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An Overview of the Molten Salt Thermal Properties Database–Thermophysical, Version 2.1.1 (MSTDB-TP v.2.1.1)

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July 2023

OAK RIDGE National Laboratory

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US Department of Energy Office of Nuclear Energy Nuclear Energy Advanced Modeling and Simulation Program

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ABBREVIATIONS

API	application programming interface
CALPHAD	Computer Coupling of Phase Diagrams and Thermochemistry
GUI	graphical user interface
MSTDB	Molten Salt Thermal Properties Database
MSTDB-TC	Molten Salt Thermal Properties Database–Thermochemical
MSTDB-TP	Molten Salt Thermal Properties Database–Thermophysical
ORNL	Oak Ridge National Laboratory
TC	thermochemical
TP	thermophysical
RK	Redlich-Kister

ACKNOWLEDGMENTS

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ABSTRACT

This report presents the current status of the Molten Salt Thermal Properties Database–Thermophysical (MSTDB-TP). Building off of MSTDB-TP and through the direction of the *Roadmap for Thermal Property Measurement of Molten Salt Reactor Systems* [1], MSTDB-TP v2.1.1 has now been released, containing a total 448 salt entries (data from 140+ independent studies), an increase compared with the original commit of 62 [2, 3], and containing thermophysical properties, including density, viscosity, thermal conductivity, and heat capacity. Along with the new database release, the further advancement of Saline, which is a C++ application programming interface, and a graphical user interface have facilitated increased user/developer interaction with the database. Furthermore, estimation techniques, first principles calculations (ab initio), and interpolation/extrapolation methods (Redlich-Kister/Muggianu) have shown great promise in the future of thermophysical property determination for filling out compositional spaces and investigating experimentally difficult salts (hazardous and/or expensive). This report describes the database composition, the development and advancement of database tools, and the strategy of advancing and implementing estimation data into future iterations of the database for MSTDB-TP.

1. INTRODUCTION

The Molten Salt Thermal Properties Database (MSTDB) is an collection of thermal properties of molten salts relevant to the nuclear industry. Fundamental thermodynamic salt properties are pivotal in facilitating the development of multiscale and multiphysics reactor models for molten salt reactors. MSTDB is split into two arms—thermochemical (TC) and thermophysical (TP)—because the data held in each category are processed in fundamentally different ways. The Molten Salt Thermal Properties Database–Thermochemical (MSTDB-TC) focuses on obtaining or generating Gibbs energy models through the Computer Coupling of Phase Diagrams and Thermochemistry (CALPHAD) method [2]. This method requires a Gibbs energy minimizer—either FactSage (licensed) or Thermochimica (open-source)—to use the data in the database. Thermodynamic data for a given composition of elements can be determined using phase equilibria, thermodynamics of phase transitions, mixing energies, and heat capacities [4]. The Molten Salt Thermal Properties Database–Thermophysical (MSTDB-TP) focuses on TP property reporting by representing the data through empirical relationships derived from experimental data gathered from literature. MSTDB-TP is managed by Oak Ridge National Laboratory (ORNL), and MSTDB-TC is managed by The University of South Carolina. Both data compilations are publicly hosted on ORNL gitlab servers (see Section 5. Access Process for access instructions). This report focuses only on the development of MSTDB-TP.

2. DATABASE COMPOSITION

Currently, the TP data held in MSTDB-TP consists of compiled data from more than 140 published experiments conducted by various national labs, industries, and university projects. The data held in the ORNL gitlab server consists of two CSV files: one containing all the property data for each salt and their respective compositions and the other containing reference data (full references and DOIs). The property data for molten salts include the following:

- Melting temperature
- Boiling temperature

- Density
- Viscosity
- Thermal conductivity
- Heat capacity

For each property, a shorthand reference, experimentally measured range, empirical constants, and an estimation of the measurement errors are provided. Although all properties are present in the database, each entry (i.e., one specific salt mixture) may not have all the properties present. Property availability for salt mixtures with up to four components are shown in Figures 1 and 2. Each table or figure shows the salt property data available in the database, as well as the number of measurements over each salt's compositional space that have been done for each property.

In addition to these two CSV files, a third CSV file containing Redlich-Kister (RK) parameters for density estimation for select pseudobinary mixtures in MSTDB-TP is also stored on the gitlab server; more details on this file are provided in Section 2.1.

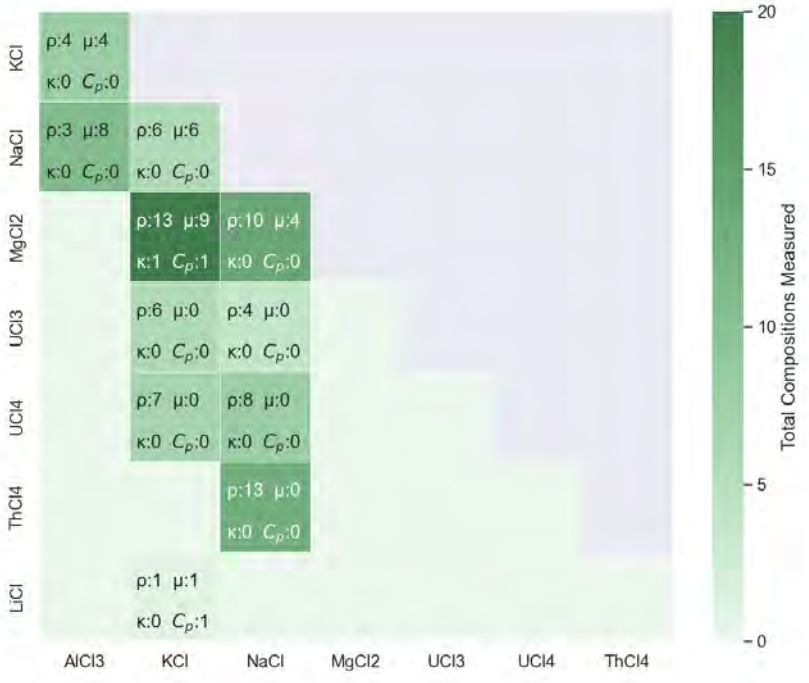
Notably, MSTDB-TP and MSTDB-TC are consistent in regards to multicomponent system melting points and pure compound heat capacities. All multicomponent salt systems that have been thermodynamically evaluated by MSTDB-TC v2.0 have had their melting points calculated for MSTDB-TP by Thermochimica software using the Gibbs energy models in the MSTDB-TC database. Those systems that have not been been evaluated by MSTDB-TC 2.0 use existing experimental melting point literature data for reporting in MSTDB-TP, if available. Heat capacity values for pure compounds have been extracted from the thermodynamic value tabulations within MSTDB-TC, and source references have been associated when available. Notably, multicomponent system heat capacities in MSTDB-TP do not mimic MSTDB-TC model outputs; rather, they are extracted directly from experimental values in literature.

2.1 REDLICH-KISTER EXPANSION AND MUGGIANU INTERPOLATION

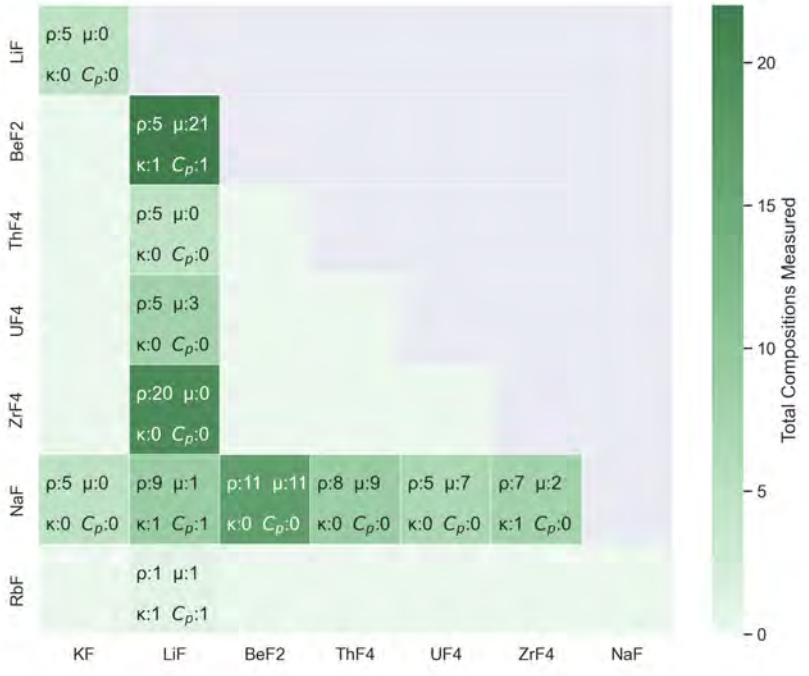
RK expansion uses well-characterized psuedobinaries and pure components of the higher-order systems to calculate binary interaction parameters. These interaction parameters can then be used to predict the density and viscosity of higher-order systems through Muggianu interpolation of any composition, making this method very attractive for filling in major gaps in MSTDB-TP. This technique has been demonstrated for predicting density in higher-order chloride and flouride molten salt systems [5, 6, 7]. The main drawback of this technique is that it relies on experimental data. To produce the interaction parameters for higher-order systems, the lower-order systems (e.g., pure or pseudobinary salts for pseudoternary) need to be well-characterized. Having experimental data in the system of interest can also drastically increase the accuracy. The development of RK estimation techniques is a focus of the maintainers of MSTDB-TP, and RK psuedobinary interaction terms have been tabulated and stored on the MSTDB-TP gitlab project.This tabulation is readable by Saline for users who want to estimate the densities of arbitrary salt mixtures.

3. DATABASE TOOLS

Currently, two tools have been developed to assist in the use of the database: an application programming interface (API) called Saline and a graphical user interface (GUI). Saline was developed to be a robust connection between MSTDB-TP to modeling/simulation codes, whereas the GUI was developed to make MSTDB-TP significantly more accessible to all types of users.



Chloride psuedobinary salts



Fluoride pseudobinary salts

Figure 1. Heat map of measured properties for chloride and fluoride pseudobinary salts in MSTDB-TP v2.1.1.

Salt	Measurements			
	ρ	μ	κ	c_p
AlCl3	1	1	0	1
BeCl2	1	0	0	0
BeF2	1	1	1	1
CaCl2	1	1	1	1
CaF2	1	1	1	1
GdCl3	1	1	0	0
GdF3	0	0	0	0
KCl	1	1	1	1
KF	1	1	1	1
LaCl3	1	1	0	0
LaF3	1	0	0	1
LiCl	1	1	1	1
LiF	1	1	1	1
MgCl2	1	1	1	1
MgF2	1	1	1	0
NaCl	1	1	1	1
NaF	1	1	1	1
NdCl3	1	1	0	0
NdF3	0	0	0	1
NpCl3	0	0	0	0
NpF3	0	0	0	0
PuCl3	0	0	0	1
PuF3	0	0	0	1
SrCl2	1	1	1	0
SrF2	1	1	1	0
ThCl4	1	0	0	0
ThF4	1	0	0	0
UCl3	1	0	0	1
UCl4	1	0	0	0
UF3	0	0	0	1
UF4	1	1	0	1
ZrCl4	1	1	0	0
ZrF4	1	0	0	0

Salt	Measurements			
	ρ	μ	κ	c_p
KCl-LiCl-NaCl	4	0	0	0
LiCl-NaCl-AlCl3	10	10	0	0
LiF-BeF2-ThF4	3	2	0	0
LiF-BeF2-ZrF4	1	0	0	0
LiF-NaF-BeF2	1	1	0	0
LiF-NaF-KF	1	1	1	1
LiF-BeF2-UF4	36	36	0	0
NaF-BeF2-UF4	79	71	0	0
NaF-KF-BeF2	1	1	0	0
NaF-KF-MgCl2	1	0	0	0
NaF-KF-UF4	1	1	1	1
NaF-KF-ZrF4	1	1	0	0
NaF-LiF-BeF2	4	4	0	0
NaF-LiF-ZrF4	10	1	0	1
NaF-ZrF4-UF4	5	3	2	3
RbF-ZrF4-UF4	2	2	1	1

Salt	Measurements			
	ρ	μ	κ	c_p
LiF-BeF2-UF4-ThF4	1	1	0	0
LiF-BeF2-ZrF4-UF4	1	0	0	0
NaF-LiF-BeF2-UF4	1	1	0	0
NaF-LiF-KF-UF4	2	2	1	1
NaF-LiF-ZrF4-UF4	1	1	0	1

Figure 2. Tables of the number of measured properties in MSTDB-TP for pure, pseudoternary, and pseudoquaternary salts.

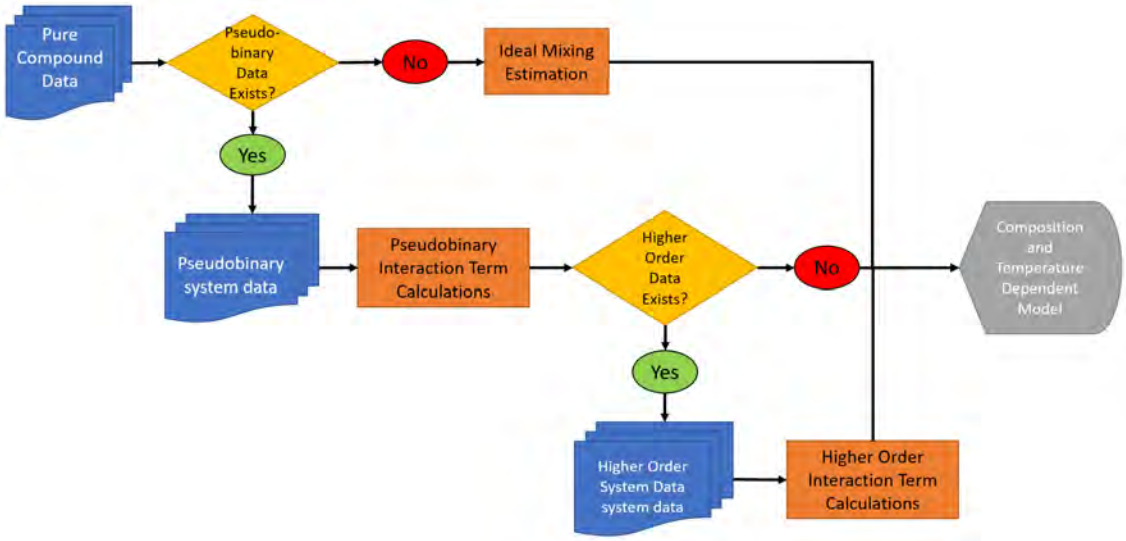


Figure 3. Diagram of the process by which a property is estimated via experimental measurements, assumption of ideal mixing, or use of system interaction terms. Blue documents represent experimental datasets, orange boxes represent calculational processes, yellow diamonds represent decision points, circles represent boolean conditions, and the final result (in grey) is the estimation of a higher-order system.

3.1 SALINE

Saline is an API for MSTDB-TP TP property data. It allows for the integration of density, viscosity, thermal conductivity, and heat capacity into client applications through a collection of classes and methods. The interface is predominately written in C++11. Additional language bindings (i.e., Fortran, Python) are supported.

Using the C++ interface, the main class that users will use is the "Thermophysical Property" class. Specific salt data are accessed through this class by the "setCompostion" method, with which each salt end member and their respective composition are passed to the model. The data are then pulled from the database, and each property (i.e., density, viscosity, thermal conductivity, and heat capacity) and their respective uncertainty can be called individually for a given temperature or enthalpy for use. The data models that are employed by "Thermophysical_properties" are implemented in a "Data_Store" class. These two modules are separated to allow the opportunity for the client to implement alternative data implementations without extensive effort to modify Saline to adopt it. Default_Data_Store is what accesses MSTDB-TP and is what is recommended for current use with Saline [3].

Saline also supports the use of estimation to fill out voids in the data. By using the "R_Kister_Data_Store", pseudobinary data can extrapolated using RK polynomials (see Section 4.1.1) to support arbitrary salt compositions. Currently, RK estimation can only be used for density, but support is being developed for viscosity. Ideal mixing will be used for any salt that does not have RK parameters. Any calculations using a model without experimentally validated data may be inaccurate, and users should be cautious when employing this module.

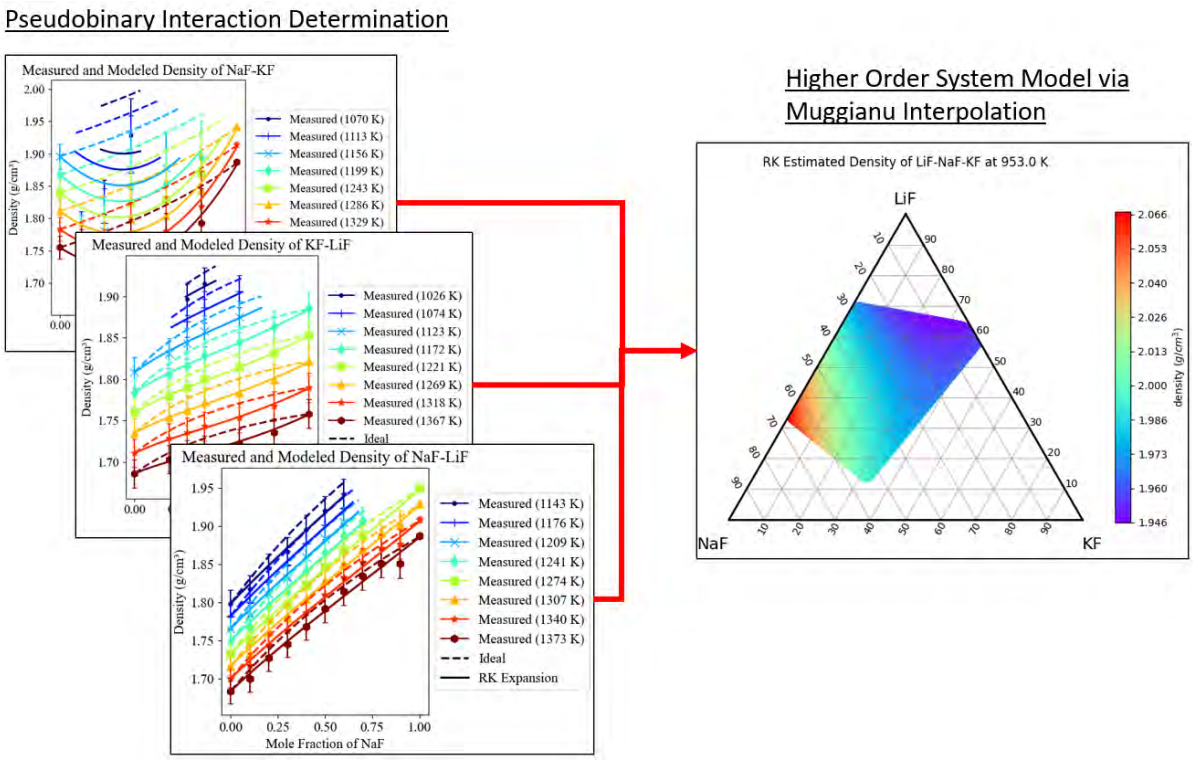


Figure 4. Example of the The RK/Muggianu interpolation process with FLiNaK density as the use case. Psuedobinary interaction terms are calculated from pseudobinary subsystem data and are used to inform the higher-order FLiNaK model through Muggianu interpolation.

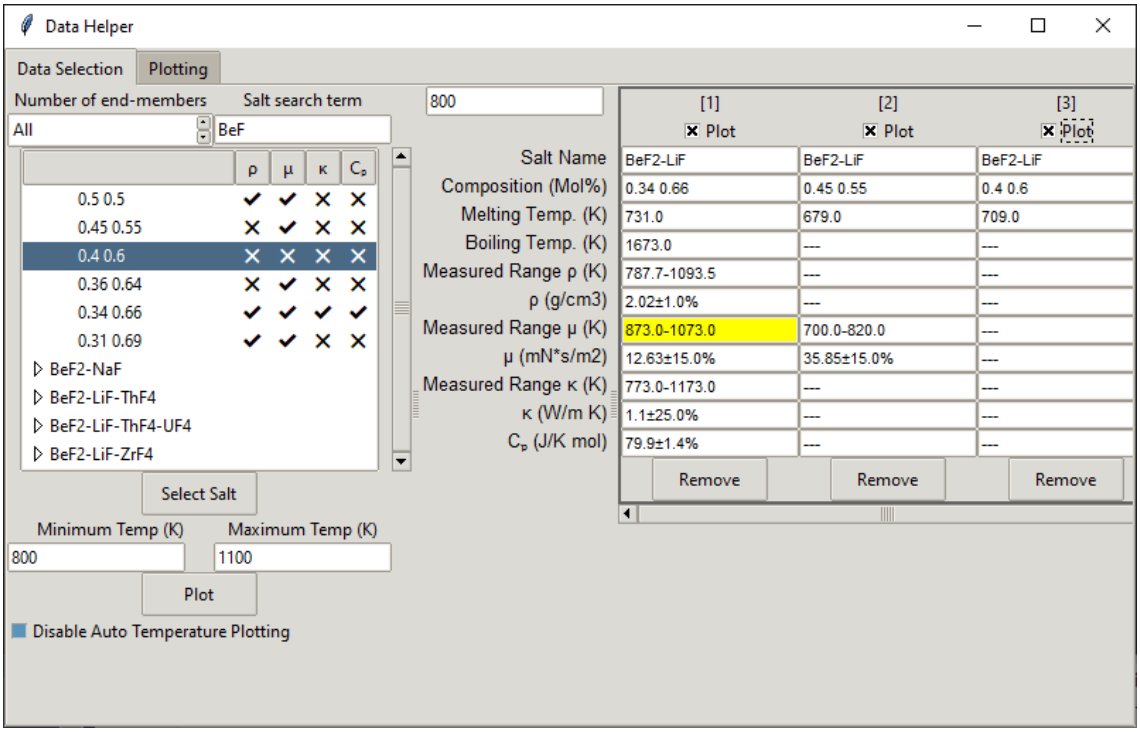


Figure 5. Screenshot of the GUI salt selection and browsing process with property calculation.

3.2 GRAPHICAL USER INTERFACE

The GUI was developed in Python to promote increased accessibility to MSTDB-TP data for all types of users. Currently, an executable file can be downloaded from MSTDB-TP for Windows, but all operating systems will be supported in future iterations. The GUI allows the user to perform a wide variety of actions, including the following:

- Browse or search salts that are present in the database. Salts can be filtered by number of salt species (e.g., pure, binary, ternary) or by search term (e.g., "Cl," "F," "NaCl").
- Calculate TP properties (i.e., density, viscosity, thermal conductivity, and heat capacity) at a user-defined temperature for user-selected salt/composition combinations. Uncertainty, experimentally measured range, melting temperature, and boiling temperature are shown, along with the calculated properties.
- Plot TP properties for user-selected salts over either an experimentally measured range (automatic) or a user-defined temperature range (manual).

3.3 MOLTEN SALT THERMAL PROPERTIES DATABASE WEBSITE

For additional access to the information contained in the database, a website has been developed for MSTDB (https://mstdb.ornl.gov/). The website is split into different sections:

- About: Section including motivation behind the database and different supporting groups for the TP arm and TC arm

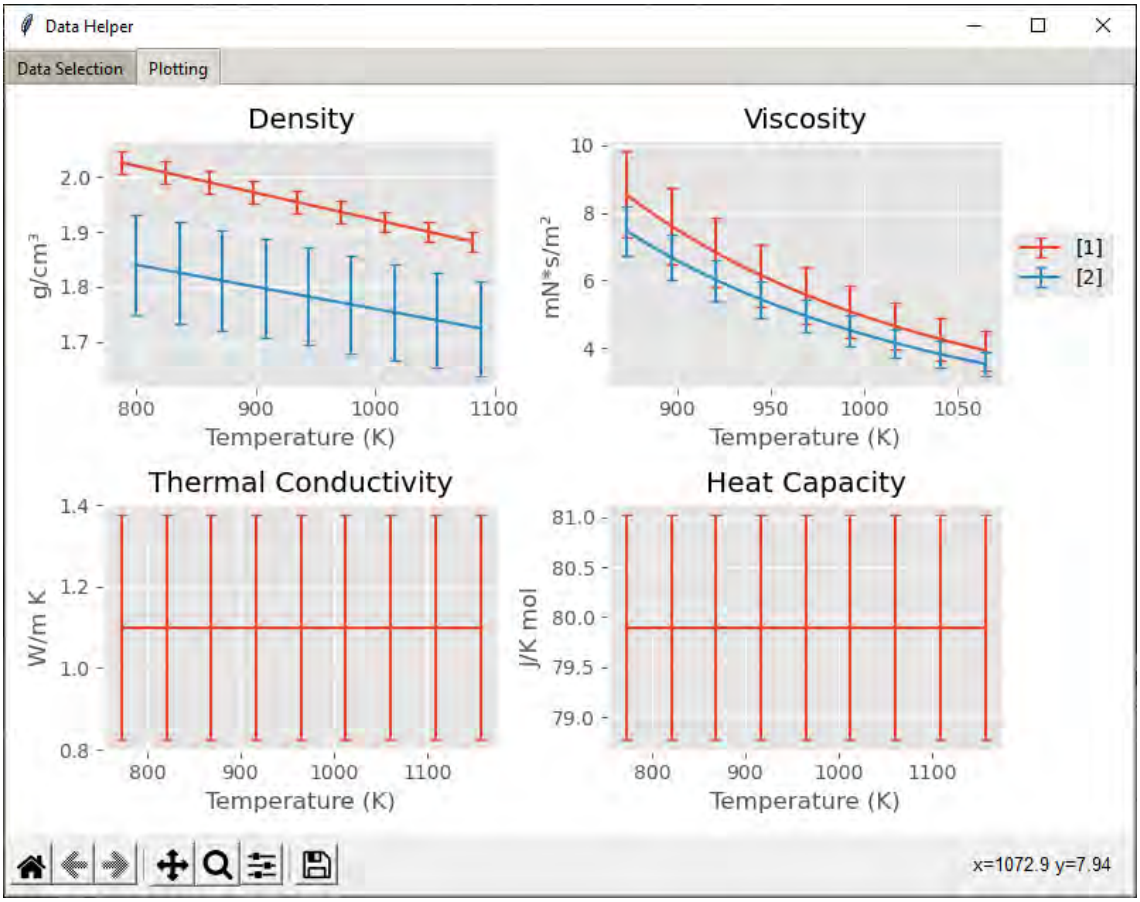


Figure 6. Screenshot of the GUI property plotting.

- TP: Multiple web pages including information on data breakdown, estimation techniques, the GUI, and Saline; serves as a landing point for MSTDB-TP arm
- TC: Contains a general description of the TC side, as well as a breakdown of data included in the database (salt systems)
- Publications: Page that includes many recent publications associated with MSTDB-TC/TP and their associated references
- News: A web page of articles covering recent events for MSTDB (e.g., workshops, new publications, version releases)
- Team: A list of profiles for all the MSTDB team members

Although the website is purely informational at the current time, expansion of the site includes potentially incorporating some of the GUI's functionality directly into the website, as well as some more interactive database visualization tools.

4. ONGOING AND FUTURE DEVELOPMENT OF MOLTEN SALT THERMAL PROPERTIES DATABASE–THERMOPHYSICAL

4.1 DATABASE EXPANSION

Generally, the database expands by introducing new salts and properties and further filling in information about existing salts (through more property data or with the compositional space). Current work is being done to expand the information about actinide chlorides for density and viscosity and should be included in the next major release for MSTDB-TP. Furthermore, future work planned for later versions includes adding a new property—surface tension—with a significant amount of work already done in gathering data in literature on salts already contained in the database. Finally, the last near-term goal being planned for the database is the introduction of computationally generated data either within MSTDB-TP or by leveraging data in MSTDB-TP. Modeling techniques are highly efficient in fleshing out compositional spaces of well-known salts and supplementing the development of experimentally challenging salt systems, but these techniques require careful scrutiny in determining their accuracy. Modeling techniques being considered for MSTDB-TP can be broken down into two categories: extrapolation/interpolation techniques (RK) and first principles calculations (e.g., ab initio). All computational data introduced into MSTDB-TP will be clearly distinguished from experimental data. To be clear, extrapolation/interpolation techniques leverage the data in MSTDB-TP to make estimations for unmeasured systems. Ab inito models would be implemented in the fundamental MSTDB-TP tabulations, assuming that the uncertainty can be quantified.

4.1.1 First Principles Calculations

First principles calculation of molten salt TP properties refers to simulating small atom-scale systems through density functional theory or other molecular dynamic simulations. The size and length of the simulation depends on the property trying to be ascertained. For example, density and heat capacity only require systems in the hundreds of atoms, whereas more complicated properties such as viscosity, diffusion, or thermal conductivity require thousands of atoms and longer simulation times to converge. An extensive collection of literature exists in which TP properties have been computed using a variety of first principles–based methods [8].

The major drawback to all first principles modeling is that the accuracy of the data comes down to how well a given theoretical model captures the interactions between atoms and the difficulty in scaling these calculations to more complicated and longer simulations. For completely unmeasured salts, determining the accuracy of the generated data is very difficult because the choice of the potential description, the method chosen, intricacies associated with the unique salt being studied, the property being studied, and simulation sizes and timescales will all affect the uncertainty or error associated with the model output. Computational data introduced to MSTDB-TP will be under extreme scrutiny, with very conservative errors for any nonvalidated data.

4.1.2 Progress on Applying Redlich-Kister to Viscosity

Although previous research established that the RK expansion is applicable to a wide variety of molten salt systems for density, for interpolating among the pseudobinary compositional space and extrapolation to the pseudoternary space, researchers aim to establish a working framework for viscosity in a similar vein. However, a few challenges exist with viscosity:

- Viscosity changes exponentially as a function of temperature, and the framework would require enough flexibility in the temperature domain to handle drastic changes in this domain.
- A well-established ideal mixing model for molten salt viscosity does not exist to use as a basis for the framework; a few potential candidate models exist, but they must all be rigorously tested to establish an appropriate ideal model.
- Viscosity can change orders of magnitude as a function of composition for mixtures because of some constituents that may be particularly viscous (e.g., BeF_2-bearing mixtures).

Three potential ideal mixing models have been identified:

- The Gambill method [9]
- The Grunburg-Nissan rule [10]
- The Katti-Chaudhri rule [11]

Nonideal mixing terms can be summatively considered in the mathematics based on the RK formalism, as outlined previously [5]. However, one small caveat is that the summative terms are constructed based on the logarithm of viscosity, which is consistent with modified viscosity models based on the aforementioned ideal methods to handle combinations of ideal and nonideal terms. Notably, nonideal mixing can be handled with a modified version of the Katti-Chaudhri rule, which involves determination of excess activation energy and falls within the quasichemical formalism. This mixing has been studied by Flores [12], but only the RK framework is considered in this work. Exact mathematical formalisms are planned to be be released in peer-reviewed journal articles in the future for the work described herein.

Some preliminary results of the application of the Grunburg-Nissan ideal model and RK expansion for nonideal mixing terms are shown in Figure 7 for LiCl-KCl and NaF-BeF_2. The isotherms shown in the plots are truncated to reflect the valid experimental ranges to which the models are fit. Experimental data for the pure compounds in LiCl-KCl come from Wakao [13] and Ejima [14], whereas the mixture data comes from Zuca [15]. Experimental data for the pure compounds in NaF-BeF_2 come from Brockner [16] and [17], whereas the mixture data came from studies of Powers [18] and Blanke [19]. The mixture of LiCl-KCl is the more ideal mixture in that the Grunburg-Nissan model without any nonideal terms

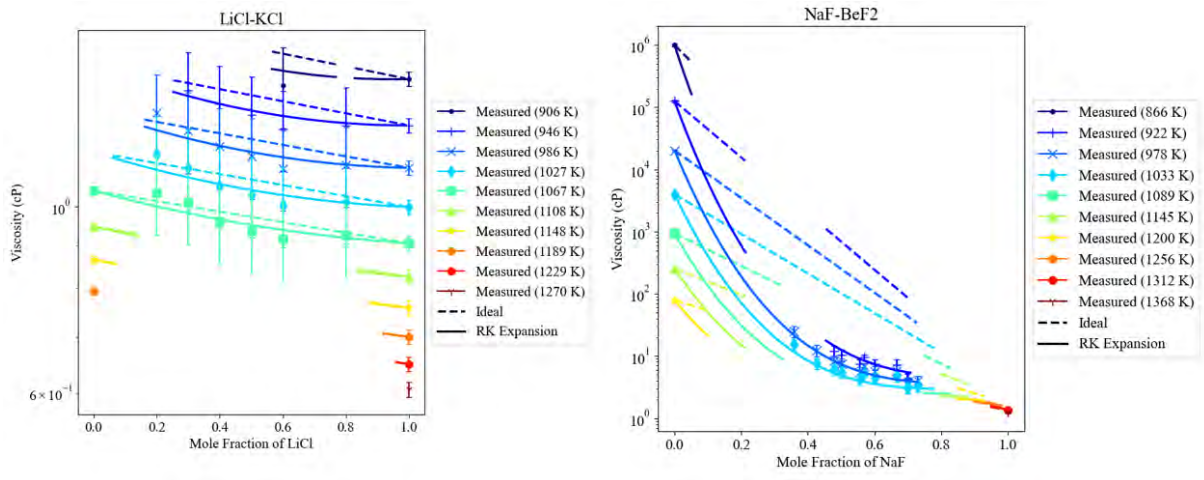


Figure 7. Measured and modeled viscosities of (left) LiCl-KCl and (right) NaF-BeF_2 as a function of temperature and composition. Models include an ideal Grunburg-Nissan model, as well as an RK model, which is based on Grunburg-Nissan but with added interaction terms following the RK framework.

represents the experimental data reasonably well. Contrarily, NaF-BeF_2 is extremely nonideal in that the mixtures are not nearly as viscous as the ideal model would suggest. Regardless, the application of the RK expansion to capture nonideality shows success for both salt mixtures to reflect experimental values as a function of composition and temperature.

Future work will inclue a more exhaustive application to many salt mixtures in MSTDB-TP where sufficient data exists, as well as application to higher-order systems. The LiF-NaF-KF is an ideal candidate because it is the only pseudoternary salt mixture with a well-validated experimental characterization of viscosity [20]. Contrarily, other higher-order systems generally lack independent experimental validation and have only been measured in one to two independent studies.

4.2 IMPROVEMENTS TO THE GRAPHICAL USER INTERFACE

The goal of the GUI is to ultimately replace casual interaction with the raw CSV files in MSTDB-TP. Currently, the GUI can only be used to browse, calculate, and plot property values at temperatures, but researchers have plans to expand the GUI to allow much more manipulation and compilation of the data in MSTDB-TP. Current features under development include the following:

- Export tool: A tool to choose any combination of salt data within the database and compile it in whatever format is useful to the user
- Estimation tool: A tool that takes advantage of existing RK parameters in MSTDB-TP to calculate applicable properties for eligible salt systems
- Quality of life improvements, such as advanced filters and more property information (reference and empirical constant data)

5. ACCESS PROCESS

MSTDB-TP is hosted on a publicly accessible server at ORNL: https://code.ornl.gov/neams/mstdb-tp. Access to this server requires an ORNL XCAMS account and MSTDB membership.

5.1 XCAMS ACCOUNT CREATION

XCAMS allows external, non-ORNL users to access code.ornl.gov. XCAMS accounts are created as follows:

1. Go to https://xcams.ornl.gov.
2. Select **"I need an account."**
3. Read and acknowledge the **"User Agreement."**
4. Enter your **email address** and **username**, following the guidelines provided on the page.
5. Enter **"Personal Information"** and **"Contact Information"** according to the guidelines provided on the page.
6. Create an XCAMS password according to the guidelines provided on the page.
7. On the final step, note the activation sequence box. Wait until each action item turns green and the box heading reads **"Transactions Complete."**
8. Log in to https://code.ornl.gov using your new XCAMS username and password.

5.2 REQUEST MOLTEN SALT THERMAL PROPERTIES DATABASE MEMBERSHIP

To acquire MSTDB membership, send an email to mstdb@ornl.gov with "MSTDB Access Request" as the subject, and include the following information:

1. Your XCAMS ID
2. A summary of the purpose of your access

You will receive an email from code.ornl.gov indicating that your access has been granted and providing a link to the project page.

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