Empirical estimation of densities in NaCl-KCl-UCl_3 and NaCl-KCl-YCl_3 molten salts using Redlich-Kister expansion

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Abstract

Densities of molten KCl-NaCl-UCl_3 and KCl-NaCl-YCl_3 ternary systems have been estimated using a multidimensional Redlich-Kister model. Temperature and composition dependent Redlich-Kister functions have been used to generate binary interaction parameters in the outlined ternary salt systems. These binary interactions have been used in the extrapolation to ternary system densities. The results of the density extrapolations by Muggianu interpolation scheme provide agreement within 2-3% for the NaCl-KCl-YCl_3 liquids and 11% in NaCl-KCl-UCl_3 liquids compared to the available experimental data. Modeling NaCl-KCl-UCl_3 molten phase density with a ternary interaction parameter improved the agreement within 4%. Thermophysical modeling used in this study has shown promising results for use in other material properties, such as viscosity, thermal conductivity, and heat capacity of the molten salts. Furthermore, the outlined modeling method applied in these specific molten salt ternaries can be used for quaternary or higher multicomponent molten salt systems.

Keywords: Molten salts, density, NaCl-KCl-UCl_3, NaCl-KCl-YCl_3, nuclear energy, Redlich-Kister model

1. Introduction

Molten salts are of interest as a fuel or a coolant in Molten Salt Reactors (MSRs). They achieve higher thermal efficiency, potentially reducing costs compared to conventional Light Water Reactors (LWRs), which makes them promising candidates for the next generation of nuclear reactors (Qualls, 2018; Serp et al., 2014; Zhang et al., 2018). Some of the perceived operational and safety advantages of MSRs over LWRs are: the fluid nature of the fuel; high thermal efficiency; online refueling and processing options; elimination of high pressure vessels as opposed to LWRs; utilization of the conversion of fertile thorium to fissile ^{233}U taking advantage of the abundance of thorium (3 times that of uranium (Degueldre and Joyce, 2020)); and reduction in transuranic wastes (LeBlanc, 2010). There are different designs, some with unique features addressing technological challenges like radionuclide retention through the use of TRISO fuel in combination with a molten salt. Others address waste by using a fast neutron spectrum to minimize long-lived, hazardous radioisotopes. MSRs are potential candidates for replacing existing fossil fueled plants. However, the great potential of MSRs over existing nuclear reactors and fossil fuel industry will remain speculative until proven through operation.

A complete understanding of thermophysical properties of molten salts is necessary for the proper design, safety analyses and operation (Greenwood et al., 2020). Thermophysical and thermodynamic properties of interest include density, viscosity, thermal conductivity, and heat capacity. These properties have been experimentally and theoretically studied in the past for specific compositions in relation with temperature and compiled in several reports (Janz, 1988; Jerden, 2019; Magnusson et al., 2020; Williams, 2006a, 2006b). The US Department of Energy (DOE) Nuclear Energy Advanced Modeling and Simulation (NEAMS) program in coordination with the Molten Salt Reactor Campaign is developing the Molten Salt Thermal Properties Database (MSTDB) (McMurray et al., 2021). The thermophysical properties version of MSTDB, that is MSTDB-TP, contains parameters for relevant models developed using experimental data from the literature. The models are thermophysical property relations with composition and temperature that can be used standalone or with multiphysics tools for MSR modeling and simulation (Betzler et al., 2019; McMurray et al., 2018).

It is well established within the materials thermodynamics community that multicomponent system behavior can be estimated by extrapolation from the lower order subsystems. The unaries

and binaries are therefore fundamental. It has been demonstrated that two species interactions generally dominate (Chartrand and Pelton, 2001; Hwang et al., 1991; Kattner, 1997). The MSTDB-TP is to be populated with property relations for pure and pseudo-binary[1] salts with respect to temperature and composition. A formalism based on a Redlich-Kister expansion (Redlich and Kister, 1948) corrects for the interactions in binary systems that can be combined for predicting property behavior in higher order systems using the Muggianu interpolation method (Hillert, 1980; Luo et al., 2019; Muggianu et al., 1975). In this way, populating the MSTDB with pure salt properties, and pseudo-binary mixture properties allows for reference data-based extrapolation into multicomponent space (i.e., properties of a base salt with fission products). Interactions can be refined with follow-on studies for more specific compositions as needed.

In this study, densities of molten NaCl-KCl-UCl_3 and NaCl-KCl-YCl_3 ternary systems have been modeled using the Redlich-Kister (RK) model (Redlich and Kister, 1948). Property measurements of the NaCl-KCl-UCl_3 salt system are underway (Rose and Thomas, 2021). YCl_3 is one of the well-known rare-earth fission products in MSRs (Iwadate et al., 2014). UCl_3-KCl and UCl_3-NaCl binary system densities were measured using the method of maximum pressure in a gas bubble (Desyatnik et al., 1976, 1975). Katyshev et al. (1983) measured the densities of NaCl-KCl-UCl_3 ternary chloride mixtures. Van Artsdalen and Yaffe (1955) measured the density of the NaCl-KCl binary system using the Archimedes method. Mochinaga and Irisawa (1974) measured the density of the NaCl-YCl_3, KCl-YCl_3 and NaCl-KCl-YCl_3 systems using dilatometry.

2. Modeling Approach

The RK model is shown in Equation 1 to represent the behavior of molten salt density with composition.

$$\rho_{mix} = \rho_{id} + \rho_{ex} \qquad \text{Equation 1}$$

Here, ρ_{mix} is the density of the mixture, ρ_{id} is the density with ideal behavior and ρ_{ex} is the excess term to account for the deviation from ideal behavior. Some reports interpret ideal density as the weighted molar average of pure salt densities indirectly (Cooper and Asfour, 1991; Guo et al., 1989). Others use weighted averages of molar volume (additive behavior) and convert the molar

[1] Since the end-members are halides, a two salt mixture is a pseudo-binary since the system is properly a ternary.

volume to density in the modeling (Chrenková et al., 2003; Kubiková et al., 2020; Mlynáriková et al., 2017). Using the additive behavior approach, the ideal behavior of density is given in Equation 2 as:

$$\rho_{id} = \frac{\sum x_i MW_i}{\sum \frac{x_i MW_i}{\rho_i}} \quad \text{Equation 2}$$

where MW_i is the molecular weight of component *i and* x_i is the mole fraction of component *i*. This expression is simply the ratio of average molar mass to average molar volume. The non-ideal behavior density term is defined using the RK expansion (Redlich and Kister, 1948), which has been used for CALPHAD Gibbs energy functions (McMurray and Besmann, 2018). Equation 3 shows the excess term via the RK expansion, which accounts for the non-ideal density with two components, A and B.

$$\boldsymbol{\rho_{ex} = x_A x_B \sum_{j=1}^{n} L_j (x_A - x_B)^{j-1}} \quad \text{Equation 3}$$

where *j* defines the order of the expansion. L_j defines the j-th order binary interaction parameter between A and B, and it has a linear temperature dependence (Equation 4).

$$\boldsymbol{L_j = A_j + B_j T} \quad \text{Equation 4}$$

Temperature units are all in Kelvin. More temperature dependent terms can be added to Eqn. 4 per the Redlich-Kister model; however, for simplicity, it was treated as a linear function. Ternary systems have been modeled using this approach. However, the ternary interactions have been considered negligible as the initial guess. The NaCl-KCl-UCl_3 required a ternary interaction parameter which was explained in the Results and Discussion part. Equation 5 shows a typical ternary RK expansion with three binary interaction terms.

$$\boldsymbol{\rho_{ex} = x_A x_B \sum_{j=1}^{n} L_j (x_A - x_B)^{j-1} + x_A x_C \sum_{j=1}^{n} L_j (x_A - x_C)^{j-1} + x_B x_C \sum_{j=1}^{n} L_j (x_B - x_C)^{j-1}} \quad \text{Equation 5}$$

Ternary halide system studies using the Redlich-Kister model at a single temperature have shown that ternary interaction parameters may be either negligible or zero depending on the mixture (Cibulková et al., 2006; Guo et al., 1989; Kubíková et al., 2016). In the final expression, ρ_{ex} in Equation 1 is composed of a summation of three binary terms between A, B and C

components. The final model estimates only the molten salt phase densities rather than the densities of the overall systems. Therefore, temperature and composition ranges of interest apply solely to regions of the corresponding phase diagrams representing molten salts rather than other combinations of phases.

3. Parameter Estimation Method

A multidimensional least squares approach has been used as the interaction parameter estimation method for the molten KCl-NaCl-UCl_3 and KCl-NaCl-YCl_3 ternary systems using the experimental data from Katyshev (1983) and Mochinaga (1974) (Katyshev et al., 1983; Mochinaga and Irisawa, 1974). MATLAB's curve fitting tool with Trust-Region and Levenberg-Marquardt (Levenberg, 1944; Marquardt, 1963) algorithms were utilized in the calculations.

KCl-NaCl, KCl-UCl_3, NaCl-UCl_3, KCl-YCl_3 and NaCl-YCl_3 binary densities have been calculated using the Redlich-Kister model outlined in Eqns 1-5. Temperature dependent linear fits taken from experimental density data at different compositions have been converted to a matrix. Each density equation had different valid temperature ranges due to experimental limitations and/or liquidus boundaries. In order to extrapolate and get a single equation for the whole composition range in a binary, the minimum and the maximum measurement temperature of all compositions were taken. Although the final equation will yield a density relation at all compositions and temperatures in the range, part of it can be unphysical due to phase relations. In the estimation of the valid temperature ranges for the final equation, the phase diagram for each system must be considered.

4. Results and Discussion

Consistent density relations for the pure salts are needed to model the binary molten salt phases. Since some experimental studies use different relations for the pure salts, a single equation had to be selected for each end-member salt given in Table 1.

Table 1

Selected pure salt density relations that are used in the modeling and interpolations

Pure Salt	Density Relation (g/cm^3), T in K	Reference
KCl	2.1360 – 5.831 x 10^{-4} T	(Van Artsdalen and Yaffe, 1955)
NaCl	2.1394 – 5.430 x 10^{-4} T	(Van Artsdalen and Yaffe, 1955)
YCl_3	3.0481 – 4.594 x 10^{-4} T	(Mochinaga and Irisawa, 1974)
UCl_3	6.3747 – 1.5222 x 10^{-3} T	(Desyatnik et al., 1975)

The fitting parameter A_1 in Eqn. 4 is the dominant term and shows the degree of non-ideality due to mixing of the liquids. It also indicates whether the deviation from the ideal behavior is negative or positive. For the molten salts discussed in this work, a parameter A_1 that is smaller than 0.5 suggests close-to-ideal behavior for engineering design purposes.

Calculated binary interaction parameters, L_j, are shown in Table 2. KCl-NaCl system experimental density data (Van Artsdalen and Yaffe, 1955) showed ideal behavior with 1 percent uncertainty range (Fig. 1), although it was shown to have a small deviation from ideality (Robelin et al., 2007). Ideal mixing volume for this system is supported by Van Arstdalen and Yaffe (1955) and Holm (1971). Non-ideal mixing volume differences of NaCl-KCl at 1073 K are well below 1 % of the ideal volumes (Holm, 1971). The calculated interaction parameters for this system supports ideal behavior. Therefore, the NaCl-KCl density was considered to have an ideal behavior in the interpolations to ternaries.

Density fit results for NaCl-YCl_3 and KCl-YCl_3 revealed ideal density behavior (Fig. 2). Reference data from both molten salt systems are fit to first order Redlich-Kister equation for comparison with the ideal behavior equation (Eqn. 2). The RK model and the ideal behavior lines show visually insignificant differences and fit well to the experimental data. A_1 terms calculated for both systems are below 0.5, which suggests close-to-ideal behavior. However, there are fluctuations in the data from Mochiaga and Irisawa (1974) depicting that the calculated deviation by RK model may be within the experimental errors. The density fit temperature ranges were 998-1278 K and 998-1276 K for NaCl-YCl_3 and KCl-YCl_3, respectively.

Using the ideal behavior and interaction parameters in Table 2, density of the KCl-NaCl-YCl_3 has been extrapolated from the pseudo-binaries and compared to existing experimental data for the pseudo-ternary (Mochinaga and Irisawa, 1974). The resulting density interpolation is shown in Fig. 3 with equimolar KCl and NaCl mole fractions and 5 % uncertainty error bars. Both the calculation results and the experimental data indicate ideal density behavior at all compositions in NaCl-KCl-YCl_3 molten phase. Ideal behavior and RK model are within 2-3 % of the available experimental data. The fact that NaCl-KCl-YCl_3 molten phase behaves ideally is useful for engineering purposes and eliminates the need for rigorous modeling for this specific system.

Table 2

Density binary interaction parameters ($L_j = A_j + B_jT$ (K) in Eqn. 4) using the Redlich-Kister model in KCl-UCl_3, KCl-YCl_3, NaCl-UCl_3 and NaCl-YCl_3 molten salt systems. Order of the salts are given consistent with Equation 3.

Binary System (Salt A – Salt B)	**L_1**		**L_2**	
	A_1	**B_1**	**A_2**	**B_2**
UCl_3-KCl	-2.3395	0.0015032	-1.8929	0.0016903
UCl_3-NaCl	-1.8722	0.0011698	-0.62805	0.00084717
KCl-YCl_3	0.083039	-1.3406×10^{-4}		
NaCl-YCl_3	0.15439	-0.00010099		
NaCl-KCl	-0.018469	-1.1918×10^{-5}		

* Although the KCl-NaCl system is included in Table 2, its parameters are not used in the interpolations. It is assumed to be an ideal system, see discussion.

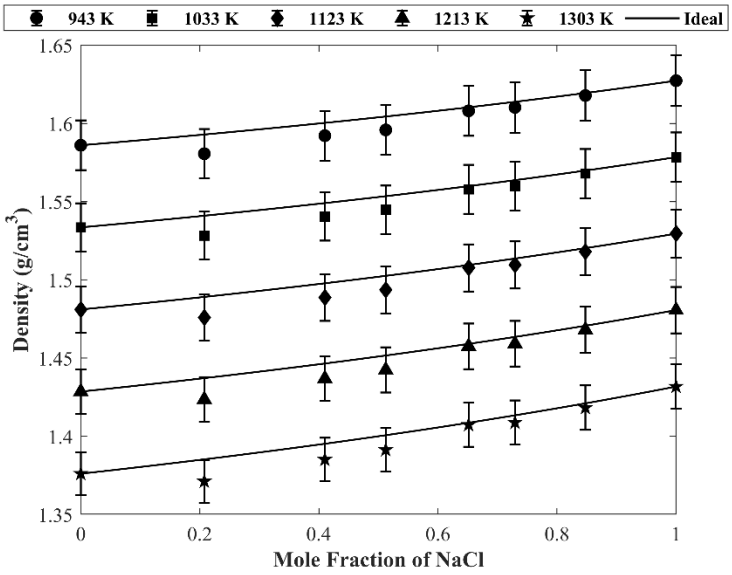


Fig. 1. Density of KCl-NaCl system comparing both ideal mixing rule and experimental data (Van Artsdalen and Yaffe, 1955) with 1 % uncertainty error bars.

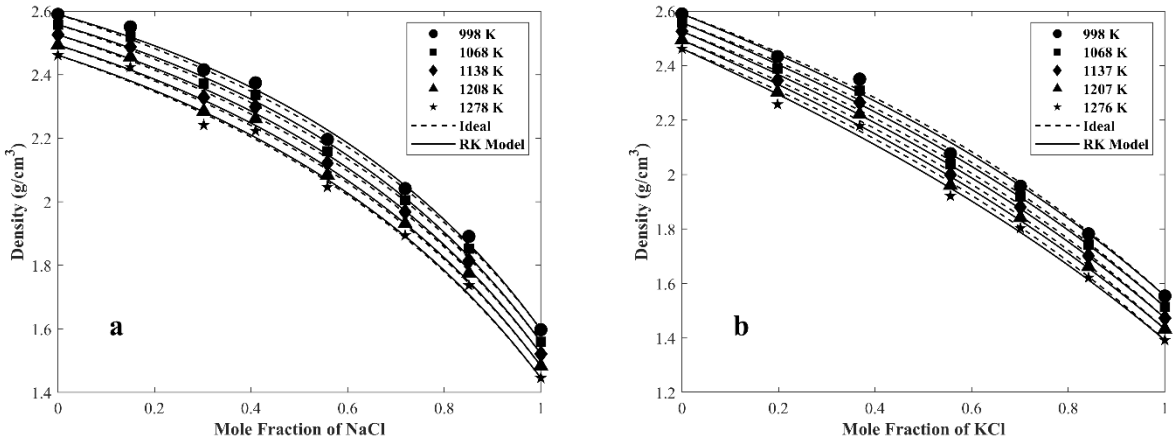


Fig. 2. Density comparisons using experimental data (Mochinaga and Irisawa, 1974), ideal behavior and Redlich-Kister model (RK Model) in **a)** NaCl-YCl_3, and **b)** KCl-YCl_3 binary systems.

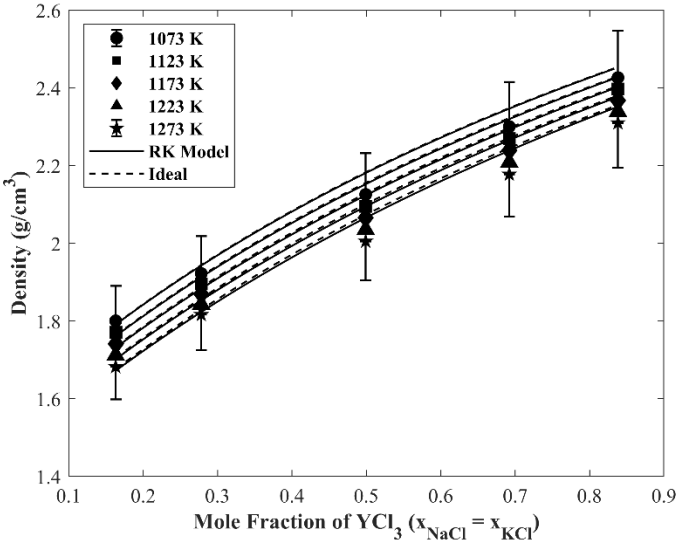


Fig. 3. Comparisons of Redlich-Kister model (shown as RK model) and ideal behavior estimation of density in KCl-NaCl-YCl_3 ternary system. Discrete symbols show the data from the literature with upper and lower 5 % uncertainty bounds added.

UCl_3-NaCl and KCl-UCl_3 binary salts have been fit to first and second order binary interaction parameters. Both systems show negative deviation from ideal behavior (Fig. 4.a-b). A_1 terms on both systems are above 1 suggesting significant nonideality. The negative density (or positive molar volume) deviation was connected to the formation of complex UCl_5^{2-} type aggregates at the intermediary compositions (Desyatnik et al., 1976, 1975). Furthermore, a first principles study of the NaCl-UCl_3 system suggested evidence of $[UCl_n]^{n-3-}$ complexes, which might support the former theory on the cause of negative density deviation (Li et al., 2019). Experimental clarification is needed to find a relationship between the complex formation and density deviations. The UCl_3-NaCl system was modeled between 892-1296 K, and UCl_3-KCl system was between 863-1296 K.

Calculations were also made for the KCl-NaCl-UCl_3 pseudo-ternary system by interpolation using the binary interaction parameters. The calculation results are compared against measured density data from Katyshev et al. (1983) who studied various KCl-NaCl-UCl_3 compositions. Fig. 5 shows all the RK model interpolations compared to experimental data. Unlike the KCl-NaCl-YCl_3 system, the measured densities deviate significantly from ideality. The interpolation resulted in negative density deviation that resembles the binary fit results. Similar to behavior in binaries,

Katyshev (1983) argued that the deviations are a result of the formation of UCl_5^{2-} complexes in the salts due to the addition of K^+ ions. The interpolations are generally in agreement with Katyshev's results within 11 %. A sharp negative density deviation is seen at 75 mol % KCl in Fig 5-c. That composition deviates 18 % from the RK model density estimation. Aside from UCl_5^{2-} ion formation, this region lies within the composition range of K_2UCl_5 endmember. This endmember congruently melts at a higher temperature (905 K) in the KCl-UCl_3 system and has been found to build up at uranium anodes during uranium electrorefining in molten chlorides (Hames et al., 2018; Rose et al., 2015). However, these results do not appear to be an artifact of the experiment because the density behavior that is measured at multiple temperatures look consistent. The sample purity could be a problem and needs to be clarified with further measurements on this specific composition. In the modeling, this sharp difference suggests that higher order terms are needed to capture non-ideal behavior.

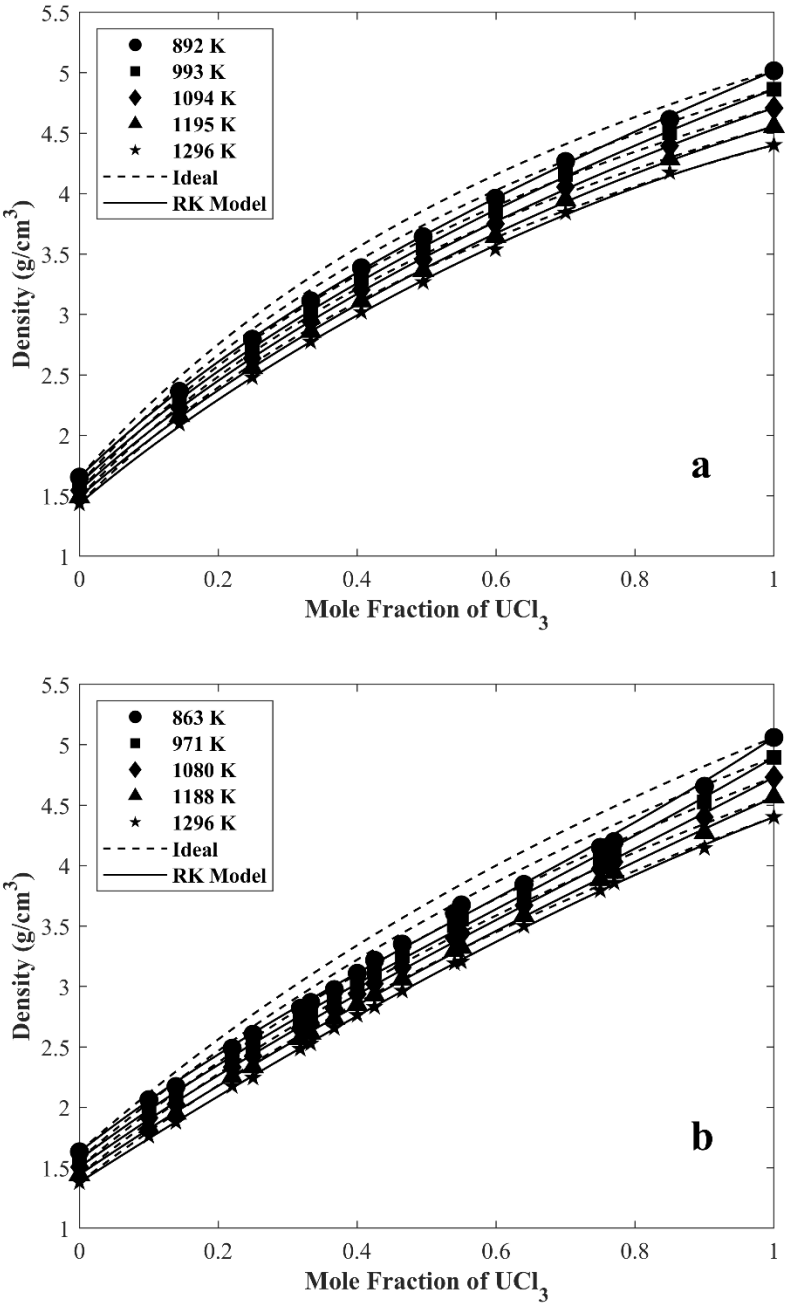
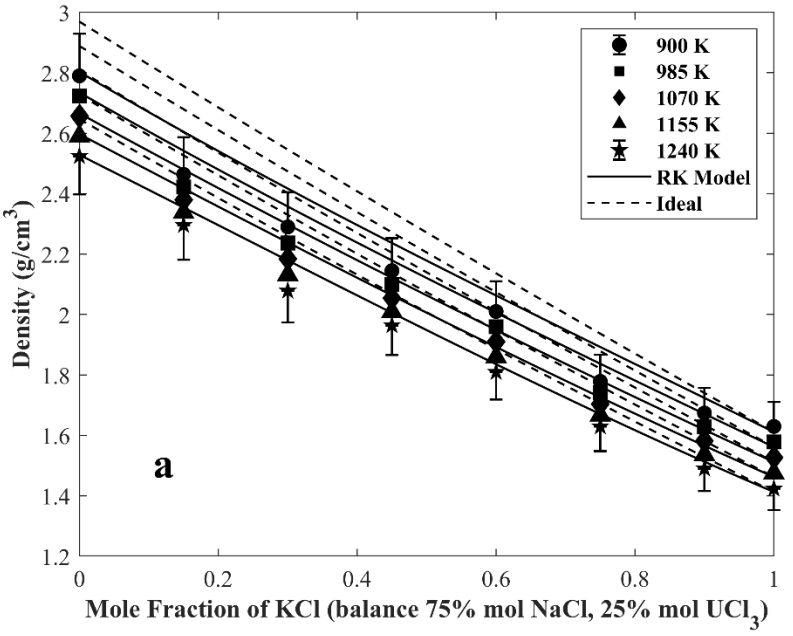
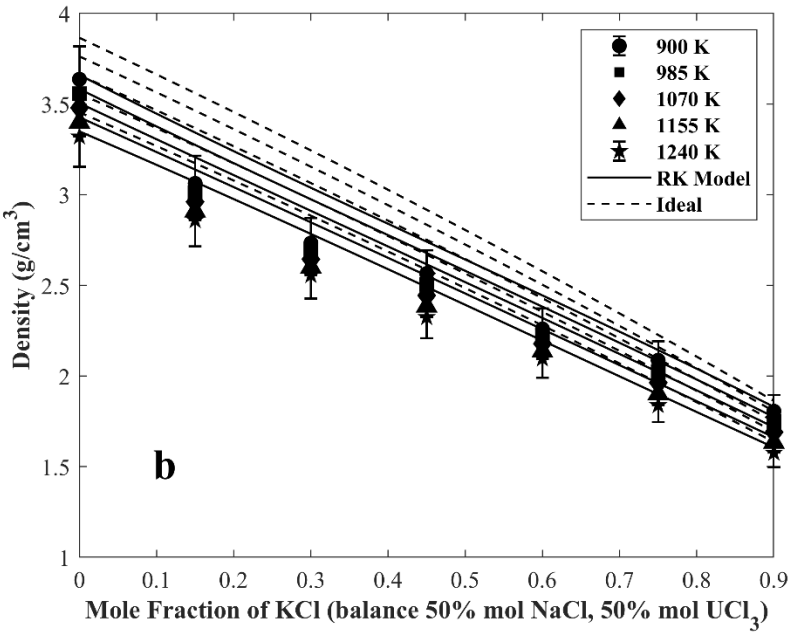


Fig. 4. Density comparisons using experimental data, ideal behavior and Redlich-Kister model (RK Model) in **a)** NaCl-UCl_3 (Desyatnik et al., 1975), and **b)** KCl-UCl_3 (Desyatnik et al., 1976) binary systems.


900 K
985 K
1070 K
1155 K
1240 K
RK Model
Ideal
Density (g/cm3)
a
Mole Fraction of KCl (balance 75% mol NaCl, 25% mol UCl3)


900 K
985 K
1070 K
1155 K
1240 K
RK Model
Ideal
Density (g/cm3)
b
Mole Fraction of KCl (balance 50% mol NaCl, 50% mol UCl3)

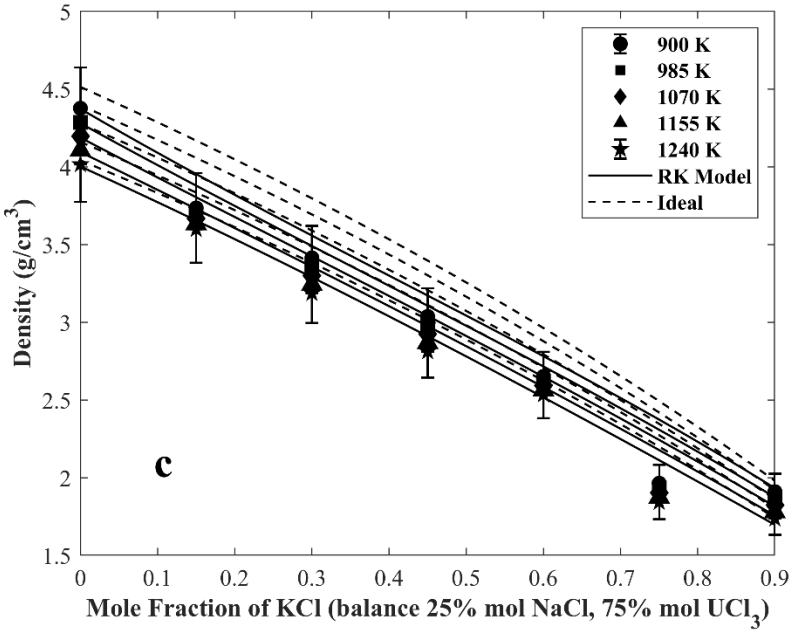


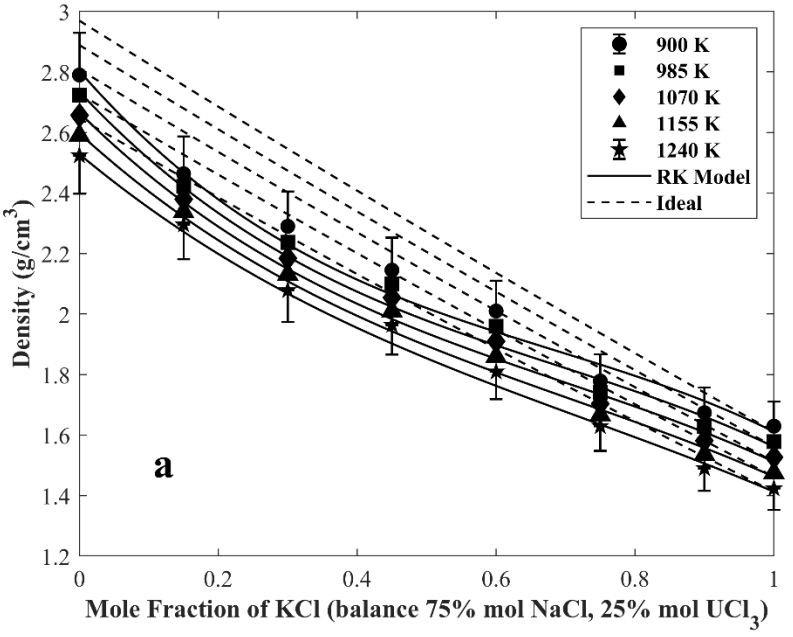
Fig. 5. KCl-NaCl-UCl_3 ternary density interpolations between KCl- **a)** 75% mol NaCl, 25% mol UCl_3; **b)** 50% mol NaCl, 50% mol UCl_3; **c)** 25% mol NaCl, 75% mol UCl_3. Experimental scatter data is from Katyshev et al. (1983). The error bars show 5% uncertainty from the experimental data.

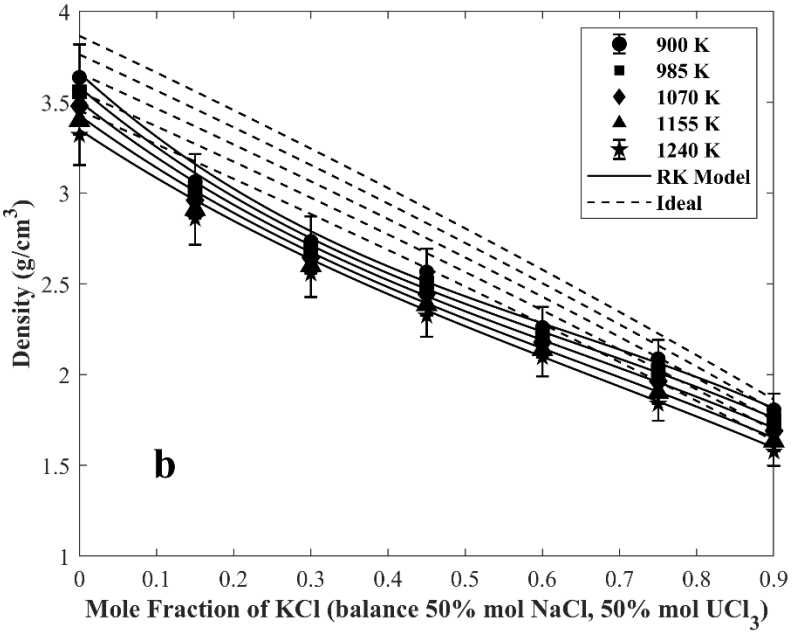
The relatively poor fit to the density of NaCl-KCl-UCl_3 molten salts imply a need for ternary interaction terms in the non-ideal part of the density of mixing. In order to accommodate the ternary interaction affects, we added a first order temperature dependent term, $\boldsymbol{\rho_{ex\_tern}}$, as shown in Equation 6 which will be the last non-ideal mixing term in Eqn. 5 in the modeling.

$$\boldsymbol{\rho_{ex\_tern} = x_A x_B x_C (A_t + B_t T)} \qquad \text{Equation 6}$$

A_t and B_t are ternary interaction parameters in NaCl-KCl-UCl_3 liquid mixture. Further fit calculations resulted in A_t = -13.96 and B_t = 0.0080. With the addition of the ternary term, the error from reference data improved to be 3.6%. The parameter A_t is almost an order magnitude higher than the binary interaction parameter A_j. This is due to ternary multiplication of mole fractions, which are less than 1. Nonetheless, the density at x_{KCl} = 0.75 in Fig 6.c had the similar error (17.8%) to the binary interpolation results. If one would like to have a more accurate model, it is best to include more complicated ternary interaction terms or a different model. For the

engineering design purposes, both Muggianu binary interpolation results and results with ternary interaction parameter will provide necessary information.





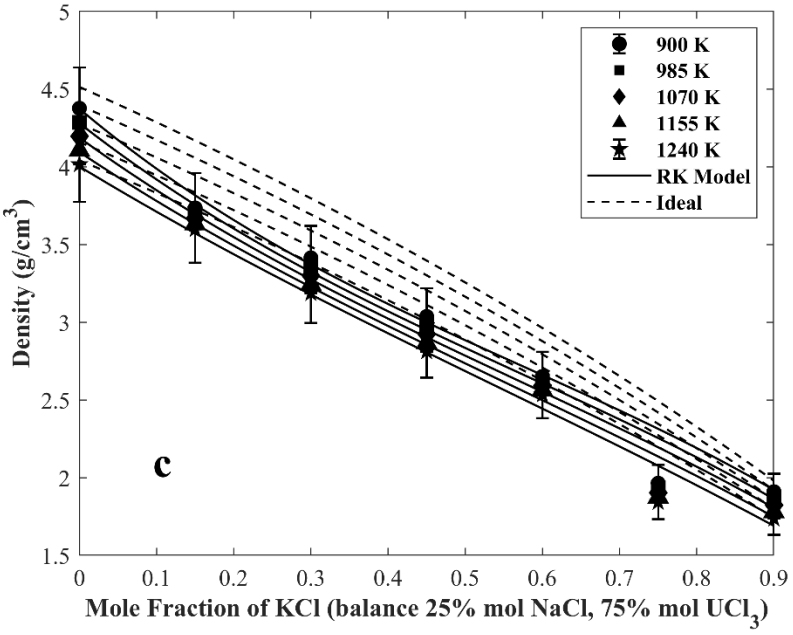


Fig. 6. KCl-NaCl-UCl_3 molten salt mixture ternary density between KCl- **a)** 75% mol NaCl, 25% mol UCl_3; **b)** 50% mol NaCl, 50% mol UCl_3; **c)** 25% mol NaCl, 75% mol UCl_3 after addition of the ternary interaction term in the non-ideal mixing term. Experimental scatter data is from Katyshev et al. (1983). The error bars show 5% uncertainty from the experimental data.

Quality Assurance (QA) of the physical properties required for supporting safety and licensing of MSRs is a critical component. An extensive literature review has been conducted revealing that for each individual system presented, the data originates from a single research groups over limited compositional space. Furthermore, generally no mention of QA is presented. While this does not preclude the use of the data-sets reproduced and modeled in this work, it does require application of a QA assessment. One strategy is experimental corroboration between research groups. It is therefore recommended that adequate, supporting measurements should be made over matching temperature-composition space for each system to validate the existing data. In addition, more expansive measurements will be required for a comprehensive experimental understanding of the fundamental pseudo-binary and pseudo-ternary systems as well as measurements for validated extrapolations into multicomponent space. These future measurements should be conducted with a documented QA program.

5. Conclusion

Molten salt density extrapolations have been performed for the KCl-NaCl-YCl_3 and KCl-NaCl-UCl_3 systems using the Redlich-Kister model. The density of molten salt mixtures is composed of an ideal and a non-ideal part modeled with temperature and composition dependent Redlich-Kister polynomials. The nonideal density has been characterized with binary system interaction parameters. These interaction parameters have been calculated in the binaries of the NaCl-KCl-YCl_3 and NaCl-KCl-UCl_3 systems using regression methods and used in the density extrapolation for the pseudo-ternary systems. While NaCl-KCl, NaCl-YCl_3 and KCl-YCl_3 binary and NaCl-KCl-YCl_3 ternary molten phases exhibit ideal density behavior, NaCl-UCl_3, KCl-UCl_3 and NaCl-KCl-UCl_3 molten salts show some negative deviation from ideality. The density estimation procedure using Muggianu interpolation method achieved overall agreement within 11 % of the experimental densities available in the literature. The agreement improved within 4% within the reference because of the addition of ternary interaction parameter.

This work represents the first in a series of publications to demonstrate the approach and associated uncertainties for extrapolating into multicomponent space the thermophysical properties of MSR relevant molten salts based on the pure and pseudo-binary systems. The parameters and models will constitute the MSTDB. The overall goal is to use the MSTDB for modeling thermophysical and thermodynamic behavior of multicomponent salts as a tool to aid in making licensing decisions as well as the design and operation of MSRs. There is currently work in progress towards viscosity, thermal conductivity and heat capacity estimation employing the method outlined here as well as a companion effort to develop the thermodynamic database MSTDB-TC.

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