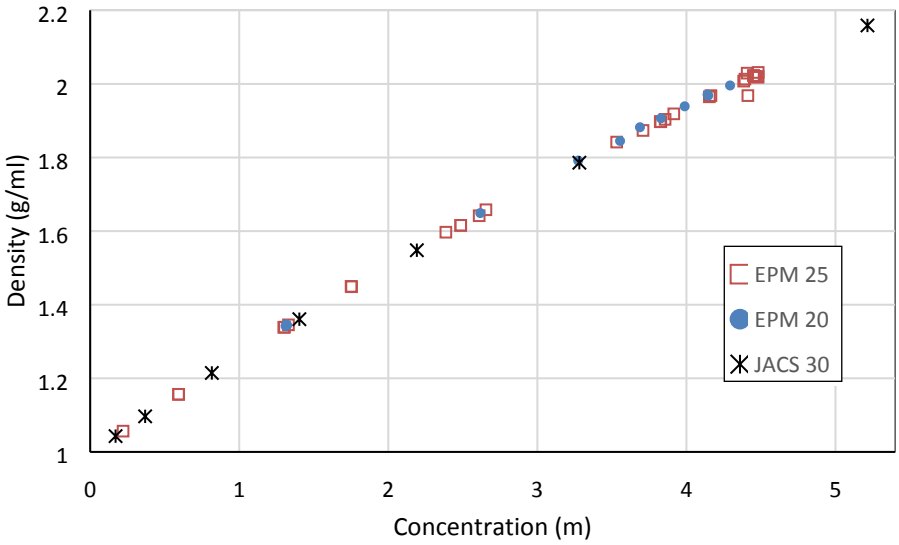

Figure 2. Density of UO_2F_2 at three temperatures. Data from Ref. [8] (EPM) at 20°C and 25°C, and Ref. [7] at 30°C.

2.2 DATA FOR HF

Because systems of UO_2F_2 often occur with excess acid, having data for hydrofluoric acid alone is necessary. Painstaking evaluations of open literature data have yielded a number of obscure sources. A plot in a marketing brochure from Honeywell Corporation (one of the largest US industrial producers of HF) shows specific gravity for temperatures 0°C, 15.6°C, 26.7°C, 37.8°C, and 48.9°C (32°F, 60°F, 80°F, 100°F, and 120°F, respectively). Queries to obtain raw data behind the plot were not successful but did identify a few additional open literature sources. Open sources only span the temperature range 0°C–25°C, although a few points taken from the Honeywell plots represent higher temperatures. All available data are provided in Appendix B.

Data at 15°C are shown in Figure 3 and demonstrate good consistency except at very high concentrations. (As mentioned, our primary concern is for concentrations below 10 m.) Similar results hold at 0°C. However, at 20°C and 25°C, we notice a distinct conflict, as several data sets deviate wildly from each other (Figure 4). The three sets at 20°C all are consistent, and in fact only the data of Winteler [15] are actual measurements. The data from Refs. [13] and [14] represent smoothed values, likely derived from the data of Winteler originally. However, the two data sets at 25°C are in direct conflict—one shows an upward shift with increasing temperature [16], whereas the three points from Ref. [12] indicate a downward shift. The difficulty is further illustrated in Figure 5, where smoothed lines describe data at different temperatures. There is a clear decrease from 0°C to 15°C, but data at 20°C are virtually unchanged from that at 15°C. As temperature rises to 26.6°C and above, densities continue to decrease in a consistent manner. However, these curves represent only the Honeywell data [12].

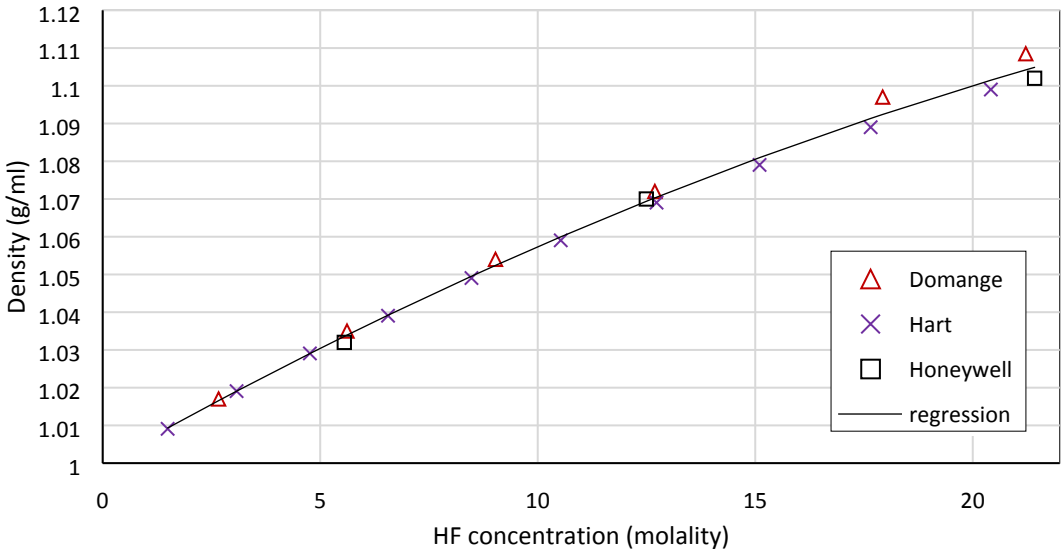

Figure 3. Hydrofluoric acid density at 15°C (Honeywell data at 15.6°C). Data from Domange [10], Hart [11], and Honeywell [12].

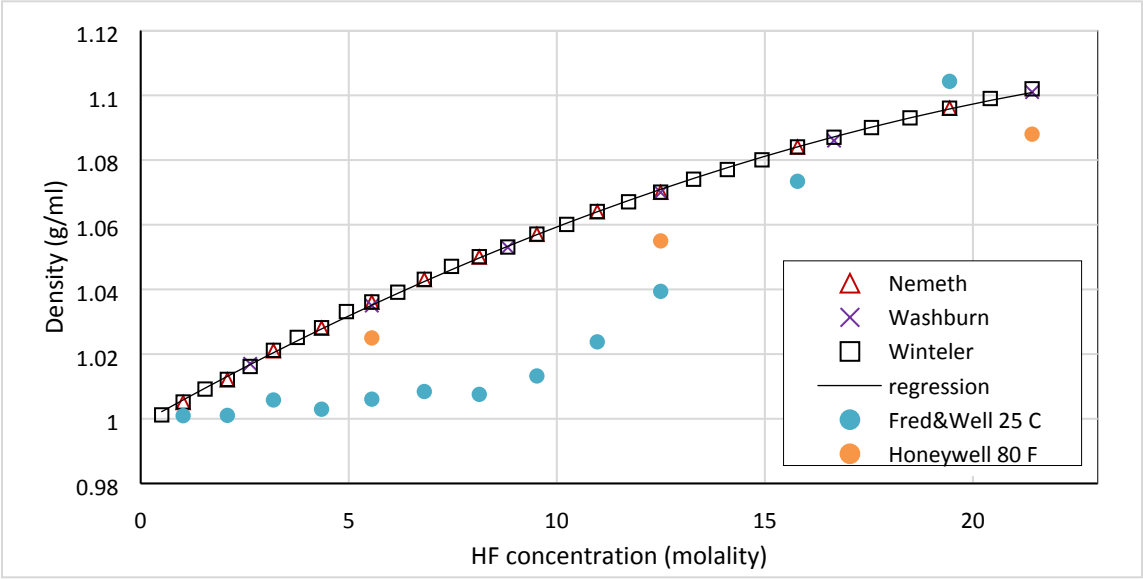

Figure 4. Hydrofluoric acid density at different temperatures. Data at 20°C (Nemeth [13], Washburn [14], Winteler [15]), 25° (Fredenhagen [16]) and 26.6°C C (Honeywell [12]).

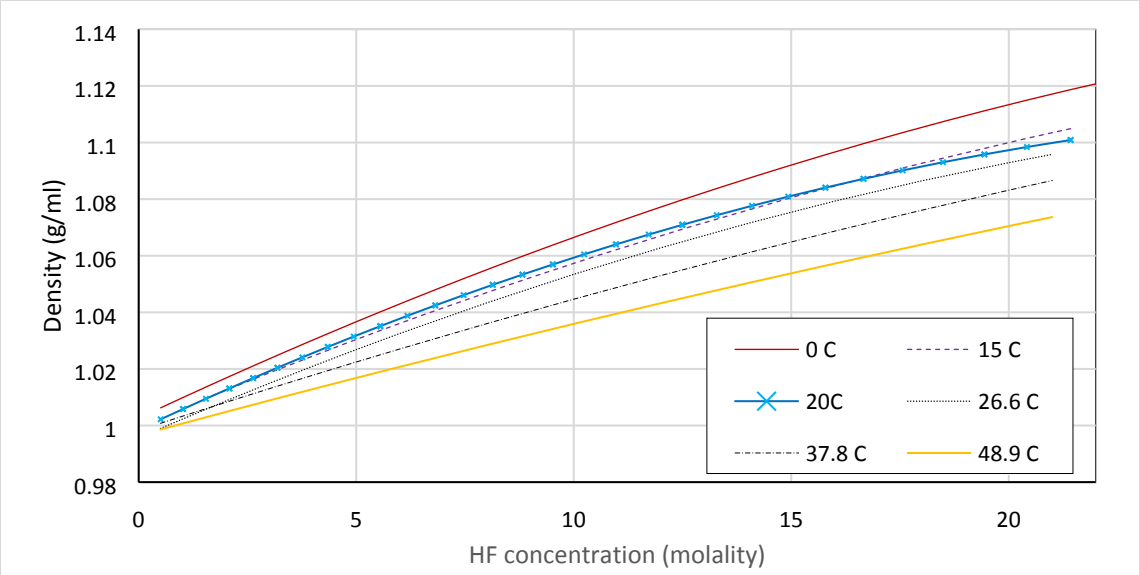

Figure 5. Hydrofluoric acid density at multiple temperatures. Data from ref. 17 (0°C), Refs. 10, 11, 12 (15°C), Refs. 13, 14, 15 (20°C) and Ref. 12 (26.6, 37.8, and 48.9°C).

2.3 DATA FOR UO_2F_2 IN HF

There are very few data for this ternary system involving the common ion F^-. Ferris [18] measured densities of saturated solutions, along with solubilities and solids formed. Since these measurements were made at the solubility limit, they are quite concentrated, as shown in Table 1 below. Hence, only the first few points will be useful in the concentration range of this work.

Table 1. Densities of saturated solutions of UO_2F_2 and HF.

Density	Molality (mol/kg H_2O)		
(g/mL)	H^+	UO_2^{2+}	F^-
2.2969	0	4.19	8.39
1.6879	1.32	2.28	5.87
1.5494	3.45	1.82	7.10
1.5029	5.37	1.70	8.77
1.4715	7.24	1.60	10.43
1.432	10.38	1.45	13.29
1.375	14.70	1.25	17.20
1.357	16.09	1.14	18.37
1.327	19.51	0.999	21.50
1.277	23.12	0.784	24.68
1.225	31.06	0.486	32.03
1.2103	46.82	0.246	47.31

2.4 DENSITIES OF UO_2SO_4 SOLUTIONS

Smoothed densities between 20°C and 90°C are presented by Söhnel and Novotny [6]. These do not represent original measurements but rather are derived from regressions of earlier published data.

Nevertheless, they were used in lieu of the original measurements. An old ORNL report [19] presents data at 25°C and 30°C, which are highly consistent at lower concentrations with the data of Söhnel and Novotny. However, deviation is noticeable at higher concentrations, as shown in Figure 6. From Figure 7, there is a clear trend of decreasing density with increasing temperature. All data for all temperatures are listed in Appendix C.

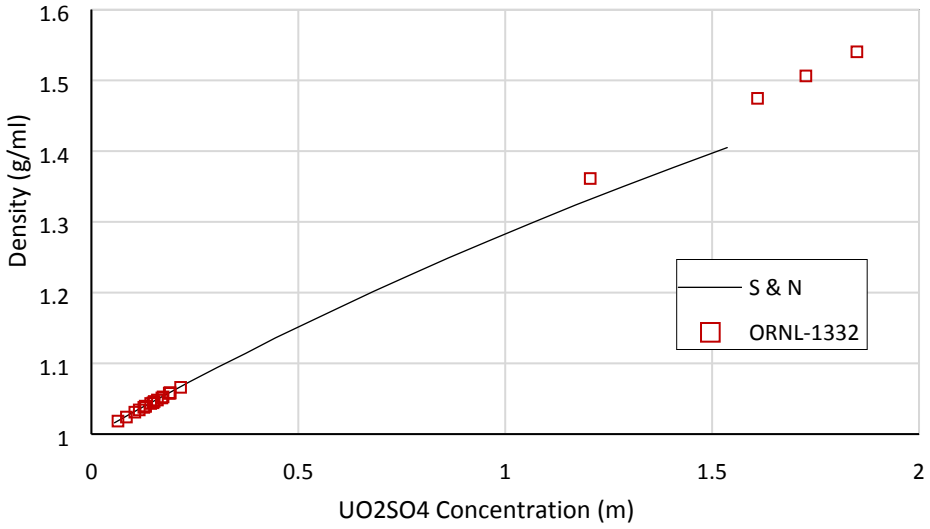

Figure 6. Density of UO_2SO_4 solutions at 25°C. Data taken from Refs. [6] and [19].

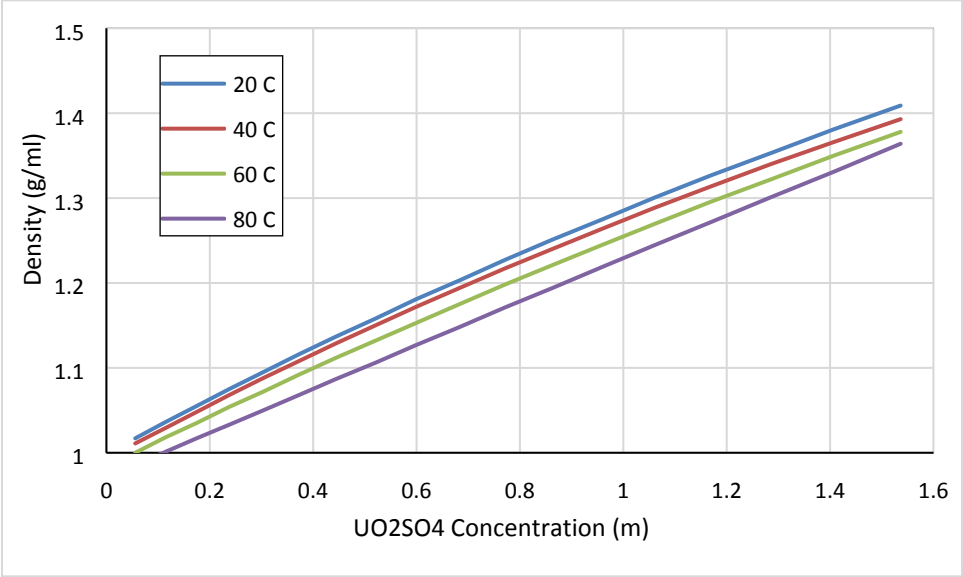

Figure 7. UO_2SO_4 density at different temperatures. Data taken from Ref. [6].

2.5 DENSITIES OF H_2SO_4 SOLUTIONS

Again, the primary source of data are the smoothed densities between 0°C and 100°C presented by Söhnel and Novotny [6]. As with the UO_2SO_4 values, these are used directly instead of the original published measurements from which they were derived. Values at 20°C given in the CRC Handbook [29] are identical to those of Ref. [6], and likely were derived from the same source; because they are redundant,

they will not be included in our data set. There are a number of more recent measurements summarized in Oca et al. [20], but these have not been pursued. One recent report from Los Alamos National Laboratory also measured densities between 7°C and 25°C [21], and these compare favorably with the other data at two temperatures in Figure 8. As expected, there is a clear trend of decreasing density with increasing temperature. All data for all temperatures are listed in Appendix D.

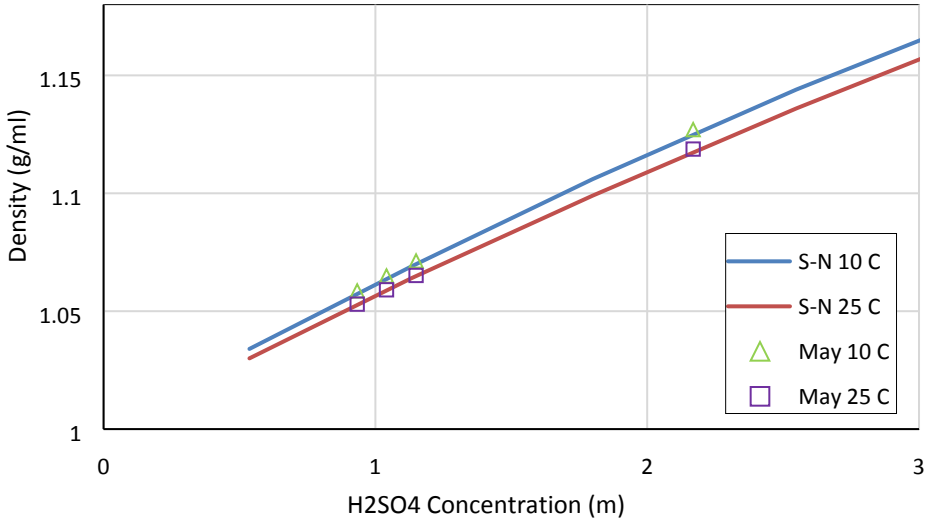

Figure 8. H_2SO_4 Densities at 10°C and 25°C. Data taken from Refs. [6] and [20].

2.6 TERNARY SYSTEM UO_2SO_4–H_2SO_4–H_2O

As was done in Section 2.3 for the fluoride system, we now examine the system involving uranyl ion in excess acid with a common sulfate ion. Two data sets are available from Refs. [19] and [21]. The latter does not fully explain concentrations in solution and will not be useful without additional follow-up. Data from Ref. [19] at 30°C are given in Table 2.

Table 2. Densities in ternary sulfate system at 30°C.

Density	Molarity (mol/L)		Molality (mol/kg H_2O)		
(g/mL)	UO_2SO_4	H_2SO_4	UO_2^{2+}	H^+	SO_4^{2-}
1.0312	0.105029	0	0.105796	0	0.105796
1.0279	0.126035	0	0.128377	0	0.128377
1.0452	0.147041	0	0.148321	0	0.148321
1.0518	0.168047	0	0.169696	0	0.169696
1.0585	0.189053	0	0.191099	0	0.191099
1.0653	0.210059	0	0.212524	0	0.212524
1.0318	0.105029	0.02	0.105942	0.040347	0.126115
1.0384	0.126035	0.02	0.12727	0.040392	0.147466
1.0457	0.147041	0.02	0.14854	0.040408	0.168744
1.0515	0.168047	0.02	0.170085	0.040485	0.190327
1.0591	0.189053	0.02	0.191363	0.040489	0.211607
1.0659	0.210059	0.02	0.212817	0.040525	0.23308

1.0332	0.105029	0.1	0.106635	0.203058	0.208164
1.0399	0.126035	0.1	0.128091	0.203262	0.229722

Table 2. Densities in ternary sulfate system at 30°C (continued).

Density	Molarity (mol/L)		Molality (mol/kg H_2O)		
(g/mL)	UO_2SO_4	H_2SO_4	UO_2^{2+}	H^+	SO_4^{2-}
1.0468	0.147041	0.1	0.149559	0.203425	0.251272
1.0532	0.168047	0.1	0.171149	0.203693	0.272996
1.0604	0.189053	0.1	0.192639	0.203794	0.294536
1.0672	0.210059	0.1	0.214238	0.203979	0.316228
1.0348	0.105029	0.2	0.107531	0.409528	0.312295
1.0416	0.126035	0.2	0.129155	0.409902	0.334106
1.0483	0.147041	0.2	0.150834	0.410318	0.355993
1.0551	0.168047	0.2	0.172539	0.410693	0.377886
1.062	0.189053	0.2	0.194264	0.411026	0.399777
1.0688	0.210059	0.2	0.216047	0.411403	0.421748
1.0376	0.105029	0.4	0.109415	0.833405	0.526117
1.0448	0.126035	0.4	0.131365	0.83383	0.54828
1.0515	0.147041	0.4	0.153417	0.834692	0.570763
1.0581	0.168047	0.4	0.175534	0.835642	0.593355
1.0648	0.189053	0.4	0.19768	0.836507	0.615933
1.0713	0.210059	0.4	0.219918	0.837549	0.638693

3. PARAMETER ESTIMATION

In this section, we evaluate the data from Section 2 to develop the parameters needed to implement the Pitzer formalism for fluoride and sulfate systems involving the uranyl ion in acid. The calculational approach is identical to that undertaken previously in earlier work [4] and will only be summarized here.

The Pitzer model for calculating densities of electrolyte solutions is based on a theoretical development that uses empirical parameters to describe ion interactions. The model is described in detail in Appendix E, but here we identify only the parameters that will be determined from density data:

$\beta_{ca}^{v(0)},\ \beta_{ca}^{v(1)},\ C_{ca}^{v} =$ parameters describing interaction of cation c and anion a
$\theta_{cc'}^{v} =$ parameter describing interaction of cations c and c'
$\psi_{cc'a}^{v} =$ parameter describing interaction of cations c and c' and anion a
$\overline{V}_i^o =$ partial molar volume (at infinite dilution) of individual salts (cation–anion pairs)

In this report, we only have cations UO_2^{2+} and H^+, and we consider systems with a single anion (either F^- or SO_4^{2-}, considered separately, but not together). Note, that the general formulation [2]–[4] would also include interactions of multiple anions, but we do not consider such systems in this report.

Each of these parameters could vary with temperature, so we consider a general dependence of the form:

$$h(T) = A + B(T - T_0) + C\left[\frac{1}{T} - \frac{1}{T_0}\right] + D\ln\left[\frac{T}{T_0}\right] + E\left[\frac{1}{T^2} - \frac{1}{T_0^2}\right], \quad T_0 = 298.15\ \text{K}. \tag{1}$$

Note that we only report the parameters that were included in the regression. Parameters that are not mentioned are assumed to be zero except where noted.

Importantly, both the fluoride and sulfate systems are notoriously ill-behaved primarily because of ion association and secondary reactions [22], [23]. In the fluoride system, the acid dissociation may only be 10%–15%, and we have the additional reaction:

$$\mathrm{HF} + \mathrm{F}^- \leftrightarrow \mathrm{HF}_2^- \ . \tag{2}$$

For the sulfate system, both dissociation reactions must be considered:

$$\mathrm{H_2SO_4} \leftrightarrow \mathrm{H}^+ + \mathrm{HSO_4^-} \tag{3a}$$

$$\mathrm{HSO_4^-} \leftrightarrow \mathrm{H}^+ + \mathrm{SO_4^{2-}} \ . \tag{3b}$$

In both cases, multiple anions are introduced for the binary system of acid alone, and this complication affects the ternary system that includes both acid and uranyl ions. This behavior is in contrast to nitrate systems, where the ions dissociate almost completely upon dissolution in water. Thus, any model of the fluoride and sulfate systems will be difficult to implement without special treatment that includes the additional aqueous species.

3.1 FLUORIDE SYSTEM

The data available for UO_2F_2 covers temperatures from approximately 17°C to 30°C and includes a total of 196 samples and solute concentrations from 0.0063 to 5.21 *m* UO_2F_2. Of the 196 samples, 108 were at 25°C. Within our limited temperature range, the data shows almost no variation with respect to temperature; hence, we performed the fit using all 196 data points disregarding temperature. The fit parameters are reported in Table 3, and the fit model is plotted against the data in Figure 9.

Table 3. Fit parameters for UO_2F_2.

Parameter	A [cf. Eq. (1)]
$\boldsymbol{\beta_{ca}^{v0}}$	1.438E-3
$\boldsymbol{\beta_{ca}^{v1}}$	1.429E-2
$\boldsymbol{C_{ca}^{v}}$	-9.430E-5
$\boldsymbol{\overline{V}_i^0}$	5.787E1

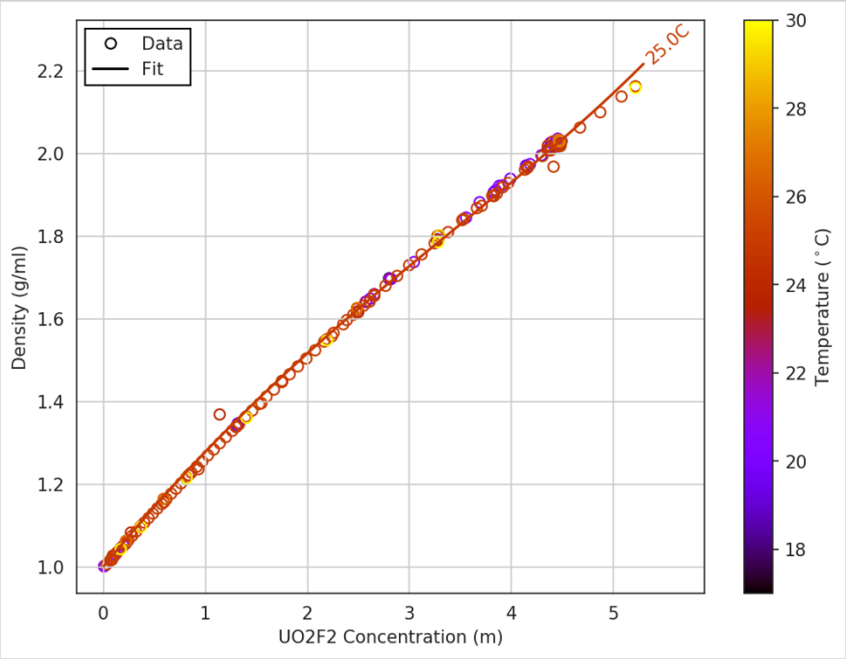

Figure 9. Plot of data and model fit for UO_2F_2.

Figure 9 shows that most of the data follows a smooth trend, which the model tracks well up to approximately 4.5 *m* UO_2F_2. The model begins to diverge for concentrations in excess of 4.5 *m*, and its use is discouraged outside this range. We were unable to find a fit that performed better in this region and provided a good fit at lower concentrations. Additionally, the fit tracks well for temperatures between 17°C and 30°C, and the very weak dependence on temperature suggests that this fit is valid over the entire temperature range and possibly beyond.

A total of 77 measurements were provided for HF with concentrations from 0.050 to 7.95 *m*. Temperatures range from 0°C to 50°C, and 10 are at 25°C. There appears to be some dependence on temperature, but we were unable to find good fits for the temperature-dependent parameters. We suspect the temperature dependence is obscured by measurement error and the need to model the additional species in Eq. (2). The fit parameters are given in Table 4, with the model and data plotted in Figure 10.

Table 4. Fit parameters for HF.

Parameter	A [cf. Eq. (1)]
$\boldsymbol{\beta_{ca}^{v0}}$	-5.041E-4
$\boldsymbol{\beta_{ca}^{v1}}$	9.5473E-3
$\boldsymbol{C_{ca}^{v}}$	2.430E-5
$\boldsymbol{\overline{V}_{i}^{0}}$	5.000E1

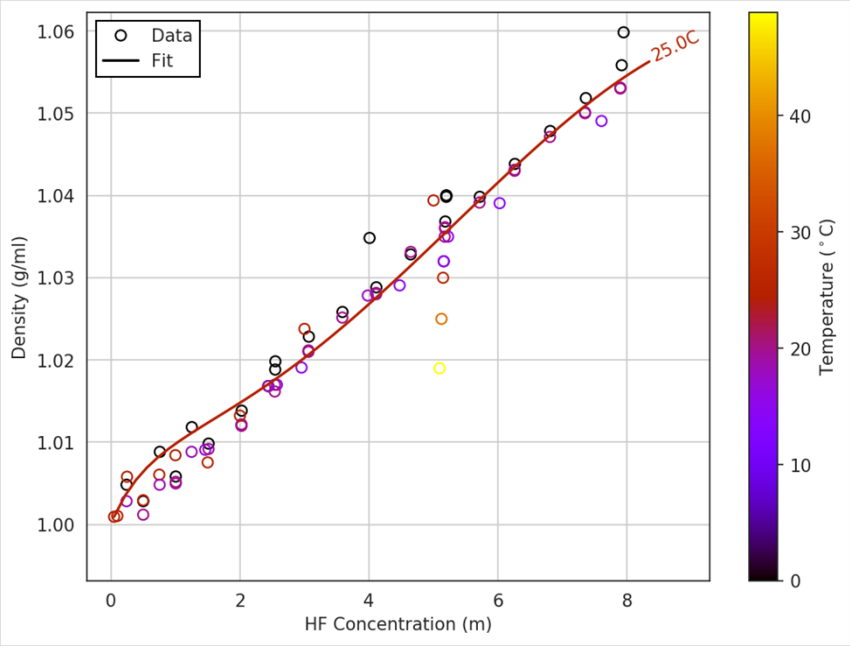

Figure 10. Plot of data and model fit for HF.

Fits for the temperature-independent coefficients were performed using the data at 25°C and agree with the data for concentrations up to 4–5 *m* and possibly even as high as 7–8 *m*. However, the unusual curvature of the model are cause for concern, and reflect the difficulties of modeling this solution that have been noted by others [2]. Note that several of the measurements with temperatures in excess of 40°C appear to have considerable variation in temperature, but there were very few data points at higher temperatures and we were unable to find a fit that agreed with the reported densities for these samples. This is likely due to the small number of samples at these higher temperatures and the high amount of variation in the reported densities.

3.2 SULFATE SYSTEM

Data for the UO_2SO_4 solutions totaled 293 samples at temperatures between 20°C and 90°C, with solute concentrations from 0.050 to 1.39 *m*. Of the 293 samples, 35 were recorded at 25°C. The data varies smoothly with concentration and contains significant variations in temperature, but we were again not able to find a fit for the temperature-dependent coefficients in Eq. 1. This indicates a potential inconsistency of the data with the model that requires additional modeling effort. Table 5 gives the fit parameters, while Figure 11 plots the fit and data. Note that the UO_2^{2+} and SO_4^{2-} ions are both doubly charged and use the special form of the model described in Appendix E.

Table 5. Fit parameters for UO_2SO_4.

Parameter	A
$\boldsymbol{\beta_{ca}^{v0}}$	3.8663E-3
$\boldsymbol{\beta_{ca}^{v1}}$	-4.0519E-2
$\boldsymbol{\beta_{ca}^{v2}}$	4.6874E0
$\boldsymbol{C_{ca}^{v}}$	2.7632E-4
$\boldsymbol{\overline{V}_{i}^{0}}$	3.7153E0

Figure 11 shows that the fit follows the data for UO_2SO_4 across the whole range of concentrations, though this range only extends to approximately 1.4 *m* UO_2SO_4. We see that the fit at 25°C is highly consistent with the corresponding data and reasonably accurate from 20°C to 35°C but diverges from reported densities at higher temperatures.

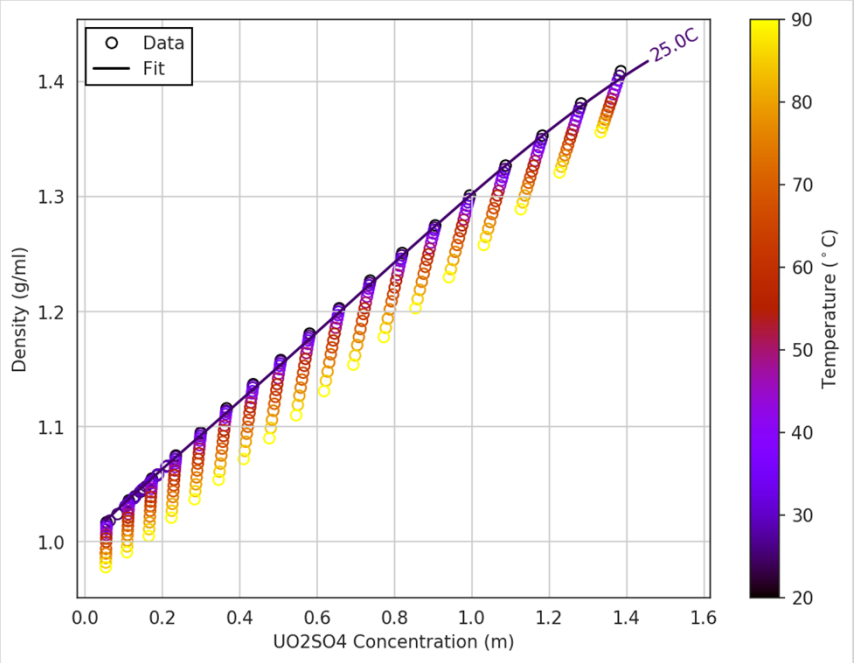

Figure 11. Plot of data and model fit for UO_2SO_4.

For sulfuric acid, 72 measurements were available at temperatures from 0°C to 100°C, and 6 were at 25°C. Concentrations varied from 0.53 to 4.37 *m* H_2SO_4. Fits suffer from the same problem as those for UO_2SO_4, where we were able to identify a good fit to the data at 25°C but were unable to find temperature-dependent parameters that agree with the data at other temperatures. Parameter values are listed in Table 6, and the fit is plotted with the data in Figure 12.

Table 6. Fit parameters for H_2SO_4.

Parameter	A
$\boldsymbol{\beta_{ca}^{v0}}$	-6.4000E-4
$\boldsymbol{\beta_{ca}^{v1}}$	3.8120E-2
$\boldsymbol{C_{ca}^{v}}$	4.7482E-5
$\boldsymbol{\overline{V}_{i}^{0}}$	4.3693E1

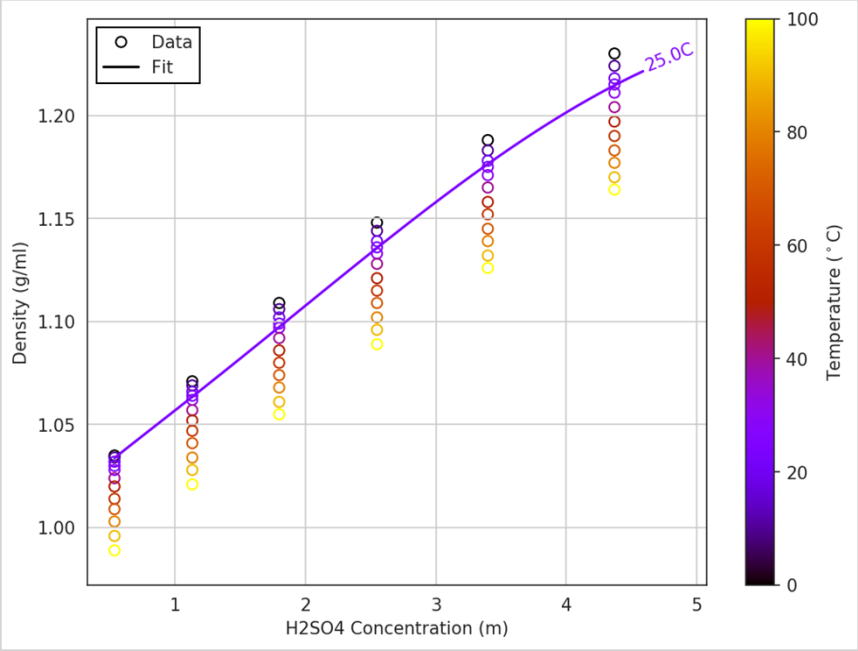

Figure 12. Plot of data and model fit for H_2SO_4.

As with the previous case, Figure 12 shows that the fit is highly consistent with the data at 25°C for all concentrations and reasonably describes the data between 15°C and 30°C.

3.3 TWO SPECIES SYSTEM

A total of 12 measurements were available for two species solutions containing UO_2F_2 and HF; however, the majority of these solutions were at concentrations well in excess of the concentrations for the corresponding binary solutions. We limited our analysis to measurements at concentrations that are consistent with the corresponding single species solution data, leaving only three measurements suitable for analysis. All of these measurements were recorded at 25°C, and no attempt was made to fit temperature-dependent coefficients. The parameters from fitting these three measurements are given in Table 7, and a comparison to the fit model predictions is listed in Table 8. Note that values for $\overline{V}_i^0$, β_{ca}^{v0}, β_{ca}^{v1}, and C_{ca}^{v} determined from the previous fits for binary solutions were fixed at the values given in Tables 3 and 4.

Table 7. Fit parameters for combined ternary fluoride solutions.

Parameter	A
$\boldsymbol{\theta^v[H^+,UO_2^{2+}]}$	-1.4560E-2
$\boldsymbol{\psi^v[H^+,UO_2^{2+},F^-]}$	2.6780E-3

Table 8. Measured and predicted densities for ternary fluoride solutions.

HF (m)	UO_2F_2 (m)	Meas. Density (g/mL)	Calc. Density (g/mL)	Difference (%)
1.319316	2.2762853	1.6879	1.6535	2.04%
3.454064	1.8240561	1.5494	1.5788	-1.90%
5.373435	1.7005814	1.5029	1.4933	0.64%

Data for solutions containing both UO_2SO_4 and H_2SO_4 consisted of a total of 29 measurements, all recorded at 30°C. Concentrations of UO_2SO_4 varied from 0.100 to 0.220 m UO_2SO_4, whereas acid concentrations ranged from 0.0200 to 0.4188 m H_2SO_4. As with the fluoride solutions, binary system parameter values were fixed at the values given in Tables 5 and 6. Unfortunately, we were not able to find values for θ^v and ψ^v that produce fits that agree with the data for the mixed UO_2SO_4–H_2SO_4–H_2O solutions. This is likely due to the dissociation behavior of H_2SO_4 as mentioned previously.

4. SUMMARY OF DATA AND MODELING NEEDS

4.1 UO_2F_2

As demonstrated in Section 3.1, data for this system are adequate for a reasonable model at room temperature (20°C–30°C). Model applications outside this range are unknown because no data exist. Potential applications may extend to near boiling because some reprocessing operations use mixtures of HF and HNO_3 to dissolve nuclear fuel at temperatures near 100°C. However, the complications of this system will also require more robust model development, especially if it is to be extended to a greater temperature range.

4.2 UO_2SO_4

Data appear to be adequate, unless systems of multiple acids (e.g., H_2SO_4 + HNO_3) are encountered. However, the difficulty in modeling this system may require additional verification data, especially for systems including excess acid. As noted in Section 3, this system is quite difficult to model, and additional work is required. The model in Sections 3.2 and 3.3 indicates that a model at room temperature could be constructed but could not reliably be extended to other temperatures.

This system is the active solution in the SHINE Medical Technologies process for production of ^{99}Mo. [24]. It is also important for other processes, and therefore additional data and modeling effort is warranted.

4.3 $PuCl_3$

There is a need for density prediction of plutonium chloride solutions so that systems that are more realistic than the fictitious metal-water system may be modeled in criticality calculations. Current methods are conservative and do not take into account any chloride. Los Alamos is pursuing data for this system and has installed equipment for density measurements. They plan to obtain data at room temperature over the next year. It would be helpful for this project to contribute to experiments and to be able to model results.

4.4 UCl_3

As with the plutonium system, density prediction for uranium chloride solutions would allow systems that are more realistic than the fictitious metal-water system to be modeled in criticality calculations. There are active needs for data and calculational support at several U.S. facilities.

4.5 MIXED ACTINIDES

There is one data set for U–Th systems in acid [4], but no data have been evaluated for mixed U–Pu systems in acid. There is also a need for data for mixed Pu-Am systems. Although there have been discussions of future work to obtain such measurements for chloride systems at Los Alamos, efforts are not currently funded.

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APPENDIX A. DATA FOR UO_2F_2 SOLUTIONS

Temp. (°C)	Density (g/mL)	Molarity (mol/L)	Molality (mol/kg-H_2O)	Reference
17	1.6979	2.554	2.802	[8] Book 48
18	1.6976	2.553	2.802	[8] Book 48
18	1.6976	2.553	2.802	[8] Book 48
19	1.7921	2.921	3.274	[8] Book 48
19	2.0176	3.789	4.455	[8] Book 96
19	2.0298	3.826	4.490	[25]
18	2.0200	3.789	4.438	[26]
18	2.0300	3.825	4.486	[26]
18.5	1.7921	2.921	3.274	[8] Book 48
19.5	2.0255	3.804	4.455	[8] Book 96
19.5	1.3474	1.268	1.325	[8] Book 99
20	1.3420	1.253	1.311	[8] Book 48
20	1.3467	1.262	1.317	[8] Book 95
20	1.3442	1.262	1.321	[8] Book 95
20	1.6491	2.390	2.617	[8] Book 95
20	1.7917	2.921	3.274	[8] Book 95
20	1.8451	3.131	3.555	[8] Book 96
20	1.8823	3.250	3.688	[8] Book 96
20	1.9067	3.350	3.829	[8] Book 96
20	1.9392	3.471	3.988	[8] Book 96
20	1.9683	3.584	4.146	[8] Book 99
20	1.9712	3.587	4.142	[8] Book 99
20	1.9958	3.689	4.293	[8] Book 99
20.3	2.0100	3.739	4.356	[8] Book 48
20.5	1.9059	3.347	3.825	[8] Book 95
20.5	1.9112	3.364	3.845	[8] Book 95
20.5	1.9220	3.397	3.879	[8] Book 95
20.5	1.9744	3.608	4.181	[8] Book 96
20.5	2.0167	3.760	4.380	[8] Book 96
20.5	2.0266	3.780	4.384	[8] Book 96
20.5	2.0280	3.800	4.431	[8] Book 96
20.8	2.0361	3.824	4.455	[8] Book 96
21	1.0011	0.006	0.006	[8] Book 3
21	1.0030	0.016	0.016	[8] Book 3
21	1.0298	0.118	0.119	[8] Book 3
21	1.0511	0.195	0.196	[8] Book 3
21	1.6966	2.552	2.816	[8] Book 48
21	1.6966	2.552	2.816	[8] Book 48

Temp. (°C)	Density (g/mL)	Molarity (mol/L)	Molality (mol/kg-H_2O)	Reference
21	1.6966	2.552	2.816	[8] Book 48
21	1.6966	2.552	2.816	[8] Book 48
21	1.6966	2.552	2.816	[8] Book 48
21	1.7374	2.729	3.043	[8] Book 95
21	1.9229	3.409	3.906	[8] Book 96
21	2.0096	3.738	4.356	[8] Book 48
21.5	2.0288	3.807	4.448	[8] Book 95
22	1.6412	2.350	2.573	[8] Book 48
22	1.6412	2.350	2.573	[8] Book 48
22	1.6412	2.350	2.573	[8] Book 48
22	1.6412	2.350	2.573	[8] Book 48
22	1.6593	2.423	2.655	[8] Book 48
22	1.6593	2.423	2.655	[8] Book 48
22	1.6593	2.423	2.655	[8] Book 48
22	1.6962	2.551	2.802	[8] Book 48
22	2.0091	3.737	4.356	[8] Book 48
22.2	2.0091	3.737	4.356	[8] Book 48
22.5	2.0091	3.737	4.356	[8] Book 48
22.5	2.0091	3.737	4.356	[8] Book 48
				[8] Book 48
23	1.6165	2.276	2.486	[8] Book 48
23	1.6165	2.276	2.486	[8] Book 48
23	1.6185	2.284	2.496	[8] Book 48
23	1.7905	2.919	3.274	[8] Book 48
23	1.7905	2.919	3.274	[8] Book 48
23	1.7905	2.919	3.274	[8] Book 48
23.3	2.0086	3.736	4.356	[8] Book 48
				[8] Book 48
24	1.0830	0.270	0.270	[8] Book 48
24	1.3687	1.512	1.140	[8] Book 48
24	1.6161	2.281	2.498	[8] Book 97
24	1.7901	2.918	3.274	[8] Book 97
24.2	2.0082	3.736	4.356	[8] Book 48
24.4	2.0191	3.756	4.356	[8] Book 48
24.5	1.6249	2.288	2.486	[8] Book 48
24.5	1.6249	2.288	2.486	[8] Book 48
24.5	1.0626	0.223	0.225	[8] Book 48
25	1.0060	0.033	0.033	[6]
25	1.0150	0.066	0.066	[6]
25	1.0250	0.100	0.100	[6]
25	1.0350	0.134	0.135	[6]

Temp. (°C)	Density (g/mL)	Molarity (mol/L)	Molality (mol/kg-H_2O)	Reference
25	1.0440	0.169	0.171	[6]
25	1.0540	0.205	0.207	[6]
25	1.0640	0.242	0.244	[6]
25	1.0750	0.279	0.282	[6]
25	1.0850	0.317	0.321	[6]
25	1.0960	0.356	0.361	[6]
25	1.1070	0.395	0.401	[6]
25	1.1180	0.436	0.443	[6]
25	1.1290	0.476	0.485	[6]
25	1.1410	0.519	0.528	[6]
25	1.1520	0.561	0.573	[6]
25	1.1640	0.605	0.618	[6]
25	1.1770	0.650	0.665	[6]
25	1.1890	0.695	0.713	[6]
25	1.2020	0.741	0.762	[6]
25	1.2150	0.789	0.812	[6]
25	1.2280	0.837	0.863	[6]
25	1.2420	0.887	0.916	[6]
25	1.2550	0.937	0.970	[6]
25	1.2700	0.990	1.025	[6]
25	1.2840	1.042	1.082	[6]
25	1.2990	1.096	1.141	[6]
25	1.3140	1.152	1.201	[6]
25	1.3290	1.208	1.263	[6]
25	1.3450	1.266	1.326	[6]
25	1.3610	1.326	1.391	[6]
25	1.3780	1.387	1.459	[6]
25	1.3940	1.448	1.528	[6]
25	1.4120	1.513	1.599	[6]
25	1.4290	1.577	1.672	[6]
25	1.4470	1.644	1.748	[6]
25	1.4660	1.713	1.826	[6]
25	1.4850	1.784	1.907	[6]
25	1.5040	1.855	1.990	[6]
25	1.5240	1.930	2.076	[6]
25	1.5450	2.006	2.164	[6]
25	1.5660	2.084	2.256	[6]
25	1.5870	2.164	2.351	[6]
25	1.6100	2.248	2.449	[6]
25	1.6320	2.331	2.551	[6]
25	1.6560	2.419	2.656	[6]

Temp. (°C)	Density (g/mL)	Molarity (mol/L)	Molality (mol/kg-H_2O)	Reference
25	1.6800	2.509	2.766	[6]
25	1.7040	2.600	2.879	[6]
25	1.7300	2.696	2.997	[6]
25	1.7560	2.793	3.119	[6]
25	1.7830	2.894	3.246	[6]
25	1.8100	2.997	3.379	[6]
25	1.8390	3.105	3.517	[6]
25	1.8680	3.214	3.661	[6]
25	1.8980	3.327	3.811	[6]
25	1.9290	3.444	3.968	[6]
25	1.9610	3.565	4.132	[6]
25	1.9940	3.690	4.303	[6]
25	2.0280	3.819	4.483	[6]
25	2.0630	3.952	4.672	[6]
25	2.1000	4.091	4.870	[6]
25	2.1380	4.234	5.078	[6]
25	1.0565	0.218	0.221	[8] Book 48
25	1.1566	0.579	0.592	[8] Book 48
25	1.1565	0.580	0.594	[8] Book 48
25	1.6157	2.275	2.486	[8] Book 48
25	1.6157	2.275	2.486	[8] Book 48
25	1.6581	2.422	2.655	[8] Book 48
25	2.0077	3.735	4.384	[8] Book 48
25	1.5971	2.196	2.386	[8] Book 89
25	1.8736	3.243	3.709	[8] Book 95
25	1.8976	3.332	3.824	[8] Book 95
25	1.8972	3.333	3.828	[8] Book 95
25	1.9037	3.356	3.857	[8] Book 95
25	2.0198	3.797	4.466	[8] Book 95
25	2.0312	3.824	4.482	[8] Book 95
25	1.3390	1.245	1.303	[8] Book 96
25	1.4499	1.649	1.750	[8] Book 96
25	1.4489	1.650	1.754	[8] Book 96
25	1.6418	2.375	2.610	[8] Book 96
25	1.8420	3.116	3.532	[8] Book 96
25	1.9187	3.405	3.915	[8] Book 96
25	1.9650	3.581	4.154	[8] Book 96
25	1.9682	3.591	4.165	[8] Book 96
25	1.9683	3.681	4.412	[8] Book 96
25	2.0130	3.758	4.393	[8] Book 96
25	2.0227	3.797	4.450	[8] Book 96

Temp. (°C)	Density (g/mL)	Molarity (mol/L)	Molality (mol/kg-H_2O)	Reference
25	2.0223	3.797	4.452	[8] Book 96
25	2.0293	3.794	4.408	[8] Book 98
25	2.0207	3.796	4.460	[8] Book 98
25	2.0181	3.799	4.479	[8] Book 98
25	2.0199	3.803	4.481	[8] Book 98
25	1.3386	1.242	1.299	[8] Book 99
25	1.3453	1.269	1.330	[8] Book 99
25	1.0202	0.083	0.083	[7]
25	1.0443	0.169	0.170	[7]
25	1.0448	0.170	0.172	[7]
25	1.0983	0.364	0.369	[7]
25	1.2164	0.793	0.816	[7]
25	1.2219	0.812	0.836	[7]
25	1.3632	1.332	1.398	[7]
25	1.3632	1.336	1.403	[7]
25	1.5509	2.029	2.192	[7]
25	1.7893	2.920	3.282	[7]
25	2.1627	4.327	5.215	[7]
25	1.0270	0.105	0.106	[9]
25	1.0170	0.084	0.085	[9]
25	1.3959	1.460	1.550	[9]
25	1.2363	0.893	0.933	[9]
25	1.0270	0.092	0.093	[9]
25.6	1.5580	2.065	2.239	[8] Book 2
26	1.8007	2.935	3.274	[8] Book 48
26.2	1.6257	2.289	2.486	[8] Book 48
26.6	1.1644	0.583	0.592	[8] Book 48
26.6	1.1644	0.583	0.592	[8] Book 48
26.6	1.0622	0.220	0.222	[8] Book 48
26.6	1.0622	0.220	0.222	[8] Book 48
26.8	2.0335	3.821	4.462	[8] Book 98
27	1.6262	2.289	2.486	[8] Book 48
27.4	2.0295	3.817	4.470	[8] Book 98
29	1.8023	2.938	3.292	[8] Book 48
30	1.0429	0.169	0.170	[7]
30	1.0967	0.363	0.369	[7]
30	1.2146	0.792	0.816	[7]
30	1.3607	1.333	1.403	[7]
30	1.5481	2.025	2.192	[7]

Temp. (°C)	Density (g/mL)	Molarity (mol/L)	Molality (mol/kg-H_2O)	Reference
30	1.7859	2.915	3.282	[7]
30	2.1589	4.320	5.215	[7]

APPENDIX B. DATA FOR HF

Temp (°C)	Density (g/mL)	Molality (mol/kg-H_2O)	Molarity (mol/L)	Reference
0	1.002842	0.504899035	0.501262	[17]
0	1.005842	1.020102133	1.005523	[17]
0	1.009841	1.545927974	1.514281	[17]
0	1.01384	2.082708521	2.027038	[17]
0	1.01884	2.63078971	2.546291	[17]
0	1.022839	3.190532202	3.067544	[17]
0	1.025839	3.762312167	3.589296	[17]
0	1.028838	4.34652213	4.114047	[17]
0	1.032837	4.943571873	4.646295	[17]
0	1.036837	5.553889389	5.18254	[17]
0	1.039836	6.177921904	5.717286	[17]
0	1.043836	6.816136977	6.261028	[17]
0	1.047835	7.469023661	6.808768	[17]
0	1.051834	8.137093756	7.360506	[17]
0	1.055834	8.820883147	7.916243	[17]
0	1.059833	9.520953238	8.475977	[17]
0	1.063833	10.23789249	9.039709	[17]
0	1.068832	10.97231806	9.616436	[17]
0	1.072831	11.7248776	10.18866	[17]
0	1.07683	12.49625112	10.76489	[17]
0	1.08283	13.28715309	11.36611	[17]
0	1.085829	14.0983346	11.94033	[17]
0	1.089828	14.93058576	12.52905	[17]
0	1.092828	15.78473826	13.10978	[17]
0	1.096827	16.66166817	13.70599	[17]
0	1.099827	17.56229888	14.29322	[17]
0	1.103826	18.4876044	14.89693	[17]
0	1.106826	19.43861286	15.49065	[17]
0	1.110825	20.41641029	16.10186	[17]
0	1.113825	21.42214479	16.70207	[17]
0	1.117824	22.45703101	17.32078	[17]
0	1.004842	0.243104045	0.243095	[27]
0	1.008841	0.763253805	0.758408	[27]
0	1.011841	1.271152698	1.254285	[27]
0	1.01684	2.520252328	2.439642	[27]
0	1.034837	4.199282221	4.008722	[27]
0	1.064832	9.414882012	8.436121	[27]
0	1.096827	16.19400318	13.41543	[27]

Temp (°C)	Density (g/mL)	Molality (mol/kg-H_2O)	Molarity (mol/L)	Reference
0	1.109825	19.90454318	15.7989	[27]
0	1.119824	21.24914756	16.69688	[27]
0	1.019839	2.63078971	2.54879	[14]
0	1.039836	5.553889389	5.197533	[14]
0	1.059833	8.820883147	7.946228	[14]
0	1.07983	12.49625112	10.79488	[14]
0	1.098827	16.66166817	13.73098	[14]
0	1.118824	21.42214479	16.77704	[14]
0	1.04	5.553889389	5.19844	[12]
0	1.08	12.49625112	10.79676	[12]
0	1.12	21.42214479	16.79496	[12]
15	1.017	2.66404174	2.572194	[10]
15	1.035	5.615667914	5.225093	[10]
15	1.054	9.029168463	8.060544	[10]
15	1.072	12.69211713	10.85056	[10]
15	1.097	17.92940379	14.47581	[10]
15	1.1085	21.21870561	16.51141	[10]
15	1.009093	1.492858013	1.46272	[11]
15	1.019084	3.077632973	2.954406	[11]
15	1.029075	4.763083671	4.475055	[11]
15	1.039066	6.559118237	6.02467	[11]
15	1.049057	8.476989067	7.603249	[11]
15	1.059048	10.52952879	9.210794	[11]
15	1.069039	12.73143778	10.8473	[11]
15	1.07903	15.09963678	12.51278	[11]
15	1.089021	17.65370254	14.20721	[11]
15	1.099012	20.41641029	15.93062	[11]
15.6	1.032	5.553889389	5.158452	[12]
15.6	1.07	12.49625112	10.69679	[12]
15.6	1.102	21.42214479	16.52504	[12]
18	1.002842	0.243104045	0.242611	[27]
18	1.004842	0.763253805	0.755401	[27]
18	1.008841	1.271152698	1.250566	[27]
18	1.01684	2.520252328	2.439642	[27]
18	1.027838	4.199282221	3.98161	[27]
18	1.057833	9.414882012	8.380672	[27]
18	1.086829	16.19400318	13.29314	[27]
18	1.102826	21.24914756	16.69688	[27]
20	1.005	1.020102133	1.004681	[13]
20	1.012	2.082708521	2.023358	[13]

Temp (°C)	Density (g/mL)	Molality (mol/kg-H_2O)	Molarity (mol/L)	Reference
20	1.021	3.190532202	3.062029	[13]
20	1.028	4.34652213	4.110696	[13]
20	1.036	5.553889389	5.178358	[13]
20	1.043	6.816136977	6.256016	[13]
20	1.05	8.137093756	7.34767	[13]
20	1.057	9.520953238	8.453319	[13]
20	1.064	10.97231806	9.572964	[13]
20	1.07	12.49625112	10.69661	[13]
20	1.084	15.78473826	13.00388	[13]
20	1.096	19.43861286	15.33914	[13]
20	1.017	2.63078971	2.541694	[14]
20	1.035	5.553889389	5.173359	[14]
20	1.053	8.820883147	7.894996	[14]
20	1.07	12.49625112	10.69661	[14]
20	1.086	16.66166817	13.5707	[14]
20	1.101	21.42214479	16.50976	[14]
20	1.001201	0.504899035	0.500442	[15]
20	1.005194	1.020102133	1.004875	[15]
20	1.009187	1.545927974	1.5133	[15]
20	1.012181	2.082708521	2.023721	[15]
20	1.016174	2.63078971	2.53963	[15]
20	1.021165	3.190532202	3.062524	[15]
20	1.025158	3.762312167	3.586915	[15]
20	1.028152	4.34652213	4.111306	[15]
20	1.033144	4.943571873	4.647672	[15]
20	1.036138	5.553889389	5.179048	[15]
20	1.039133	6.177921904	5.713418	[15]
20	1.043126	6.816136977	6.256769	[15]
20	1.047118	7.469023661	6.804112	[15]
20	1.050113	8.137093756	7.348461	[15]
20	1.053108	8.820883147	7.895803	[15]
20	1.0571	9.520953238	8.454122	[15]
20	1.060095	10.23789249	9.007951	[15]
20	1.064088	10.97231806	9.573755	[15]
20	1.067083	11.7248776	10.13407	[15]
20	1.070077	12.49625112	10.69738	[15]
20	1.07407	13.28715309	11.27416	[15]
20	1.077065	14.0983346	11.84395	[15]
20	1.080059	14.93058576	12.41674	[15]
20	1.084052	15.78473826	13.0045	[15]
20	1.087047	16.66166817	13.58378	[15]

Temp (°C)	Density (g/mL)	Molality (mol/kg-H_2O)	Molarity (mol/L)	Reference
20	1.090041	17.56229888	14.16604	[15]
20	1.093036	18.4876044	14.75131	[15]
20	1.096031	19.43861286	15.33956	[15]
20	1.099025	20.41641029	15.93081	[15]
20	1.10202	21.42214479	16.52506	[15]
25	1.000948	0.050002594	0.05	[16]
25	1.001048	0.100095355	0.1	[16]
25	1.005815	0.249796879	0.25	[16]
25	1.002958	0.503547725	0.5	[16]
25	1.006063	0.756766967	0.75	[16]
25	1.008431	1.011710607	1	[16]
25	1.007556	1.534453708	1.5	[16]
25	1.013241	2.055015173	2	[16]
25	1.023785	3.112786885	3	[16]
25	1.039398	5.32272665	5	[16]
25	1.073432	11.4498702	10	[16]
25	1.104331	18.65113082	15	[16]
25	1.13165	27.33997548	20	[16]
25	1.159083	37.94010881	25	[16]
25	1.232083	47.47567451	30	[16]
25	1.210887	97.40736401	40	[16]
26.7	1.03	5.553889389	5.148455	[12]
26.7	1.065	12.49625112	10.64681	[12]
26.7	1.097	21.42214479	16.45006	[12]
37.8	1.025	5.553889389	5.123463	[12]
37.8	1.055	12.49625112	10.54684	[12]
37.8	1.088	21.42214479	16.31511	[12]
48.9	1.019	5.553889389	5.093472	[12]
48.9	1.045	12.49625112	10.44687	[12]
48.9	1.075	21.42214479	16.12016	[12]

APPENDIX C. DATA FOR UO_2SO_4 SOLUTIONS

Temp (°C)	Density (g/mL)	Molarity (mol U/L)	Molality (mol/kg-H_2O)	Reference
20	1.017	0.05556	0.055746	[6]
20	1.036	0.113196	0.113815	[6]
20	1.055	0.172908	0.174355	[6]
20	1.075	0.234914	0.237527	[6]
20	1.095	0.299106	0.303507	[6]
20	1.116	0.36581	0.372485	[6]
20	1.137	0.43481	0.444672	[6]
20	1.158	0.506103	0.520297	[6]
20	1.181	0.580675	0.59961	[6]
20	1.203	0.657213	0.68289	[6]
20	1.227	0.737357	0.77044	[6]
20	1.251	0.820123	0.862598	[6]
20	1.275	0.905512	0.959737	[6]
20	1.301	0.995052	1.062273	[6]
20	1.327	1.087434	1.170668	[6]
20	1.353	1.182656	1.285439	[6]
20	1.381	1.282576	1.407167	[6]
20	1.409	1.385556	1.536502	[6]
24.8	1.126166	0.470637		[28]
24.9	1.262961	0.941481		[28]
24.9	1.396087	1.410522		[28]
24.8	1.525314	1.881869		[28]
25	1.656117	2.351785		[28]
25	1.720905	2.586666		[28]
25.2	1.794098	2.821788		[28]
25	1.851797	3.057215		[28]
25	1.901969	3.29237		[28]
25	2.042403	3.763061		[28]
25	1.016	0.055505	0.055746	[6]
25	1.035	0.113087	0.113815	[6]
25	1.054	0.172744	0.174355	[6]
25	1.074	0.234696	0.237527	[6]
25	1.094	0.298833	0.303507	[6]
25	1.114	0.365155	0.372485	[6]
25	1.136	0.434427	0.444672	[6]
25	1.157	0.505666	0.520297	[6]
25	1.179	0.579691	0.59961	[6]
25	1.202	0.656667	0.68289	[6]
25	1.225	0.736155	0.77044	[6]

Temp (°C)	Density (g/mL)	Molarity (mol U/L)	Molality (mol/kg-H_2O)	Reference
25	1.249	0.818812	0.862598	[6]
25	1.273	0.904091	0.959737	[6]
25	1.298	0.992758	1.062273	[6]
25	1.324	1.084975	1.170668	[6]
25	1.35	1.180033	1.285439	[6]
25	1.377	1.278861	1.407167	[6]
25	1.405	1.381623	1.536502	[6]
25	1.0184	0.063858	0.064177	[19]
25	1.0241	0.084023	0.084587	[19]
25	1.031	0.104189	0.104939	[19]
25	1.0342	0.114692	0.115592	[19]
25	1.0378	0.125615	0.126652	[19]
25	1.0395	0.128976	0.129979	[19]
25	1.0393	0.129816	0.130893	[19]
25	1.0433	0.142	0.143244	[19]
25	1.0443	0.147461	0.148903	[19]
25	1.0461	0.150402	0.151762	[19]
25	1.0482	0.157964	0.1595	[19]
25	1.0511	0.168467	0.170267	[19]
25	1.0526	0.171408	0.173166	[19]
25	1.0576	0.186112	0.188093	[19]
25	1.0586	0.187792	0.189718	[19]
25	1.058	0.188212	0.190287	[19]
25	1.0661	0.212999	0.21556	[19]
25	1.3613	1.138517	1.205419	[19]
25	1.4746	1.493558	1.609747	[19]
25	1.5063	1.593504	1.726567	[19]
25	1.5404	1.698953	1.849849	[19]
30	1.014	0.055396	0.055746	[6]
30	1.033	0.112868	0.113815	[6]
30	1.053	0.17258	0.174355	[6]
30	1.072	0.234258	0.237527	[6]
30	1.092	0.298286	0.303507	[6]
30	1.113	0.364827	0.372485	[6]
30	1.134	0.433662	0.444672	[6]
30	1.155	0.504792	0.520297	[6]
30	1.177	0.578708	0.59961	[6]
30	1.2	0.655574	0.68289	[6]
30	1.223	0.734953	0.77044	[6]
30	1.246	0.816845	0.862598	[6]
30	1.271	0.902671	0.959737	[6]

Temp (°C)	Density (g/mL)	Molarity (mol U/L)	Molality (mol/kg-H_2O)	Reference
30	1.295	0.990463	1.062273	[6]
30	1.321	1.082517	1.170668	[6]
30	1.347	1.177411	1.285439	[6]
30	1.374	1.276075	1.407167	[6]
30	1.401	1.377689	1.536502	[6]
30	1.0312	0.105029	0.105796	[19]
30	1.0379	0.126035	0.127082	[19]
30	1.0452	0.147041	0.148321	[19]
30	1.0518	0.168047	0.169696	[19]
30	1.0585	0.189053	0.191099	[19]
30	1.0653	0.210059	0.212524	[19]
35	1.013	0.055341	0.055746	[6]
35	1.032	0.112759	0.113815	[6]
35	1.051	0.172252	0.174355	[6]
35	1.07	0.233821	0.237527	[6]
35	1.09	0.29774	0.303507	[6]
35	1.111	0.364171	0.372485	[6]
35	1.131	0.432515	0.444672	[6]
35	1.153	0.503918	0.520297	[6]
35	1.175	0.577725	0.59961	[6]
35	1.197	0.653935	0.68289	[6]
35	1.22	0.73315	0.77044	[6]
35	1.244	0.815534	0.862598	[6]
35	1.268	0.90054	0.959737	[6]
35	1.292	0.988169	1.062273	[6]
35	1.317	1.079239	1.170668	[6]
35	1.343	1.173915	1.285439	[6]
35	1.37	1.27236	1.407167	[6]
35	1.397	1.373756	1.536502	[6]
40	1.011	0.055232	0.055746	[6]
40	1.029	0.112431	0.113815	[6]
40	1.048	0.17176	0.174355	[6]
40	1.068	0.233384	0.237527	[6]
40	1.088	0.297194	0.303507	[6]
40	1.108	0.363188	0.372485	[6]
40	1.129	0.43175	0.444672	[6]
40	1.15	0.502607	0.520297	[6]
40	1.172	0.57625	0.59961	[6]
40	1.194	0.652296	0.68289	[6]
40	1.217	0.731348	0.77044	[6]

Temp (°C)	Density (g/mL)	Molarity (mol U/L)	Molality (mol/kg-H_2O)	Reference
40	1.24	0.812912	0.862598	[6]
40	1.264	0.8977	0.959737	[6]
40	1.289	0.985874	1.062273	[6]
40	1.314	1.076781	1.170668	[6]
40	1.34	1.171292	1.285439	[6]
40	1.366	1.268645	1.407167	[6]
40	1.393	1.369822	1.536502	[6]
45	1.008	0.055068	0.055746	[6]
45	1.027	0.112212	0.113815	[6]
45	1.046	0.171433	0.174355	[6]
45	1.065	0.232729	0.237527	[6]
45	1.084	0.296101	0.303507	[6]
45	1.105	0.362205	0.372485	[6]
45	1.125	0.430221	0.444672	[6]
45	1.146	0.500859	0.520297	[6]
45	1.168	0.574283	0.59961	[6]
45	1.19	0.650111	0.68289	[6]
45	1.213	0.728944	0.77044	[6]
45	1.236	0.81029	0.862598	[6]
45	1.26	0.894859	0.959737	[6]
45	1.284	0.98205	1.062273	[6]
45	1.31	1.073503	1.170668	[6]
45	1.336	1.167796	1.285439	[6]
45	1.362	1.26493	1.407167	[6]
45	1.389	1.365889	1.536502	[6]
50	1.006	0.054959	0.055746	[6]
50	1.024	0.111885	0.113815	[6]
50	1.043	0.170941	0.174355	[6]
50	1.062	0.232073	0.237527	[6]
50	1.081	0.295282	0.303507	[6]
50	1.101	0.360894	0.372485	[6]
50	1.121	0.428691	0.444672	[6]
50	1.142	0.49911	0.520297	[6]
50	1.164	0.572316	0.59961	[6]
50	1.186	0.647926	0.68289	[6]
50	1.208	0.725939	0.77044	[6]
50	1.232	0.807667	0.862598	[6]
50	1.255	0.891308	0.959737	[6]
50	1.28	0.978991	1.062273	[6]
50	1.305	1.069405	1.170668	[6]

Temp (°C)	Density (g/mL)	Molarity (mol U/L)	Molality (mol/kg-H_2O)	Reference
50	1.331	1.163426	1.285439	[6]
50	1.358	1.261215	1.407167	[6]
50	1.385	1.361955	1.536502	[6]
55	1.003	0.054795	0.055746	[6]
55	1.021	0.111557	0.113815	[6]
55	1.039	0.170285	0.174355	[6]
55	1.058	0.231199	0.237527	[6]
55	1.077	0.294189	0.303507	[6]
55	1.097	0.359582	0.372485	[6]
55	1.117	0.427161	0.444672	[6]
55	1.137	0.496925	0.520297	[6]
55	1.159	0.569858	0.59961	[6]
55	1.181	0.645194	0.68289	[6]
55	1.203	0.722934	0.77044	[6]
55	1.227	0.804389	0.862598	[6]
55	1.25	0.887757	0.959737	[6]
55	1.275	0.975167	1.062273	[6]
55	1.301	1.066127	1.170668	[6]
55	1.327	1.159929	1.285439	[6]
55	1.354	1.2575	1.407167	[6]
55	1.382	1.359005	1.536502	[6]
60	1	0.054631	0.055746	[6]
60	1.018	0.111229	0.113815	[6]
60	1.035	0.16963	0.174355	[6]
60	1.054	0.230325	0.237527	[6]
60	1.072	0.292823	0.303507	[6]
60	1.092	0.357943	0.372485	[6]
60	1.112	0.425249	0.444672	[6]
60	1.132	0.49474	0.520297	[6]
60	1.153	0.566908	0.59961	[6]
60	1.175	0.641916	0.68289	[6]
60	1.198	0.71993	0.77044	[6]
60	1.221	0.800456	0.862598	[6]
60	1.245	0.884206	0.959737	[6]
60	1.27	0.971342	1.062273	[6]
60	1.296	1.06203	1.170668	[6]
60	1.322	1.155559	1.285439	[6]
60	1.35	1.253786	1.407167	[6]
60	1.378	1.355072	1.536502	[6]

Temp (°C)	Density (g/mL)	Molarity (mol U/L)	Molality (mol/kg-H_2O)	Reference
65	0.99	0.054085	0.055746	[6]
65	1.014	0.110792	0.113815	[6]
65	1.031	0.168974	0.174355	[6]
65	1.049	0.229232	0.237527	[6]
65	1.068	0.29173	0.303507	[6]
65	1.087	0.356305	0.372485	[6]
65	1.106	0.422955	0.444672	[6]
65	1.126	0.492118	0.520297	[6]
65	1.147	0.563958	0.59961	[6]
65	1.169	0.638638	0.68289	[6]
65	1.192	0.716324	0.77044	[6]
65	1.215	0.796523	0.862598	[6]
65	1.239	0.879944	0.959737	[6]
65	1.264	0.966753	1.062273	[6]
65	1.29	1.057113	1.170668	[6]
65	1.317	1.151188	1.285439	[6]
65	1.345	1.249142	1.407167	[6]
65	1.374	1.351138	1.536502	[6]
70	0.994	0.054303	0.055746	[6]
70	1.01	0.110355	0.113815	[6]
70	1.027	0.168319	0.174355	[6]
70	1.044	0.22814	0.237527	[6]
70	1.062	0.290092	0.303507	[6]
70	1.081	0.354338	0.372485	[6]
70	1.1	0.42066	0.444672	[6]
70	1.12	0.489495	0.520297	[6]
70	1.141	0.561008	0.59961	[6]
70	1.163	0.635361	0.68289	[6]
70	1.185	0.712117	0.77044	[6]
70	1.209	0.792589	0.862598	[6]
70	1.233	0.875683	0.959737	[6]
70	1.258	0.962164	1.062273	[6]
70	1.285	1.053016	1.170668	[6]
70	1.312	1.146818	1.285439	[6]
70	1.341	1.245427	1.407167	[6]
70	1.371	1.348188	1.536502	[6]
75	0.99	0.054085	0.055746	[6]
75	1.006	0.109918	0.113815	[6]
75	1.022	0.167499	0.174355	[6]
75	1.039	0.227047	0.237527	[6]
75	1.056	0.288453	0.303507	[6]

Temp (°C)	Density (g/mL)	Molarity (mol U/L)	Molality (mol/kg-H_2O)	Reference
75	1.075	0.352371	0.372485	[6]
75	1.094	0.418366	0.444672	[6]
75	1.114	0.486873	0.520297	[6]
75	1.134	0.557566	0.59961	[6]
75	1.156	0.631536	0.68289	[6]
75	1.178	0.707911	0.77044	[6]
75	1.202	0.788	0.862598	[6]
75	1.226	0.870712	0.959737	[6]
75	1.252	0.957575	1.062273	[6]
75	1.278	1.04728	1.170668	[6]
75	1.307	1.142447	1.285439	[6]
75	1.336	1.240783	1.407167	[6]
75	1.367	1.344255	1.536502	[6]
80	0.986	0.053866	0.055746	[6]
80	1.001	0.109372	0.113815	[6]
80	1.017	0.16668	0.174355	[6]
80	1.033	0.225736	0.237527	[6]
80	1.05	0.286814	0.303507	[6]
80	1.068	0.350077	0.372485	[6]
80	1.087	0.415689	0.444672	[6]
80	1.106	0.483377	0.520297	[6]
80	1.127	0.554124	0.59961	[6]
80	1.148	0.627166	0.68289	[6]
80	1.171	0.703704	0.77044	[6]
80	1.194	0.782756	0.862598	[6]
80	1.219	0.86574	0.959737	[6]
80	1.245	0.952221	1.062273	[6]
80	1.272	1.042363	1.170668	[6]
80	1.301	1.137203	1.285439	[6]
80	1.331	1.23614	1.407167	[6]
80	1.364	1.341305	1.536502	[6]
85	0.982	0.053648	0.055746	[6]
85	0.996	0.108825	0.113815	[6]
85	1.011	0.165696	0.174355	[6]
85	1.027	0.224425	0.237527	[6]
85	1.044	0.285175	0.303507	[6]
85	1.061	0.347782	0.372485	[6]
85	1.079	0.412629	0.444672	[6]
85	1.099	0.480317	0.520297	[6]
85	1.119	0.550191	0.59961	[6]
85	1.14	0.622795	0.68289	[6]

Temp (°C)	Density (g/mL)	Molarity (mol U/L)	Molality (mol/kg-H_2O)	Reference
85	1.162	0.698296	0.77044	[6]
85	1.186	0.777511	0.862598	[6]
85	1.211	0.860059	0.959737	[6]
85	1.237	0.946103	1.062273	[6]
85	1.265	1.036627	1.170668	[6]
85	1.295	1.131958	1.285439	[6]
85	1.326	1.231496	1.407167	[6]
85	1.36	1.337371	1.536502	[6]
90	0.978	0.053429	0.055746	[6]
90	0.991	0.108279	0.113815	[6]
90	1.005	0.164713	0.174355	[6]
90	1.021	0.223114	0.237527	[6]
90	1.037	0.283263	0.303507	[6]
90	1.054	0.345488	0.372485	[6]
90	1.072	0.409952	0.444672	[6]
90	1.09	0.476384	0.520297	[6]
90	1.11	0.545765	0.59961	[6]
90	1.131	0.617879	0.68289	[6]
90	1.154	0.693488	0.77044	[6]
90	1.178	0.772266	0.862598	[6]
90	1.203	0.854377	0.959737	[6]
90	1.23	0.940749	1.062273	[6]
90	1.258	1.03089	1.170668	[6]
90	1.289	1.126713	1.285439	[6]
90	1.321	1.226852	1.407167	[6]
90	1.356	1.333438	1.536502	[6]

APPENDIX D. H_2SO_4 DATA

Temp (°C)	Density g/mL	Molarity mol/L	Molality mol/kg-H_2O	Reference
7.01	1.05953	0.9	0.933043	[21]
7.01	1.06594	1	1.040852	[21]
7.01	1.07239	1.1	1.149634	[21]
7.01	1.1286	2	2.169043	[21]
10.01	1.0586	0.9	0.933043	[21]
10.01	1.06494	1	1.040852	[21]
10.01	1.07131	1.1	1.149634	[21]
10.01	1.127	2	2.169043	[21]
13.01	1.05759	0.9	0.933043	[21]
13.01	1.06387	1	1.040852	[21]
13.01	1.07017	1.1	1.149634	[21]
13.01	1.12536	2	2.169043	[21]
16	1.05652	0.9	0.933043	[21]
16	1.06274	1	1.040852	[21]
16	1.06896	1.1	1.149634	[21]
16	1.12372	2	2.169043	[21]
19.01	1.05536	0.9	0.933043	[21]
19.01	1.06158	1	1.040852	[21]
19.01	1.06773	1.1	1.149634	[21]
19.01	1.12208	2	2.169043	[21]
22	1.05421	0.9	0.933043	[21]
22	1.06034	1	1.040852	[21]
22	1.06646	1.1	1.149634	[21]
22	1.12043	2	2.169043	[21]
25	1.05299	0.9	0.933043	[21]
25	1.05907	1	1.040852	[21]
25	1.06516	1.1	1.149634	[21]
25	1.11875	2	2.169043	[21]
0	1.035	0.527633	0.536622	[6]
0	1.071	1.091972	1.132868	[6]
0	1.109	1.696073	1.799261	[6]
0	1.148	2.340959	2.548953	[6]
0	1.188	3.028156	3.398604	[6]
0	1.23	3.762255	4.369634	[6]
10	1.034	0.527124	0.536622	[6]
10	1.069	1.089932	1.132868	[6]
10	1.106	1.691485	1.799261	[6]
10	1.144	2.332802	2.548953	[6]
10	1.183	3.015412	3.398604	[6]

Temp (°C)	Density g/mL	Molarity mol/L	Molality mol/kg-H_2O	Reference
10	1.224	3.743902	4.369634	[6]
20	1.032	0.526104	0.536622	[6]
20	1.066	1.086874	1.132868	[6]
20	1.102	1.685368	1.799261	[6]
20	1.139	2.322606	2.548953	[6]
20	1.178	3.002667	3.398604	[6]
20	1.218	3.72555	4.369634	[6]
25	1.03	0.525084	0.536622	[6]
25	1.064	1.084834	1.132868	[6]
25	1.099	1.68078	1.799261	[6]
25	1.136	2.316489	2.548953	[6]
25	1.175	2.99502	3.398604	[6]
25	1.215	3.716374	4.369634	[6]
30	1.028	0.524065	0.536622	[6]
30	1.062	1.082795	1.132868	[6]
30	1.097	1.677721	1.799261	[6]
30	1.133	2.310371	2.548953	[6]
30	1.171	2.984824	3.398604	[6]
30	1.211	3.704139	4.369634	[6]
40	1.024	0.522026	0.536622	[6]
40	1.057	1.077697	1.132868	[6]
40	1.092	1.670074	1.799261	[6]
40	1.128	2.300175	2.548953	[6]
40	1.165	2.96953	3.398604	[6]
40	1.204	3.682728	4.369634	[6]
50	1.02	0.519986	0.536622	[6]
50	1.052	1.072599	1.132868	[6]
50	1.086	1.660898	1.799261	[6]
50	1.121	2.285901	2.548953	[6]
50	1.158	2.951688	3.398604	[6]
50	1.197	3.661316	4.369634	[6]
60	1.014	0.516928	0.536622	[6]
60	1.047	1.067502	1.132868	[6]
60	1.08	1.651722	1.799261	[6]
60	1.115	2.273666	2.548953	[6]
60	1.152	2.936394	3.398604	[6]
60	1.19	3.639905	4.369634	[6]
70	1.009	0.514379	0.536622	[6]
70	1.041	1.061384	1.132868	[6]
70	1.074	1.642545	1.799261	[6]
70	1.109	2.261431	2.548953	[6]

Temp (°C)	Density g/mL	Molarity mol/L	Molality mol/kg-H_2O	Reference
70	1.145	2.918551	3.398604	[6]
70	1.183	3.618494	4.369634	[6]
80	1.003	0.51132	0.536622	[6]
80	1.034	1.054247	1.132868	[6]
80	1.068	1.633369	1.799261	[6]
80	1.102	2.247157	2.548953	[6]
80	1.139	2.903258	3.398604	[6]
80	1.177	3.600141	4.369634	[6]
90	0.996	0.507751	0.536622	[6]
90	1.028	1.04813	1.132868	[6]
90	1.061	1.622664	1.799261	[6]
90	1.096	2.234922	2.548953	[6]
90	1.132	2.885415	3.398604	[6]
90	1.17	3.57873	4.369634	[6]
100	0.989	0.504183	0.536622	[6]
100	1.021	1.040992	1.132868	[6]
100	1.055	1.613487	1.799261	[6]
100	1.089	2.220648	2.548953	[6]
100	1.126	2.870121	3.398604	[6]
100	1.164	3.560378	4.369634	[6]

APPENDIX E. EQUATIONS OF THE PITZER METHOD

For a solution, the apparent molar volume (L/mol) is defined as

$$\varphi_v = \frac{1}{n}V - \frac{n_1}{n}\overline{V}_1^0 \ , \tag{E.1}$$

where $n =$ total inventory of dissolved species (mol),
$V =$ total solution volume (L),
$n_1, \overline{V}_1^0 =$ inventory (mol) and partial molar volume at infinite dilution (L/mol) of solvent.

Throughout this study, the solvent is water, and so $\overline{V}_1^0$ is just the molar volume of pure water, which is readily available [2]. The density can be calculated from these quantities as

$$d = 10^{-3}\frac{n_1 M_1 + nM}{n_1\overline{V}_1^0 + n\varphi_v} \ , \tag{E.2}$$

where $d =$ total solution density (g/mL),
$M = \frac{1}{n}\sum_{i=2}^{N} n_i M_i =$ average molecular weight of all solutes (g/mol),
$M_i =$ molecular weight of component i (g/mol); $i = 1$ denotes water.

Note that in this definition, components refer to dissolved salts (ion pairs) and the solvent (water). The Pitzer method derives a representation of the apparent molar volume based on empirical coefficients and the inventories of all solution components:

$$\varphi_v = \sum_{i=2}^{N} x_i\overline{V}_i^0 + \frac{RT}{m}\left\{\frac{\partial f}{\partial P} + 2\sum_c\sum_a m_c m_a\left(B_{ca}^v + ZC_{ca}^v\right) + \sum_{c<c'} m_c m_{c'}\left(2\Phi_{cc'}^v + \sum_a m_a\psi_{cc'a}^v\right)\right\} \ , \tag{E.3}$$

where $x_i, \overline{V}_i^0 =$ mole fraction and partial molar volume at infinite dilution (L/mol) of components,
$m_c, m_a =$ inventories of cations and anions, respectively (molality),
$m = m_c + m_a =$ total moles in solution (molality),
$R =$ universal gas constant,
$T =$ absolute temperature (K),
$f =$ Debye–Huckel term (see below),
$Z =$ total charge in solution, calculated below,
$B_{ca}^v, C_{ca}^v, \Phi_{cc'}^v, \psi_{cc'a}^v =$ ion-interaction parameters, determined empirically as described below.

The concentrations (*molality* = mol solute per kg water, abbreviated with lower-case "*m*") should not be confused with *molarity* (mol solute per liter of solution, abbreviated with upper-case "*M*"), which is often the measured quantity. The pressure derivative of the Debye–Huckel term appearing in Eq. (E.3) has been calculated and can be found in tables [2]:

$$RT\frac{\partial f}{\partial P}. \tag{E.4}$$

The total charge from all ions in solution is calculated as:

$$Z = \frac{1}{2}\sum m_i|z_i| = \sum_{\text{cations}} m_i z_i = -\sum_{\text{anions}} m_i z_i \,, \tag{E.5}$$

where $z_i =$ charge on individual ion, listed in Table E.1.

Table E.1. Ionic charges.

Ion	Charge
UO_2^{2+}	+2
H^+	+1
F^-	-1
SO_4^{2-}	-2

Of the ion-interaction parameters, B_{ca}^{v} and C_{ca}^{v} are termed "binary" parameters because they are determined from solutions of a single salt. The C_{ca}^{v} are determined directly from regressions, but the B_{ca}^{v} have the representation:

$$B_{ca}^{v} = \beta_{ca}^{v0} + \beta_{ca}^{v1} g\left(\alpha\sqrt{I}\right) , \tag{E.6}$$

where $I = \frac{1}{2}\sum m_i z_i^2 =$ ionic strength,
$\alpha = 2 =$ an empirically determined constant,
$\beta_{ca}^{v0}, \beta_{ca}^{v1} =$ parameters to be obtained through regressions,
$g(x) =$ an empirically determined function given below.

$$g(x) = \frac{2}{x^2}\left[1 - (1+x)e^{-x}\right] . \tag{E.7}$$

For the special case of doubly charged cation and anion, Eq. (E.6) takes the special form [2]

$$B_{ca}^{v} = \beta_{ca}^{v0} + \beta_{ca}^{v1} g\left(\alpha_1\sqrt{I}\right) + \beta_{ca}^{v2} g\left(\alpha_2\sqrt{I}\right) , \tag{E.6a}$$

where $\beta_{ca}^{v2} =$ additional parameter to be regressed,
$\alpha_1 = 1.4$ and $\alpha_2 = 12$ are empirical constants.

In our present study, Eq. (E.6a) is only relevant for the UO_2SO_4 binary system because upon dissolution this results in the doubly charged ions UO_2^{2+} and SO_4^{2-}.

The last term in Eq. (E.3) involves "ternary" parameters $\Phi_{cc'}^{v}$ and $\psi_{cc'a}^{v}$, so named because they only arise when three or more components are present (the solvent H_2O being one of them). The parameters $\psi_{cc'a}^{v}$ are determined directly from regressions, whereas the terms $\Phi_{cc'}^{v}$ are defined by the function:

$$\Phi_{cc'}^{v} = \theta_{cc'}^{v} + {}^{E}\theta_{cc'}^{v}(I) , \tag{E.8}$$

where $\theta_{cc'}^{v} =$ parameter obtained through regression of systems involving the cations c and c',

${}^{E}\theta_{cc'}^{v}(I) =$ a function arising from statistical mechanics that must be calculated.

We note that Eq. (E.3) only involves ternary systems involving multiple cations c and c', together with a single anion a. The theory is applicable to multiple anions (e.g., NO_3^- and F^-) in the same solution, but this study has not involved any such systems. Hence, the appropriate terms have not been included in Eq. (E.3).

In summary, the terms listed in Table E.2 must be determined by empirical regression involving density data with known temperatures and concentrations. All other variables in Eqs. (E.1)–(E.8) can be calculated using methods or published data from open literature.

Table E.2. Parameters determined from density data.

Parameter	Symbol	Equation
Binary ion-interaction	β_{ca}^{v0}	E.6
Binary ion-interaction	β_{ca}^{v1}	E.6 or E.6a
Binary ion-interaction	β_{ca}^{v2}	E.6a
Binary ion-interaction	C_{ca}^{v}	E.3
Like-charged ion-interaction	$\theta_{cc'}^{v}$	E.8
Triple ion-interaction	$\psi_{cc'a}^{v}$	E.3
Partial molar volume at infinite dilution	$\overline{V}_i^0$	E.3