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Molten Salt Thermal Property Uncertainty Workshop Report

Chemical and Fuel Cycle Technologies Division

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1. INTRODUCTION

Operation of a molten salt fueled reactor is governed, more so than any other type of reactor, by the thermal properties of the molten salt fuel because it is in a liquid form. The composition of the fuel salt will evolve during use in a reactor, as fission products are generated and corrosion of structural materials release corrosion products into the salt. The recently released NUREG/CR-7299 states that fuel qualification for molten salt fuels will be based upon maintaining fuel salt properties within an acceptable range that results in achievement of fundamental safety functions (NUREG/CR-7299). Understanding the dependence of fuel properties on temperature and composition across the anticipated range of operating temperatures, burn-up and anticipated levels of contamination is required to qualify molten salt fuels. Measuring aspects of salt chemistry is appropriately a major thrust of the Molten Salt Reactor (MSR) campaign under the Advanced Reactor Technology program.

A lack of broadly accepted nuclear quality assurance level 1 (NQA-1) qualified property data for molten salts is an identified knowledge gap emphasized by MSR developers in an open letter to the director of the Gateway for Accelerated Innovation in Nuclear (GAIN) Program in August 2020. The MSR campaign and the Nuclear Energy Advanced Modeling and Simulation (NEAMS) campaign have collaboratively worked to address this gap in the last several years by supporting the development of a set of two centralized databases for molten salt properties, the Molten Salt Thermal Databases (MSTDB). These databases organize property information into two categories with a separate interface for each, thermochemical (MSTDB-TC) and thermophysical (MSTDB-TP) properties. Thermochemical properties include phase behavior, vapor pressure and heat capacity, while thermophysical properties include density, viscosity, thermal conductivity and diffusivity, and surface tension. The construction of the MSTDB-TP and MSTDB-TC differ. The MSTDB-TP tabulates measured property data on a variety of salt compositions of interest to MSR developers (Agca et al., 2021). In contrast, the MSTDB-TC is a database of thermodynamic models of salt compositions that have been optimized using available measurements of phase transition temperatures, enthalpies of mixing, and other relevant data (Ard et al., 2022 and Besmann et al., 2021). NEAMS supports the development of the databases while the MSR campaign supports the generation of high-quality property data to fill gaps in the databases.

Both databases incorporate published property data collected at varying levels of quality as well as data being actively generated under the MSR campaign. Frequently, measured property values for the same salt composition can vary widely, depending on the measurement method and its application. A lack of measured property data also limits the composition regions for which the MSTDB-TC can reliably predict phase behavior, as noted in the documentation associated with the latest version of the MSTDB-TC (Version 3.0). This is illustrated in an example presented during the workshop and summarized in Table 1, where phase transition temperature measurements were made of a ternary NaCl-KCl-UCl_3 composition and compared to predicted values from two different versions of the MSTDB-TC database (V1.2 and V2.0) and a simple model constructed from literature values (Rose and Thomas, 2021). The comparison between measured and predicted values highlights that the MSTDB-TC performance is not adequate for some salt compositions. This can be attributed to the absence of measured data in the specific composition region of this ternary salt, which leads to high uncertainty in predicted values. Both measuring salt properties and bridging data gaps through modeling have inherent uncertainty which should be quantified to provide confidence in system models of MSRs and in fuel qualification.

Table 1: Comparison of measured (Rose et al., 2021) and predicted phase transition temperatures for 50.9 mol % NaCl, 24.4 mol % KCl, 24.7 mol % UCl_3, delta T (measured-calculated) values listed in parentheses.

Transition	Temp. (°C)				Phase transition reaction
	measured	Rose and Thomas (2021)	MSTDB-TC V1.2	MSTDB-TC V2.0	
T1	**474 ± 2**	460 (+13)	465 (+8)	361 (+112)	K_2UCl_5 + (Na,K)Cl + UCl_3 → Liquid + (Na,K)Cl + K_2UCl_5
T2	**505 ± 2**	517 (-12)	478 (+27)	410 (+95)	Liquid + (Na,K)Cl + K_2UCl_5 → Liquid + (Na,K)Cl
T3	**587 ± 2**	559 (+28)	500 (+87)	556 (+31)	Liquid + (Na,K)Cl → Liquid
	Total absolute deviation:	53	122	238	

Sources of uncertainty in property values include uncertainty in both measurements and modeling. Sources of measurement uncertainty include uncertainty in salt composition and purity, the measurement method selected and its application, and the data analysis method. Uncertainty in modeling property values occurs because models are often optimized or interpolated over large composition ranges including regions where no measured data exist. Quantifying confidence levels for property model predictions applied to multicomponent salts based on uncertainty in the input data and uncertainty in the models used to predict property values is essential to understanding the reliability of these property values when used to design, license, and safely operate MSRs and in qualifying MSR fuels (Holcomb et al., 2021).

A workshop was held to gather MSR community experts on measuring and modeling molten salt properties to discuss how uncertainty should be quantified and reported and to identify best practices for taking the uncertainty in modeled property values into account in system model outputs.

2. Workshop Organization

A virtual workshop was held on July 25th, 2023 to address quantifying uncertainty in the thermal properties of molten salts. The workshop was supported by the DOE Office of Nuclear Energy Advanced Reactor Technology Programs Molten Salt Reactor Campaign. The workshop was organized into four sessions. Three speakers were invited to give talks in each session on the subject of each session and the talks were followed by robust discussions. The subject of the four sessions were:

Session 1: Quality assessment of measured property values

Session 2: Quantifying uncertainty in property models

Session 3: Quantifying consistency of property predictions and measured values

Session 4: Quantifying uncertainty in system models

Prior to the start of the sessions supportive statements were made by Patricia Paviet, the MSR campaign National Technical Director, and Janelle Eddins, the acting Federal Manager for the MSR campaign, stressing the importance of the workshop to the deployment of MSRs. Attendees were invited from universities, national laboratories, and industry. Appendix A contains the agenda for the workshop. Appendix B contains a list of attendees. Fifty-eight people attended the workshop. Seven national laboratories were represented including the Canadian Nuclear Laboratory, seven universities were represented and representatives from six MSR developers were in attendance. Representatives from both Department of Energy and the Nuclear Regulatory Commission were also in attendance.

3. Quality assessment of measured property values

The first session of the workshop centered around data-based quality assessment approaches for existing and new measured thermochemical and thermophysical property values and application of quality assessments and ranking in campaign-managed databases. The goal of the session was to highlight best practices in reporting uncertainty for measured property values and in assessing MSTDB entries for quality.

Recently published recommendations for quality assessment of molten salt thermal property data were presented (Rose, 2022 and Rose 2023). Argonne National Laboratory and Oak Ridge National Laboratory have been collaborating to implement a documented quality assessment process for MSTDB-TP for unary salts. An assessment of density values available for LiF was presented as an example. Table 2 summarizes the assessment across six aspects as outlined in Rose, 2022. Individual aspects were ranked either high quality (H), moderate quality (M), or insufficient quality (I). An overall ranking was applied to each data set as either high quality (A), moderate quality (B), computed in the absence of measured data (C) or insufficient for quantitative use (U).

This assessment highlighted several key issues in the reporting of legacy data. Limited information is usually provided about uncertainty in the measured values. In many cases measurement precision is reported incorrectly as uncertainty. There is no standard method for measuring the density of a molten salt. Several established methods exist for measuring density but the application of each of these methods vary. For example, a hydrostatic method based on Archimede's principle is often used, where a plummet is suspended in the salt and the mass change upon immersion is measured. This mass change can be measured in a variety of ways including spring elongation or using a balance with an underweighing hook. Each method of measuring the change in mass requires different calibration procedures to determine the uncertainty in the measured mass. Additionally, the surface tension effect on the suspension wire is not always considered by the authors.

These variations in application of the methods and the use of a variety of methods can lead to wide variation in not only the measured property value but also variation in the temperature dependence of measured property values. Raw unprocessed data is rarely reported, in some cases only a final correlation with no accompanying measured values are reported. This prevents a reviewer from verifying the measured property value or proper application of the measurement method. All five reviewed sources of LiF density measurements lacked sufficient data to verify proper application of the method and determination of density values.

Table 2: Quality assessment of publicly available density measurements of LiF molten salt

Source	Overall Rank	Method	Calibrations	Composition Analysis	Environmental Controls	Measurement Precision	Verifiability
Smirnov & Stepanov 1982	**U**	**M**-Capillary and Archimedes	**M**-calibrated with KNO_3 and KCl	**M**-No direct composition measurements – salts dried under vacuum, fused, bubbled with HF and UHP Argon	**M**-Inert atmosphere- no reported O_2 and H_2O limits Temperature to 1°C	**M**-0.05% capillary 0.04% Archimedes	**I**-No raw data provided – report correlation and precision only
Katyshev, Artemov & Desyatnik 1987	**U**	**M**-maximum bubble pressure	**H**- Manometer was calibrated to ±0.015mm and micrometer to 0.01mm	**H**- Composition confirmed by chemical analysis post-test, Reagents dried with HF and vacuum distilled	**H**- Details of Argon gas purification included Temperature to 2°C	**M**- 1%	**I**- no raw data provided, reported correlation and method of least squares for fitting
Porter & Meaker 1966	**U**	**M**- Archimedes method, mass change measured by spring elongation	**M**- spring elongation constant was predetermined to ±0.1% Density of bob measured to ±0.05%	**H**- Composition confirmed by chemical analysis post-test, Reagents vacuum dried, reported oxygen and trace metal content	**I**- conducted under helium purge No reported uncertainty in Temperature, measurements made at ascending and descending temperatures	**M**- ±0.1 %	**I**- no raw data provided, reported correlation and method of least squares for fitting

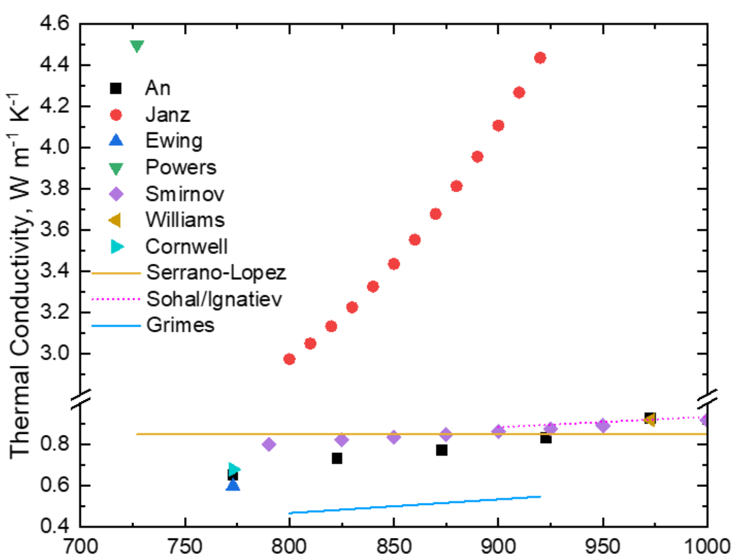
Table 2 Cont.: Quality Assessment of Publicly Available Density Measurements of LiF Molten Salt

Source	Overall Rank	Method	Calibrations	Composition Analysis	Environmental Controls	Measurement Precision	Verifiability
Brown & Porter 1964	**U***	**M**- Archimedes method, mass change measured by spring elongation	**H**- calibrated with KCl within an average deviation of ±0.4 %	**M**- trace metal and oxygen content were reported pre-test, salts were vacuum dried	**H**- Conducted under helium purge Temperature to ±1 °C	**H**- replicate measurements at each temperature in both ascending and descending temperatures, average deviation of ±0.4 %	**I**- no raw data provided, reported correlation and method of least squares for fitting
Mellors & Senderoff 1964	**U**	**M**- Archimedes method, assumed effect of surface tension is negligible	**I**- no calibrations or uncertainty information reported	**I**- no composition analyses reported, no purification of salts reported	**H**- Conducted under a purified argon purge Temperature to ±2 °C	**I**- no precision of measurements reported	**I**- no raw data provided, no correlations provided, only plots provided.

The assessment found no data sets of sufficient documentation for quantitative use for the density of LiF. However, the best choice data set was selected for use based on the six aspect assessments while higher quality data is sought. The evaluation of measured LiF density values from different sources highlights the need for a transparent, thorough, and documented quality assessment of all data entered into the MSTDB-TP. It also highlights a need for more thorough reporting of measurements, including raw unprocessed data, and determinations of uncertainty.

Criteria for accepting data for incorporation into the MSTDB was discussed during this session. All new data accepted into MSTDB must be publicly available to enable robust assessment of data quality.

An in-depth discussion of the uncertainties associated with specific thermal property measurement methods for molten salts highlighted the need to standardize methods of measurement for molten salt properties. For example, three established methods for measuring density of molten salts exist, a hydrostatic method based on Archimedes principle, a maximum bubble pressure method, and dilatometry. Each method has different sources of uncertainty that must be understood and quantified. In the case of the hydrostatic method, the effect of surface tension on the supporting wire must be quantified and any degradation of the suspended plummet will increase uncertainty. In the case of maximum bubble pressure measurements, the uncertainty in the measurement of immersion depth and the effects of surface tension will increase uncertainty in the measurement. In the case of dilatometry measurements, unusual meniscus effects and the formation of void bubbles will increase uncertainty in the measurement.



Temperature, K

Figure 1: Available thermal conductivity measurements of eutectic LiF-NaF-KF (An et al. 2015, Janz et al. 1981, Ewing et al. 1962, Powers et al. 1963, Smirnov et al. 1987, Williams et al. 2006, Cornwell, 1971, Serrano-Lopez et al. 2013, Ignatiev et al. 2002, Grimes et al. 1960)

Figure 1 is a summary of available thermal conductivity values for eutectic LiF-NaF-KF (FLiNaK) and demonstrates the variation observed in available measured property values. Published values in Figure 1 range over a full order of magnitude and have varying temperature dependencies. An assessment of the quality of each data set across the six aspects of measurement method, salt composition, calibrations, environmental control, measurement precision and verifiability, will be required to discern which values can be used quantitatively.

A thorough analysis of sources of uncertainty in each measurement method and consistent application of the method are required to generate high-quality property data. This is best achieved through the use of a standardized measurement method published through a standards body such as American Society for Testing and Materials (ASTM). Standard methods exist for measuring some thermal properties but require adaptations for application to molten salts. **A set of standard methods for measuring the thermal properties of molten salts will enable the generation of high quality property data needed to qualify molten salt fuel and to design, license, and safely operate MSRs.** Argonne National Laboratory has begun this effort by proposing a new ASTM standard for measuring viscosity of molten salts using a rotational viscometer at the ASTM International Meeting in June 2023.

Several best practices in reporting uncertainty of measurements were discussed during this session. First, that uncertainty in analytical measurements of salt composition by spectroscopy and mass spectrometry (typically 10%) is inherently much greater than the uncertainty in batching molten salts which is limited to the uncertainty of the balance used (Rose et al., 2021). Therefore, the uncertainty in batched salt compositions should be reported. Second, the standard deviation for replicate measurements or measurements of replicate samples often reported is not a measure of accuracy but of precision or repeatability. In order to determine the accuracy of a measurement method, one must measure the properties of a reference material of known properties in the same range as the material being investigated. A reference salt ideally would be a pure material rather than a mixture, with melting behavior that is similar to that of a variety of other molten salts. **The identification and production of a standard reference salt will enable researchers to quantify the accuracy of property measurement methods.**

On-going work in applying quality assessments to MSTDB-TP was summarized during this session. An assessment of the 448 systems currently included in the database is ongoing. In the absence of this assessment maximum values for uncertainty are assigned to data entries by experts with experience with measurement methods. Figure 2 shows density data entries for pure KCl with these assigned uncertainty bands (Birri, 2023). The measurement method, author and year published are given for each data set in the legend. A complete quality assessment of these data entries will eliminate suspect data sets and may reduce the size of the uncertainty bands providing confidence in values for the density of KCl as a function of temperature.

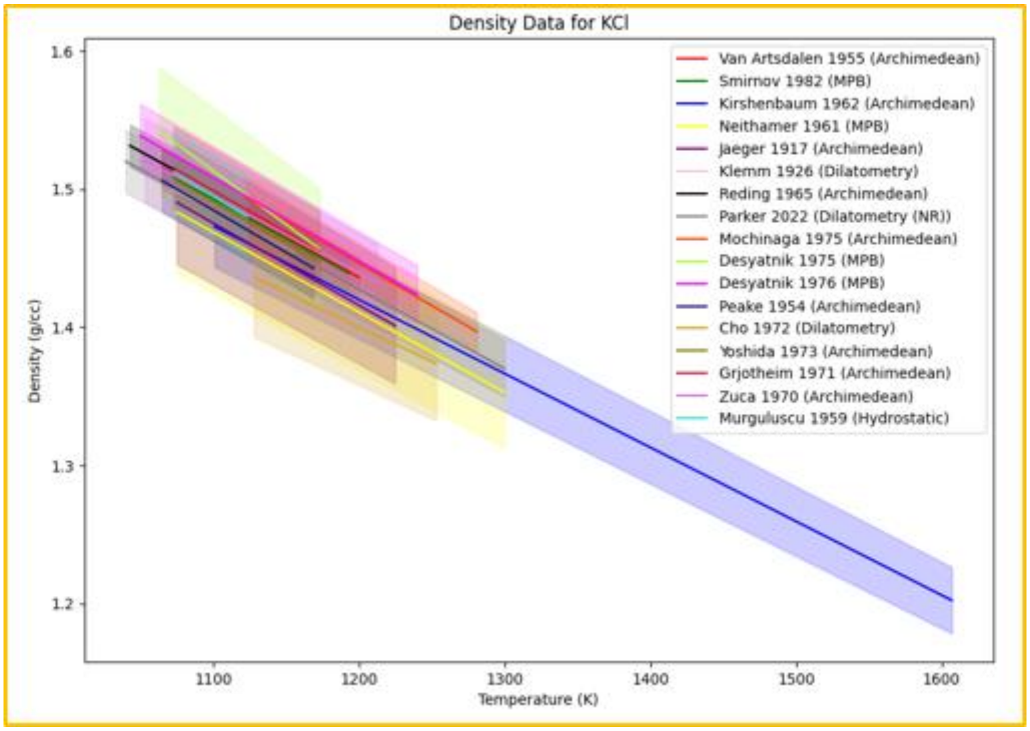


Figure 2: Density of KCl from MSTDB-TP with uncertainty bands assigned at maximum values by experts with experience with measurement methods (Image Courtesy of Oak Ridge National Laboratory).

The quality assessment performed for data included in the MSTDB-TC differs from the assessment performed for MSTDB-TP. All available data are evaluated for measurement errors, but are not included in MSTDB-TC. Only a final model of salt behavior is recorded into the database with a discussion of the assessment with a citation. The full assessment and all considered data is therefore not available to users of the database within the database. Assessments are being published in the open literature to aid users of the database (Ard, 2022). The MSTDB-TC is a modeling tool rather than a compilation of measured values and therefore it would not be possible in its current form to include more than one model or to tabulate measured values. In the quality assessments for MSTDB-TC the source of the salt measured and the uncertainty assumptions are considered. Values obtained using established methods, such as drop-calorimetry for heat capacity measurements are given higher priority. Some weighting of data sets is done in the assessments and a model is entered into the database which captures the behavior of the system (Ard, 2022). Tabulated values and assessments of their quality should be made accessible to users of the database within the database to enhance confidence in the reported model values.

MSTDB-TC relies on published values of invariant compositions and temperatures for unary, and binary systems to model higher order systems in the absence of measured values for ternary and higher order systems. A recent analysis of measurements of the ternary LiF-NaF-UF_4 system revealed that many published data sets do not measure the invariant compositions directly but extrapolate the invariant compositions and temperatures from measurements of other compositions in the ternary system (Benes, 2010; Capelli, 2014 and Thoma, 1959). Without measurements of the actual eutectic compositions these invariant compositions and temperatures must be considered unverified. These interpolations are not historically reported with a quantification of uncertainty. In many cases the measured compositions are either not reported or are reported without uncertainty. This makes evaluation of the available thermochemical data challenging.

The main findings of this session are that a significant amount of high quality data is needed to fill gaps in the available data and replace data of insufficient quality to improve the utility of the MSTDB. **Quality assessment processes should be applied to data in both MSTDB-TC and -TP.** These quality assessments should be transparent and documented in the databases, as described in ANL/CFCT-22/26 and ANL/CFCT-23/14. To improve the quality of measured property values standard measurement methods should be developed through an international standards body such as ASTM. To support the development of these standard methods, a reference salt should be selected, produced and supplied to all researchers developing measurement methods to determine the accuracy achievable with the measurement methods being developed.

4. Quantifying uncertainty in property models

A lack of measured values for all compositions of a multicomponent diagram can result in unreliable predictions in those regions of the phase diagram less well characterized (Paz Soldan Palma et al., 2023). Figure 3 illustrates this concept with a generic A-B-C ternary system, where data is available for binary compositions A-B, B-C and A-C (edges of the triangle) and two pseudo-binary compositions at low concentrations of C (black dashed lines). In this case confidence in predictions of properties in the region of high concentrations of C (grey box) would be low. This highlights a need for uncertainty quantification to inform users of the database that the compositions they are using are either in well measured regions of composition or in regions where data is lacking and confidence is low. The goal of the second session of the workshop was to identify approaches to uncertainty quantification for models of molten salt properties.

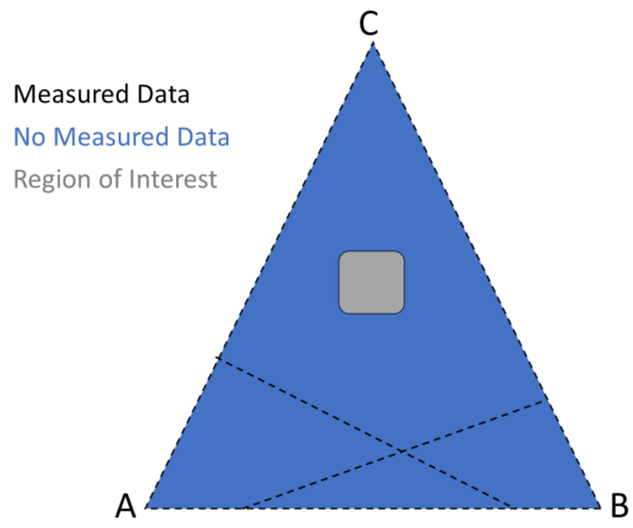


Figure 3. A generic A-B-C ternary phase diagram. Data is available for binary compositions A-B, B-C and A-C (edges of the triangle) and two pseudo-binary compositions at low concentrations of C (black dashed lines). The region of interest is marked as a grey box (Adapted from Paz Soldan Palma et al., 2023).

During the second session of the workshop the uncertainty inherent to modeling thermochemical properties of molten salts included in the MSTDB-TC was discussed. The MSTDB-TC is not a database by the strict definition that a database is a tabulation of measured values with sources. MSTDB-TC is a collection of models calculated from pooled interpolated invariant compositions and temperatures drawn from available literature. It does not contain sets of measured values with defined uncertainty. This makes assigning uncertainty to its predictions of molten salt thermochemical properties challenging. Nevertheless, MSR developers are using the predicted values from MSTDB-TC in making design decisions for their reactors. It is therefore necessary to quantify for developers the uncertainty in these predicted values.

Approaches to quantifying uncertainty for model predictions based on uncertainty inherent in the modeling were discussed. Incorporation of Bayesian statistics was recommended to determine uncertainty in model parameters. Uncertainty in the parameters (probability curve) can then be used to propagate uncertainty into properties of interest. The CALculation of PHAse Diagrams (CALPHAD) approach used in MSTDB-TC can be improved by leveraging Bayesian automated weighting and inference tools. Figure 4 highlights how this enhances the use of expert intuition and results in a predicted value with associated uncertainty.

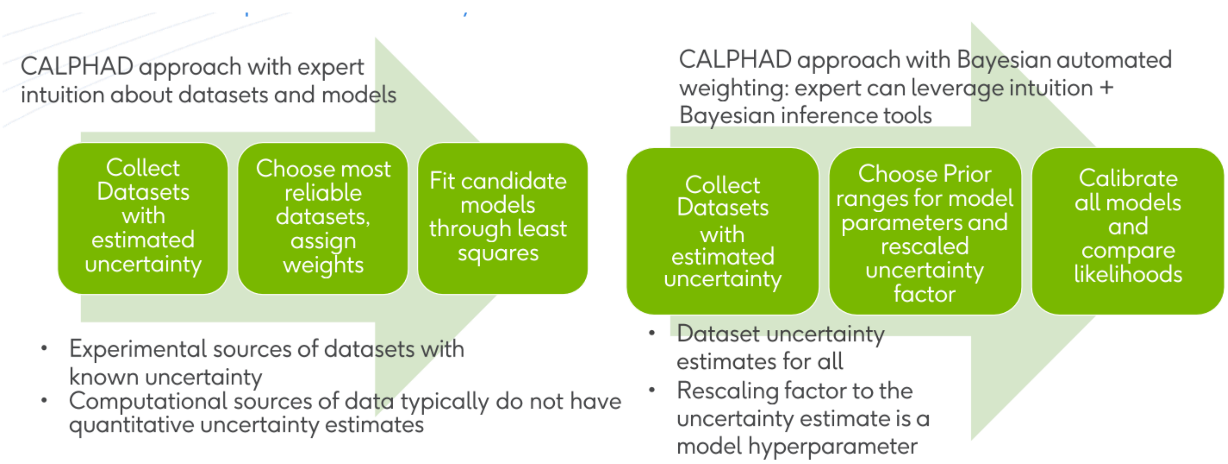


Figure 4: Schematic comparing the CALPHAD approach with and without Bayesian statistics (Figure taken from J. Gabriel, M. Poschmann presentation on "Towards Uncertainty Quantification of Molten Salt Property Models- inspirations from Bayesian thermodynamic modeling using diverse dataset sources")

Several examples of the success of this method applied to metallic materials were presented and its ability to assess both measured and computationally produced values. Assigning uncertainty to individual model parameters which may be used in multiple calculations in the database, particularly in higher order systems which rely on unary and binary data, allows uncertainty propagation to be done.

Multiple presenters recommended that Bayesian statistics be used to perform uncertainty quantification and propagation for thermochemical models of molten salts. Markov Chain Monte Carlo method was recommended to analytically solve for the probability term in Bayes theorem. The usual model for determining properties of higher order salt systems from data for unary and binary salt systems is a modified quasi-chemical model in the quadruplet approximation (MQMQA). Three interfaces are available which have capabilities to perform these calculations, FactSage, Thermochimica and PyCalphad. PyCalphad has a tool for uncertainty quantification which utilizes Markov Chain Monte Carlo method called Extensible Self-optimizing Phase Equilibria Infrastructure (ESPEI). Developers of MSTDB-TC and collaborators are currently exploring using this tool to perform uncertainty quantification and propagation for the thermodynamic models in MSTDB-TC.

Another strategy for predicting molten salt properties is to use ab initio molecular dynamics simulations of properties of molten salts. Ab initio molecular dynamics simulations for molten salts use input parameters from atomic scale calculations and the change in free energy as the driving force for reactions to simulate several isothermal behaviors which are stitched together to yield the temperature dependent molten salt properties. Sources of uncertainty in this type of modeling include approximations made in applying density functional theory, such as exchange-correlation approximations, and molecular dynamics parameters such as time steps, thermostats, cell size and simulation time. Bayesian statistics are again recommended to quantify uncertainty in this type of modeling and simulation to assign uncertainty to individual parameters used in the modeling and to propagate that uncertainty to the predicted property value.

Bayesian statistics can be used to identify data sets that are needed to reduce uncertainty. Discussion during this session concluded that more **regular interaction between modelers and those measuring properties of molten salts are needed to communicate identified needs for specific data**. Measuring properties of compositions that will reduce the overall uncertainty of modeling a salt system would be an efficient use of resources. Suggestions for increasing these interactions included regular scheduled teleconferences, GitLab forums and email blasts.

The main findings of this session were that Bayesian statistics can be used to quantify uncertainty in model parameters and these uncertainties can then be propagated to determine an uncertainty in predicted molten salt properties, whether the modeling is done by AIMD, or the CALPHAD method. Recently developed tools for doing this uncertainty propagation were identified. The Bayesian method for propagating uncertainty in predicted values can also identify data needed to reduce uncertainty and it was recommended that modelers and those measuring properties have regular communication to convey these data needs to make the best use of the resources available.

5. Quantifying consistency of property predictions and measured values

The goal of the third session of the workshop was to identify approaches for determining consistency of property predictions with measured values which consider both uncertainty in the model as discussed in the second session and uncertainty in the measured values as discussed in the first session. Presenters in this session were asked to outline approaches for representing consistency. As an example, Table 1 in section 1 of this report used an absolute difference between the predicted and measured value to represent consistency of the models with measured values. Ted Besmann of University of South Carolina shared a similar approach at the workshop where he calculated an average absolute difference across multiple data points from published literature to compare a simple model constructed from only unary and binary data (Rose and Thomas, 2021) to the predictive capability of MSTDB-TC V2.0. Table 3 shows the measured liquidus and eutectic compositions and temperatures, the predictions of transition temperatures by each model, and the absolute differences between each model and the measured values from Ted Besmann's analysis of the ternary NaCl-KCl-UCl_3 system. Table 4 summarizes these comparisons with average absolute differences across each data set and model. This approach is able to summarize the performance of models across the entire measured compositional space of the ternary system. This initial analysis only considered the liquidus and eutectic transitions while it is known that this system has several lower temperature transitions. Expanding this analysis to include lower temperature transitions would be valuable.

Table 3: Comparison of measured and computed liquidus and eutectic temperatures from two models for the ternary NaCl-KCl- UCl_3 system. Measured values taken from two sources, Desyatnik et al. and Yingling et al.

			Measured Transition Temperature, °C		**Computed Transition Temperatures, °C**				**Absolute Difference Between Computed and Measured Transition Temperatures**			
Mole Fraction			**Desyatnik et al.**		**Simple Model**		**MSTDB-TC V 2.0**		**Simple Model**		**MSTDB-TC V 2.0**	
NaCl	**KCl**	**UCl_3**	**Liq.**	**Eut.**	**Liq.**	**Eut.**	**Liq.**	**Eut.**	**Liq.**	**Eut.**	**Liq.**	**Eut.**
0.096	0.606	0.298	581		613	547	585	521	32		4	
0.090	0.61	0.300		506	613	547	586	521		41		15
0.188	0.544	0.268		530	593	548	553	521		18		9
0.193	0.541	0.266			592	548	551	521				
0.290	0.476	0.234	553		569	548	536	521	16		17	
0.285	0.479	0.236		504	570	548	532	521		44		17
0.390	0.409	0.201		502	560	547	605	521		45		19
0.389	0.409	0.202	605		559	547	605	521	46		1	
0.484	0.346	0.170		509	608	547	647	520		38		11
0.589	0.275	0.136		513	655	547	683	519		34		6
0.695	0.204	0.101		515	698	547	716	516		32		1
0.794	0.138	0.068		511	734	545	746	512		34		1
0.493	0.340	0.167	662		612	547	651	520	50		11	
0.591	0.274	0.135	692		656	547	683	519	36		9	
0.697	0.203	0.100	733		699	547	716	516	34		17	
0.911	0.060	0.029		503	773	539	778	498		36		5
0.806	0.130	0.064	753		729	545	749	511	24		4	
0.922	0.052	0.026	791		776	538	781	495	15		10	
			Yingling et al.		**Simple Model**		**MSTDB-TC V 2.0**		**Simple Model**		**MSTDB-TC V 2.0**	
			Liq.	**Eut.**	**Liq.**	**Eut.**	**Liq.**	**Eut.**	**Liq.**	**Eut.**	**Liq.**	**Eut.**
0.475	0.475	0.050	646		636	528	640	521	10		6	
0.437	0.437	0.126	613	514	610	536	625	521	3	22	12	7
0.148	0.566	0.286	578	515	601	547	568	521	23	32	10	6
0.285	0.478	0.237	532		570	547	531	521	38		1	
0.546	0.302	0.152	655		636	547	669	519	19		14	

Table 4: Average absolute deviation of computed liquidus and eutectic temperatures from measured values from two sources for the NaCl-KCl-UCl_3 system

Data Source	# of data points	Simple Model		MSTDB-TC V 2.0	
		Liquidus	Eutectic	Liquidus	Eutectic
Desyatnik et al.	34	31.4	36.0	9.0	9.4
Yingling et al.	14	18.7	27.2	8.8	6.3

This analysis, when compared with the analysis shown in Table 1, highlights that a model may predict the system performance well for compositions for which measured data exist and still fail to predict well for compositions where no data exists. This emphasizes the need for more measurements of the properties of molten salts, particularly for compositions of interest to reactor developers. This approach does not account for uncertainty in either the measured values or the modeled values.

A Bayes factor can be used to compare different models against the same measured data to determine which performs better. The Bayes factor, K, is a ratio of the probability, Pr, of an outcome from a model, M, given a certain set of input data, D, as in Equation 1 (Gong, 2023). Bayes factors above 10 provide strong evidence to choose model 1 over model 2 as described in Table 5.

$$K = \frac{\Pr(D|M_1)}{Pr(D|M_2)} \quad (1)$$

Table 5: Bayes Factor Indications (adapted from Gong, 2023).

Bayes Factor, K	Strength of Evidence
1 to 3.2	Insignificant
3.2 to 10	Substantial
10 to 100	Strong
>100	Decisive

A dashboard testing capability for MSTDB-TC has been developed to enhance stakeholder confidence in the predictive abilities of the database. Comprehensiveness of the database across both fluoride and chloride systems were evaluated using this dashboard as high, medium-high, medium, medium-low or low (Piro and Poschmann, 2023). It was stressed that the application of this assessment to MSTDB-TC is an on-going process which will require consistent support.

Two options for applying uncertainty quantification to molten salt properties were outlined during this session, first a conventional approach wherein uncertainty is quantified for a specific application such as in the MSTDB-TC or in severe accident codes. This approach is less general and requires expert judgement by the user. The second option is to propagate uncertainty from measurements and modeling up through thermodynamic codes to system codes. This should lead to more meaningful quantifications of uncertainty but will require an enormous amount of work to the database and associated software packages to accommodate the propagation calculations. This approach may also lead to very high uncertainty in cases where data is missing.

A novel method for measuring thermal conductivity was presented to demonstrate best practices in uncertainty quantification applied to the resultant model for the property value. When fitting a model to the measured data a goodness of fit error was calculated to determine the consistency of the model with measured values. Measurements of a standard reference material can be used to quantify the true uncertainty of measured values. Variance in measurements should be considered in the uncertainty as well. Propagation of uncertainty for each parameter in the model fit to the data was conducted to yield an overall uncertainty for the model used to represent the measured values.

A model of a higher order system may predict salt behavior well for compositions for which measured data exist and still fail to predict well for compositions where no data exists. **It will therefore be necessary for MSR developers to measure the properties of their fuel and/or coolant salt compositions under a quality assurance program to produce high-quality property values suitable for use in licensing their reactor and qualifying their fuel.** A Bayes factor can be used when deciding between different models for representing measured data, and uncertainty should be propagated from thermodynamic models through to system models, though it will require an enormous amount of work on the database and software packages to do so. Missing data is likely to cause very high uncertainty in thermodynamic property models and therefore effort should be made to fill existing data gaps.

6. Quantifying uncertainty in system models

System performance models for MSRs will be relied upon during design, licensing and operation of deployed reactors. The reliability of these models must be quantified for developers and regulators to have confidence in the model predictions. Presenters during the fourth session were invited to describe approaches for quantifying uncertainty in system models which accounted for both uncertainty in property models and the data upon which they were based.

Recent work applying MELCOR to study accident scenarios and perform uncertainty analysis was summarized. It was suggested that uncertainty in properties may be swamped by the uncertainty inherent to phenomenological or event scenario modeling. Sensitivity analyses provide insight into which properties of molten salts are likely to have a significant effect on reactor safety and performance. Dymola, Modelica and Transient Simulation Framework of Reconfigurable Modules (TRANSFORM) tools were used to study the effect of variation in thermal properties of molten salts on temperatures at select locations in a generic MSR model (Creaseman, 2022). The main finding was that variation in heat capacity produced the largest effect on output temperatures. It was concluded that all effects seen when varying properties up to 30% of their value were not safety significant. However, developers present during the session added the perspective that though these variations in properties were not safety significant they would strongly affect the design and economics of reactors.

The uncertainty in regions of the phase diagrams without adequate data to map the phase space, and insufficient resources to conduct an extensive measurement program to characterize all molten salts of interest, will require developers to measure the properties of their chosen molten salt system across the compositional range anticipated during use.

Modeling of a solid-fueled salt cooled high temperature reactor and how uncertainty in property values for molten salt might impact this type of reactor were discussed. While temperatures in the reactor might not change dramatically with variation in molten salt properties, the flow conditions will. An observation echoed by developers present at the workshop. A change in viscosity, heat capacity or density will result in a change in pressure drop across the loop and resultant velocity of the salt. These changes can be accommodated by changing pump power to ensure a constant flow rate to actively manage flow conditions. This will require reactor developers to design in additional capacity in order to accommodate such changes in real-time, resulting in increased costs.

Uncertainty in molten salt properties can be propagated to system performance models. In the case of phenomenological accident scenario modeling this uncertainty in property values may be swamped by uncertainty inherent to phenomenological modeling. Despite this, developers expressed a strong interest in understanding how uncertainty in bulk properties such as viscosity, density and heat capacity will impact uncertainty in flow conditions, as this will have an impact on design decisions.

7. Conclusions

In order to design, license and safely operate an MSR and qualify MSR fuels, the properties of the molten salt fuel must be well understood across the temperature and composition range anticipated during normal and transient reactor conditions. The MSR and NEAMS campaigns have been collaborating to close gaps in the understanding of molten salt properties by developing the MSTDB-TC and -TP databases. Work is ongoing to incorporate transparent, thorough, and objective quality assessments into the databases. As part of this process, evaluating the uncertainty of measured and modeled property values became a focus for researchers under the MSR campaign. This workshop was held to bring together experts in the MSR community to discuss best practices in quantifying and reporting uncertainty in measured and modeled property values and to identify best practices for taking the uncertainty in property values into account in system model outputs.

The discussions across the four sessions yielded several recommendations:

- Continue to apply transparent, thorough, and documented quality assessment processes to data in both MSTDB-TC and -TP.
- Quantify the uncertainty in model predictions where data gaps must be bridged by modeling; Bayesian statistics were recommended.
- Develop a set of standard methods for measuring the thermal properties of molten salts to enable the generation of high quality property data.
- Identify and produce a standard reference salt to enable researchers to quantify the accuracy of property measurement methods and cross-compare work from different labs and using different methods.
- Promote regular interaction between modelers and those measuring properties of molten salts to communicate identified needs for specific data.

Developers will require a quantitative assessment of the reliability of the property models being used in their system level models. Property models of multi-component salts will always have increased uncertainty due to necessary extrapolation from binary and unary data. Models for higher order systems used to compute values in regions where no data is available for use in fitting model parameters, may provide inaccurate phase behavior. It will therefore be necessary for MSR developers to measure the properties of their fuel and/or coolant salt compositions under a quality assurance program to produce high-quality property values suitable for use in licensing their reactor and qualifying their fuel. The development of standard measurement methods and standard reference salts for determining the accuracy of methods will enable the generation of these high-quality measurements. Developers will need access to world-class property measurement facilities operated under a quality assurance program using standard measurement methods to verify model predictions for thermal properties of their complex fuel salts at beginning and end of life conditions.

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Appendix A: Workshop Agenda

Agenda for Workshop on Uncertainty in Molten Salt Thermal Property Values and Predictions

July 25th, 2023

All times in Central Standard Time

8:30 – 8:40 am Attendees Log in

8:40 – 8:45 am Opening Remarks by Patricia Paviet

8:45 – 8:50 am Statement of Support from Janelle Eddins

8:50 – 9:00 am Goal of the Workshop

9:00 – 10:20 am Session 1: Quality assessment of measured property values

9:00 - 9:15 am Melissa Rose (ANL) – Quality Assessment Process for Property Data

9:15 - 9:30 am Tony Birri (ORNL) – MSTDB-TP: Dealing with Uncertainty

9:30 – 9:45 am Juliano Schorne Pinto (University of South Carolina) – Assessing Phase Equilibria and Thermochemical Properties for MSTDB-TC

9:45 - 10:20 am Discussion

10:20 – 10:30 am Break

10:30 – 11:45 am Session 2: Quantifying uncertainty in property models

10:30 – 10:45 am Joshua Gabriel (CNL) – Towards Uncertainty Quantification of Molten Salt Property Models: Inspirations from Bayesian Thermodynamic Modeling Using Diverse Dataset Sources

10:45 – 11:00 am Jorge Pas Soldan Palma (University of South Carolina) – Quantifying Uncertainties in Thermochemical Properties and Resultant Computed Equilibrium States

11:00 -11:15 am David Andersson (LANL) – Uncertainty Quantification for Ab Initio Molecular Dynamics (AIMD) Simulations of Molten Salt Properties

11:15 – 11:45 am Discussion

11:45 – 12:30 pm Lunch Break

12:30 – 1:45 pm Session 3: Quantifying consistency of property predictions and measured values

12:30 – 12:45 pm Rushi Gong (Penn. State University/ ANL) – Thermodynamic Modeling and Model Selection for LiF-LnF3 Molten Salts with Uncertainty Propagation

12:45 – 1:00 pm Markus Piro (Ontario Tech University) – Uncertainty Associated with Thermodynamic Calculations of Molten Salts

1:00 – 1:15 pm Troy Munro (Brigham Young University) – Quantifying Uncertainty in Molten Salt Thermal conductivity Measurements

1:15 – 1:50 pm Discussion

1:50 – 2:00 pm Break

2:00 – 3:30 pm Session 4: Quantifying uncertainty in system models

- 2:00 – 2:15 pm David Luxat (SNL) -
- 2:15 – 2:30 pm Sarah Creaseman (INL) – Sensitivity Analysis of Thermophysical Properties of Molten Salts Using a MSDR Model in TRANSFORM
- 2:30 – 2:45 pm Max Fratoni (Berkeley) -
- 2:45 – 3:20 pm Discussion

3:20 – 3:50 pm Wrap-up Discussion

3:50 – 4:00 pm Closing Remarks

Appendix B: List of Attendees

Attendee List

Name	Affiliated Institution
James Johnstone	Unknown
Amber L. Polke	Argonne National Laboratory
Kyle Chamberlain	Argonne National Laboratory
Levi Gardner	Argonne National Laboratory
Melissa A Rose*	Argonne National Laboratory
Nora Shaheen	Argonne National Laboratory
Sara A. Thomas	Argonne National Laboratory
Sarah Stariha	Argonne National Laboratory
Shayan Shahbazi	Argonne National Laboratory
Timothy Lichtenstein	Argonne National Laboratory
William L. Ebert	Argonne National Laboratory
Troy Munro*	Brigham Young University
Joshua Gabriel* (CNL)	Canadian Nuclear Laboratory
Brian Robinson	Department of Energy
James Willit	Department of Energy
Janelle L (Zamore) Eddins	Department of Energy
Rodolfo Vaghetto	Electric Power Research Institute
Viola Aureggi	Exodys Energy Inc.
Carl Perez	Exodys Energy, Inc.
Kurt Harris	Flibe Energy Inc.
David E. Holcomb	Idaho National Laboratory
Jacob A. Yingling	Idaho National Laboratory
James A. King	Idaho National Laboratory
Mauricio E. Tano Retamales	Idaho National Laboratory
Samuel A. Walker	Idaho National Laboratory
Sarah E. Creasman*	Idaho National Laboratory
Toni Y. Karlsson	Idaho National Laboratory
William C. Phillips	Idaho National Laboratory
Ryan Gallagher	KAIROS
Anders David Ragnar Andersson*	Los Alamos National Laboratory
Aditya Savara	Nuclear Regulatory Commission
Alexander Chereskin	Nuclear Regulatory Commission
Steve Bajorek	Nuclear Regulatory Commission
Wendy Reed	Nuclear Regulatory Commission
Tony Birri*	Oak Ridge National Laboratory
Markus Piro* (Ontario Tech)	Ontario Tech
Kyle A Makovsky	Pacific Northwest National Laboratory
Manh Thuong Nguyen	Pacific Northwest National Laboratory
Michaella S. Swinhart	Pacific Northwest National Laboratory
Patricia D. Paviet*	Pacific Northwest National Laboratory, NTD
Rushi Gong*	Pennsylvania State University
David Lyle Luxat*	Sandia National Laboratory
Matthew Scott Christian	Sandia National Laboratory

Attendee List Continued

Name	Affiliated Institution
Brandon Wilson	Terrapower
Hilary Fitzgerald	Terrapower
SeungMin Lee	Terrapower
Daniel Carleton	Terrestrial Energy USA
Max Fratoni*	University of California Berkeley
Randall Chiu	University of California Berkeley
Adam Burak	University of Michigan
Soumya Sridar	University of Pittsburgh
Amir Mofrad	University of South Carolina
Jack Wilson	University of South Carolina
Jorge Paz Soldan Palma*	University of South Carolina
Juliano Schorne Pinto*	University of South Carolina
Mina Aziziha	University of South Carolina
Ronnie Booth	University of South Carolina
Theodore Besmann	University of South Carolina

***speaker**

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