

immediate

Predicting thermodynamic and thermophysical properties of molten chloride salts from
ab-initio molecular dynamics simulations

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1 Introduction

2 Case study: Ab initio molecular dynamics simulations of select thermophysical and thermodynamic properties of NaCl, UCl_3 and NaCl- UCl_3 molten salts

2.1 Review of existing experimental and modeling studies on NaCl, UCl_3 and NaCl- UCl_3

Experimental characterization of UCl_3 densities have been reported by Janz et al. [1] and Desyatnik et al. [2]. The density correlations derived from these two experimental data sets deviate significantly from each other. Desyatnik et al. [2] also investigated the density of NaCl- UCl_3 mixtures. They identified a negative deviation from an ideal solution across most of the composition range, with the high-temperature UCl_3 region deviating slightly from this trend. Maatsura et al. [3] measured the mixing energies of NaCl- UCl_3 at 1173 K, which highlighted a negative deviation from ideal solution behavior, which implies an exothermic reaction, with a shallow minimum of -0.07 to -0.08 eV per formula unit close to the eutectic composition ($x_{\text{UCl}_3} \approx 0.35$), but perhaps slightly shifted towards the 50-50 composition. There are two Calphad assessments of the NaCl- UCl_3 system [4, 5]. The magnitude and shape of the NaCl- UCl_3 mixing energies are noticeably different between the two assessments [5]. The assessment by Benes et al. [4] arrived at a form with a minimum close to the eutectic composition ($x_{\text{UCl}_3} \approx 0.35$), while Yin et al. [5] used the experimental data due to Maatsura et al. [3] as input, which resulted in a more negative mixing energy that is shifted slightly closer to the 50-50 composition. The magnitude of the mixing energy is off by about 50% between the two assessments.

The sparse and sometimes contradictory experimental data on molten salts in general and the NaCl- UCl_3 system in particular provide justification for pursuing modeling and simulations as a complementary approach to gain improved understanding. This opportunity is already acknowledged in the literature. Molecular dynamics simulations based on both classical potentials and AIMD simulations have been used to study chloride salts involving actinides [6–9]. Li et al. [6] used AIMD simulations to study the local structure of UCl_3 , UCl_4 and mixtures of UCl_3 , UCl_4 and NaCl at 1173 K. This study showed good agreement with experiments for the radial distribution function of the first coordination shell and identified network formation of UCl_3 units in mixed NaCl- UCl_3 salts [6]. In order to study temperature dependent thermo-physical properties, a semi-empirical potential was developed [7]. This potential successfully predicted density, thermal conductivity and viscosity, though validation is challenged by the lack of experimental data. Nam et al. [8] studied the solution thermodynamics of dilute concentrations of UCl_3 in a base salt and investigated the properties of base salts for different Van der Waals interaction

models. Song et al. [9] performed AIMD simulations of densities and transport properties in a LiCl-KCl eutectic salt with a small concentration of UCl_3 .

In the present study, AIMD simulations relying on different models for the Van der Waals interactions were used to predict temperature dependent thermophysical (density) and thermochemical (mixing energy and heat capacity) properties of NaCl, UCl_3 and NaCl- UCl_3 . The standard PBE exchange-correlation potentials typically used were extended to include a Hubbard U model for the actinide 5f electrons. The purpose of the study was first to determine with what accuracy fundamental properties can be predicted with AIMD simulations for actinide containing salts, second to populate some of the data gaps that exist in the literature and third to provide understanding of the link between coordination chemistry and properties.

The next sections are organized as follows. The methodology is described in Sec. 2.2, followed by results and discussion in 2.3. First the benchmark for NaCl is presented, next the UCl_3 results are reviewed, which is followed by NaCl- UCl_3 mixtures. The connection of our results to local chemistry and pair distribution functions are discussed in Sec. 2.4.

2.2 Methodology

The AIMD simulations were performed with the VASP code [10]. The simulations used a range of supercell sizes with the largest consisting of 216 (NaCl), 216 (UCl_3) and 134-184 (NaCl- UCl_3 mixtures) atoms. The smallest cells for the same systems contain 64, 64 and 60-72 atoms, respectively. For NaCl and UCl_3 these were created by expansion of the crystalline unit cells followed by melting of the lattice by performing high temperature MD simulations. The mixed supercells were created by replacing Na ions with U ions or vice versa, in some cases additional molecular units were added to ensure that a suitable number of atoms were maintained in the supercells. The differently sized supercells were investigated in order to understand and optimize the compromise between computational efficiency, enabling sampling in the time domain, and accuracy with respect to long range interactions. The radial distribution function (RDF) was used as the most basic measure of the adequacy of the supercell size, with convergence with respect to the targeted thermophysical and thermodynamic properties following suite. All the supercells investigated properly capture the expected radial distribution function in the liquid state. This behavior is exemplified in Figures 2.1a) and 2.1b) for two UCl_3 and NaCl supercells of different size, respectively. The smaller cells predict essentially identical radial distribution functions to the larger cells, but come with improved computational efficiency. Although the larger cells may still be more accurate for, e.g., mixed salts exhibiting more complex radial distribution functions, the situation in mixed salts is further complicated by the need to sample sufficient configurations to resolve the preferred short and intermediate range distribution of ionic species in network forming salts such as UCl_3 [6]. This requires fairly long simulation times. Proper sampling is easier to achieve in smaller supercells given the computational cost of AIMD simulations, even though the radial distribution itself as well as other properties may be better described in a larger supercell. For this reason, our production runs tend to use supercells of intermediate size. Based on the verification against large supercells for the radial distribution function above and further examples in Sec. 2.3, the results in the present study are considered sufficiently converged with respect to supercell size.

All simulations used the Γ point for integration in reciprocal space. The accurate simulation setting was utilized in VASP, but the plane wave cut-off energy was increased above the standard setting to 400 eV. Gaussian smearing with a smearing parameter of 0.05 eV was used for the partial occupancies of the wave functions. The convergence criteria for the electronic minimization was at least 10^{-3} eV for NaCl and 5×10^{-3} eV for salts containing uranium.

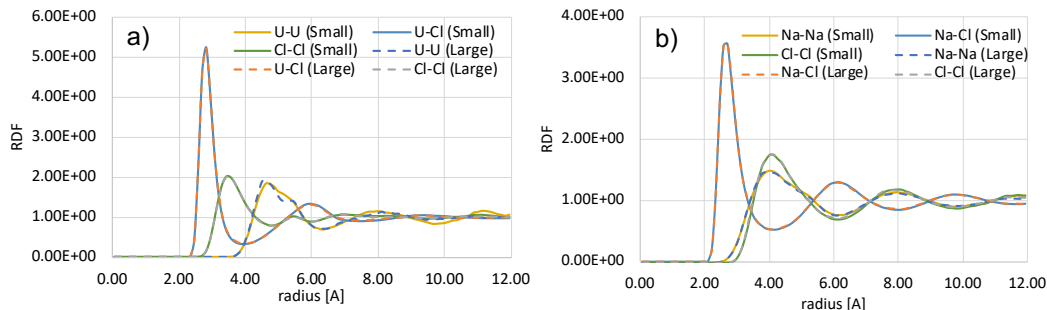


Figure 2.1: The predicted radial distribution function in a) UCl₃ and b) NaCl obtained by supercells containing 64 (small) and 216 (large) atoms at 1250 K.

The Projector Augmented Wave (PAW) method was used to describe the core electrons [11, 12]. For each element, the PAW potentials supplied with VASP for the PBE exchange-correlation potential were utilized. For Na, the version that only includes the s electron(s) in the valence shell was used. The PAW potential for Cl also included p electrons and for U it included the outer s, p and f electrons in the valence shell. Calculations for U ions were performed with a Hubbard U term, in order to capture the impact of accounting for strong electron correlation effects. The Lichtenstein approach [13] was used for the Hubbard U methodology. An approximate U value range of 3.0-4.5 eV was determined by using scoping constrained DFT linear-response method for crystalline UCl₃ [14]. The J value was set to 0.51 eV. These values are similar to those proposed for UO₂ [15]. After confirming that the values for UCl₃ were close to those for UO₂, the values for UO₂ were adopted in the present study. Future work may consider further optimization of the Hubbard U (and J) parameters, but the results and conclusions are not expected to significantly change based on this choice, as long as the value is sufficiently large to ensure an insulating ground state. It should be noted that the effective U value depends on the coordination environment and consequently could differ between crystalline UCl₃ and molten salts. It could also be a function of time in the simulations as the environment may change. Future work may consider these questions in more detail, but it is beyond the scope of the present study. The effect of the Hubbard U parameter for molten uranium chloride salts is the same as for crystalline UO₂; without the U parameter the salts are predicted to be metallic, which is contrary to the expected behavior. Even though useful results may certainly be obtained while ignoring the strong correlations captured by the Hubbard U model and accepting the resulting metallic character predicted for the salts, there are limitations to this approach. For reference, a few simulations were performed without the Hubbard U model (not shown).

Uranium ions in its 3+ state have localized magnetic moments. In order to mimic the disordered arrangement of magnetic moments expected at temperatures where the salts are molten, the spins were arranged in an anti-ferromagnetic (AFM) pattern and then allowed to relax during

the simulations. In the context of molten salts, the AFM option is similar to a random distribution with a total magnetic moment in the supercell close to zero. The predicted properties are not strongly influenced by the magnetic ordering.

It is well-established in the literature that Van der Waals or dispersion interactions are critical for reproducing the density of molten salts by DFT methods [6, 8, 16]. Previous simulations have used both the DFT-D3 method [6, 17] and the Langreth & Lundqvist [8, 18, 19] methodologies for various molten chloride salts. Both methods were used in the present study, although the Langreth & Lundqvist method was dropped after concluding that it was not very accurate for pure UCl_3 . In addition, the density-dependent energy correction (dDsC) method [20, 21] was used with the goal of improving accuracy. The dDsC method does not include parameters for f elements in the standard VASP version. In order to enable simulations the corresponding parameters were taken from Ref. [22].

The AIMD simulations for the molten salt supercells were performed using isobaric conditions (NPT) conditions. The primary intent of the NPT simulations (with the pressure set to zero) is to evaluate density, thermal expansion, heat capacity and mixing energy. All NPT ensemble simulations used the Langevin thermostat in VASP. For the NPT simulations, the temperature friction coefficient was set to 10 ps^{-1} and the friction coefficient for the lattice degrees of freedom to 1 ps^{-1} . The time step was set to 2 fs for production runs between 1000 K and 1500 K. Around and below 1000 K a larger time step, up to 5 fs, is warranted due to the slow dynamics of uranium ions. In principle, the larger time step can also be applied at higher temperatures as long as the structures have been properly converged.

The simulations used pre-equilibrium and equilibration runs that involve melting the lattice and ensuring convergence of the total energy and pair distribution function for the temperature of interest. After pre-equilibration and equilibration, production runs follow for at least 20 ps. Some simulations used longer production runs, in particular this applies to the simulations based on smaller supercells (up to 50 ps) and systems containing a mixture of UCl_3 and NaCl (up to 40 ps). NPT simulations often require long equilibration runs and may sometimes leave the equilibrium state due to distortions of the supercells. All simulations were inspected to avoid sampling such regimes.

Properties were calculated by averaging over the production run (not including the equilibration or pre-production time). Densities were trivially obtained from the supercell volume and heat capacities from the slope of the total internal energy (E_{tot}) as function of temperature.

$$C = \frac{\partial E_{tot}}{\partial T} \quad (2.1)$$

For the NPT ensemble, this corresponds to heat capacity at constant pressure (C_p). Mixing energies were calculated from the potential energy (E_{pot}) of the mixed salt with pure NaCl and UCl_3 at the same temperature as the reference.

$$E_{mix} = E_{pot}(\text{U}_x\text{Na}_y\text{Cl}_{3x+y}) - xE_{pot}(\text{UCl}_3) - yE_{pot}(\text{NaCl}). \quad (2.2)$$

2.3 Results

2.3.1 Ab initio molecular dynamics simulations on NaCl

Density and structure

Figure 2.2a plots the predicted density of molten NaCl as function of temperature for the DFT-D3 and DFT-dDsC Van der Waals models as well as simulations without any dispersion interaction. The results refer to the supercell size that we consider best converged for each methodology, see caption and below for further discussion. A correlation derived from experimental data is also shown [1]. All simulations reproduce the temperature dependence of the density obtained from experiments. However, as expected, only the simulations that account for dispersion interactions are within 10% of the experimental density correlation. The best agreement is obtained for the dDsC dispersion model, which is within 5% or less of the experimental correlation across an extended temperature range. The calculated (dDsC) correlation for the density as function of temperature is listed in Table 2.1. The radial pair distribution function at 1250 K is reported in Figure 2.1, which highlights first, second and third-shell coordination distances of 2.70 Å, 3.78 Å and 4.14 Å, respectively. The predicted pair distribution function is in good agreement with experimental values [23].

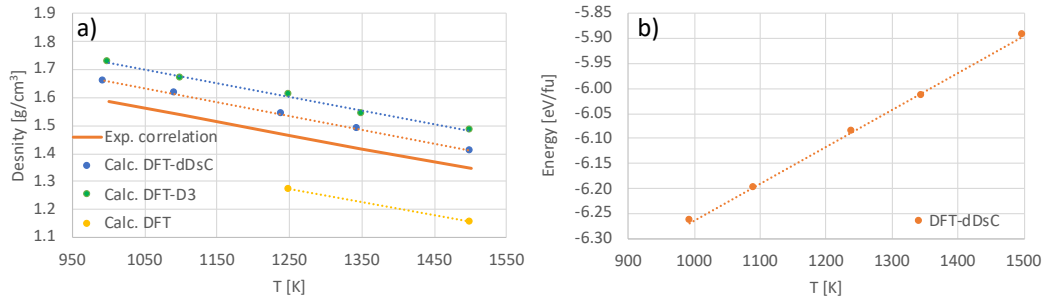


Figure 2.2: a) Density of NaCl predicted with two different models for dispersion forces (D3 and dDsC) and one without (small cell for dDsC, large cell for others). Experimental data is represented by the correlation plotted as an orange line [1]. b) Calculated total energy of NaCl as function of temperature (small cell). The line is a least-squares fit to data points and the slope represents the heat capacity.

	Density (g/cm ³)	Heat capacity (J/mol/K)
NaCl Calculated (dDsC)	$2.1594 - 0.0004993T$	71.6
NaCl Experiment	$2.061 - 0.0004759T$ [1]	66.9 [24]
UCl ₃ Calculated (dDsC)	$6.1066 - 0.001383T$	145.3
UCl ₃ Experiment	$6.375 - 0.001522T$ [2]	150 [25]

Table 2.1: Calculated and experimental correlations and values for density and heat capacity of NaCl and UCl₃.

Heat capacity

Figure 2.2b plots the total energy per formula unit of molten NaCl as function of temperature. The derivative equals the heat capacity of NaCl, which is also tabulated in Table 2.1 together with an experimental reference value [24]. The results refer to the supercell size that we consider best converged for each methodology, see caption and below for further discussion. The simulation results indicate a constant heat capacity, though in order to identify small deviations from this behavior a denser temperature mesh would be required. The good agreement between simulations and experiments for the heat capacity (see Table 2.1) further emphasizes the accuracy of the AIMD simulations.

Impact of supercell size and other simulation settings

The results in Figure 2.2 refer to simulations based on the supercell that we consider best converged. Figure 2.3 compares these results with those obtained from other supercells. Density and heat capacity are accurately represented by all supercell sizes in the temperature range investigated. These results suggest that for NaCl, the larger supercell is not required to achieve converged results for density and heat capacity at moderate temperature, which is consistent with the discussion of radial distribution functions in Sec. 2.2. The small variations between supercells are more likely related to sampling differences, rather than due to the supercell size. Consequently, the converged results in Figure 2.2 as well as the correlations in Table 2.1 are representative of all simulation results in this study, regardless of the supercell size.

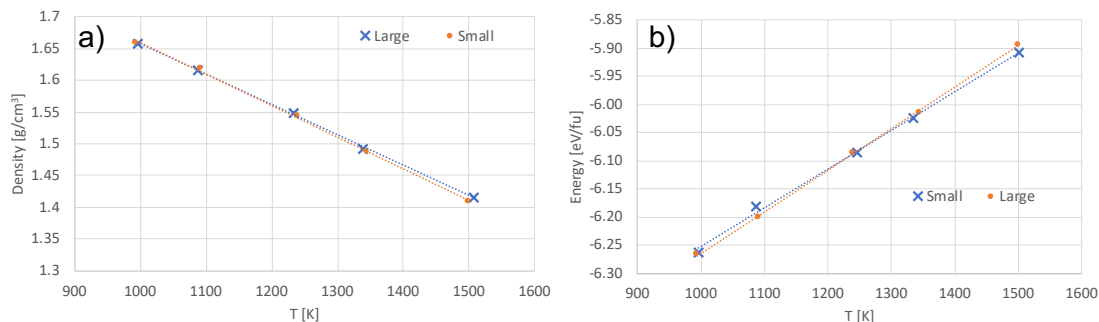


Figure 2.3: a) Calculated density and b) total energy of NaCl as function of temperature for the large and small supercells. The lines are least-squares fits to data points and the slope represents the heat capacity.

2.3.2 Ab initio molecular dynamics simulations on UCl_3

Density and structure

Following the results for NaCl, the best performing Van der Waals model (dDsC) was applied to UCl_3 . Figure 2.4a) plots the predicted density for UCl_3 as function of temperature for the dDsC dispersion model. These results refer to the supercell size that we deem to be best converged and

most representative of the UCl_3 system, see below for further discussion. The figure also compares the predicted densities to two literature correlations derived from experiments [1, 2]. The two experimental correlations for density are surprisingly different and cannot both be correct, except in a narrow temperature range. The temperature dependence is predicted to be close to linear across the full temperature range investigated and agrees very well with the experimental data due to Desyatnik et al. [2]. The densities in Figure 2.4 were fitted to linear correlations and summarized in Table 2.1.

The radial pair distribution function at 1250 K is reported in Figure 2.1, which highlights a first-shell coordination distance of 2.82 Å. The coordination distance is in excellent agreement with the experimental values of 2.82 Å [26] and 2.84 Å [27] measured at 1113 K and 1200 K, respectively, while it is higher than the AIMD simulations by Li et al. [6], which can likely be ascribed to the application of the Hubbard U methodology in the present study. The predicted coordination numbers are within the 6 to 8 range reported in experiments and previous simulations [6, 26, 27].

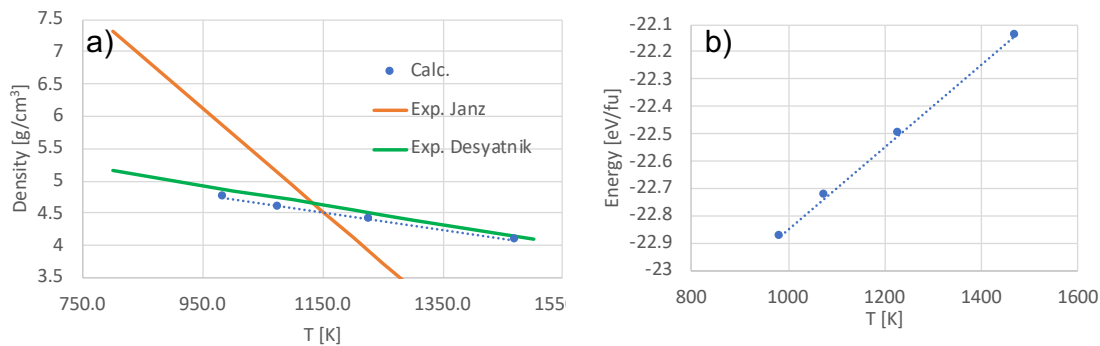


Figure 2.4: a) Density of UCl_3 predicted by the dDsC model for the dispersion forces (small cell). Experimental data is represented by the correlations plotted as green [2] and orange lines [1]. The dashed line is a least-squares fit to the calculated data points, the equations of which are summarized in Table 2.1. b) Calculated total energy of UCl_3 as function of temperature (small cell). The line is a least-squares fit to data points and the slope represents the heat capacity.

Heat capacity

Figure 2.4 plots the total energy as function of temperature, from which the heat capacity can be derived by calculating the slope. The total energy closely follows a linear relation as function of temperature and, consequently, the heat capacity can be approximated as a constant in the temperature range investigated. In order to resolve small deviations from the linear relation, a denser temperature mesh would have to be used. Experimental data for the heat capacity of UCl_3 has not been identified, but our results compare very well with the value derived from the MSTDB CALPHAD assessment of the UCl_3 thermodynamics (see Table 2.1) [25].

Impact of supercell size and other simulation settings

The results discussed above for UCl_3 refer to simulations using the supercell size that we deem to be best converged and most representative of the UCl_3 system for each simulation methodology. Select results obtained from different supercell sizes are compared in Figure 2.5, which exhibits fairly good agreement with each other. Any difference between supercell sizes is primarily ascribed to sampling appropriate configurations rather than an effect of increasing the numbers of ions in the simulation box, although additional simulations would be required to fully certify this conclusion. Compared to NaCl it is much more challenging to reach long simulation times for the large UCl_3 simulation cells.

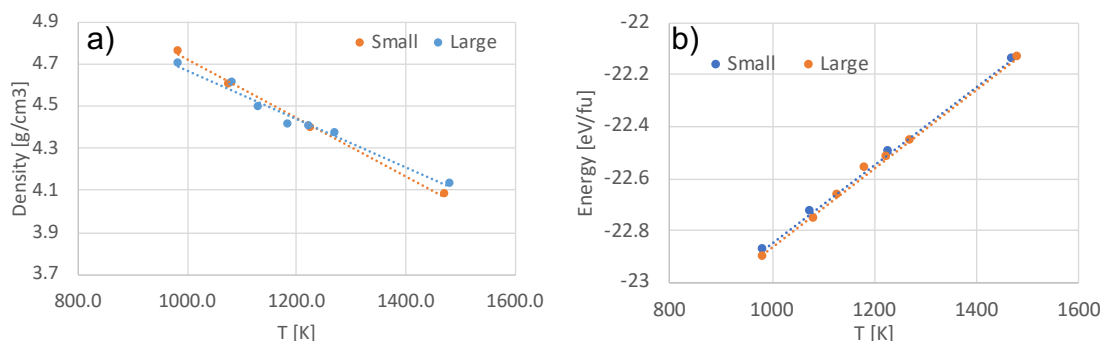


Figure 2.5: Calculated density and total energy of UCl_3 as function of temperature for the large and small supercells. The line is a least-squares fit to data points and the slope represents the heat capacity.

2.3.3 Ab initio molecular dynamics simulations on NaCl-UCl_3

Density and structure

The density of NaCl-UCl_3 mixtures were calculated for the dDsC dispersion model at three or four (depending on composition) different temperatures between 800 K and 1500 K, as shown in Figure 2.6a). This figure also includes densities at the same temperatures as those of the simulations obtained from correlations derived from experimental data due to Desyatnik et al. [2]. Figure 2.6b) highlights the fractional deviation from ideal solution behavior as calculated from simulations and experiments [2]. It is challenging to converge the density for mixed salt solutions to an accuracy better than around one per cent of the absolute density using AIMD simulations, which gives rise to some scatter in the data points. Nevertheless, a few trends are discernible from Figure 2.6. The simulated data points are within a few per cent of the experimental data. The simulations suggest a negative deviation from an ideal solution (lower density than predicted by an ideal solution behavior) by up to 2-3%, except close to pure UCl_3 at high temperature where a positive deviation is observed. According to our simulations, the magnitude of the deviation from ideal solution behavior is a function of composition and varies some with temperature starting at $x_{\text{UCl}_3} \approx 0.35$ and continuing in the UCl_3 rich composition range, while it is almost independent of temperature in the NaCl rich range. The maximum deviation from ideal

solution behavior occurs close to the eutectic composition of 35% UCl_3 . These predictions are qualitatively similar to the correlations derived from experiments by Desyatnik et al. [2], though the experimental correlations predict a larger magnitude for the deviation from an ideal solution and also exhibit a stronger temperature dependence than the simulations.

Figure 2.7a) plots the density as function of temperature for each composition, which emphasizes a close to linear temperature dependence, similar to the pure end-members. The coefficients describing the linear dependence on temperature for each composition are plotted in Figure 2.8a). Although there is some scatter, a weak non-linear dependence on composition for both the linear and constant term (not shown) is identified. The non-linear dependence is most pronounced in the UCl_3 rich range. The density correlations identified in Figures 2.7a) and 2.8a) can, in principle, be used for calculating densities at temperatures and compositions not explicitly investigated by AIMD simulations.

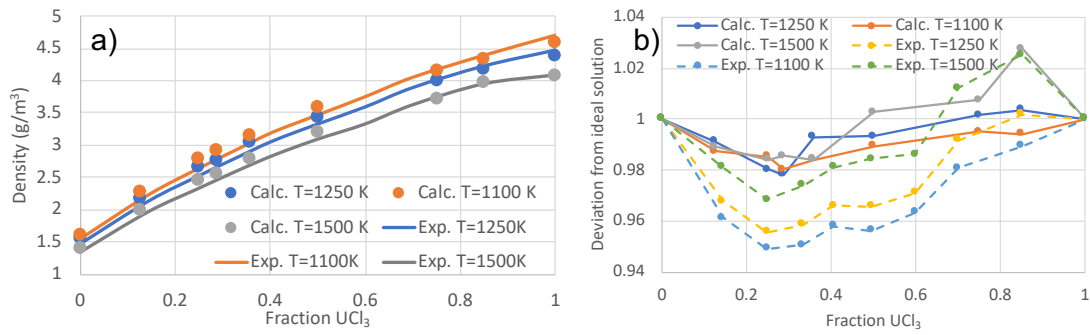


Figure 2.6: a) Density of NaCl-UCl_3 mixtures as obtained from simulations and experimental data [2] at temperatures between 1100 K and 1500 K. b) The fractional deviation from ideal solution behavior plotted as function of composition at temperatures between 1100 K and 1500 K.

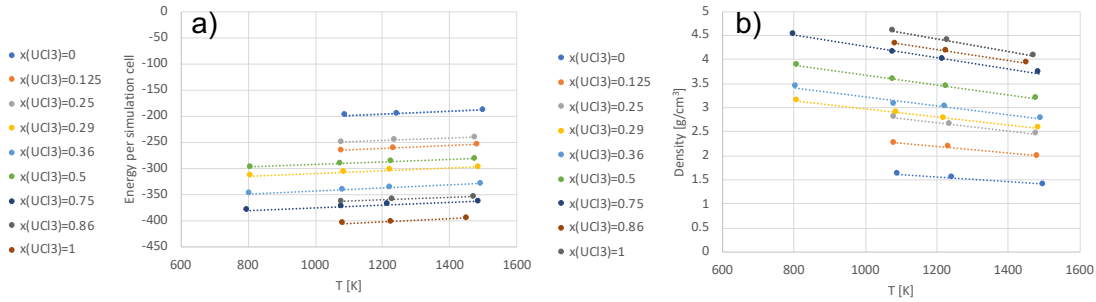


Figure 2.7: a) Calculated temperature dependent densities as function of composition. b) Calculated energies per simulation cell as function of composition. In both a) and b) the lines represent least-squares fits to the calculated data.

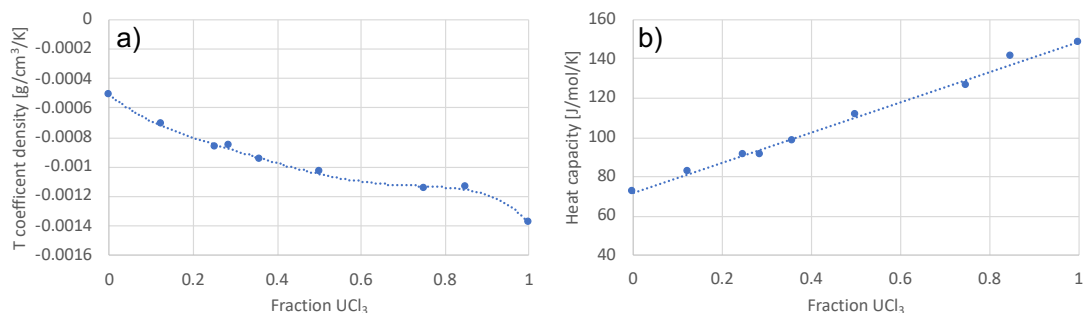


Figure 2.8: a) Coefficient describing the linear temperature dependence of density as function of the UCl_3 fraction. The line represents a least-squares fit of a fifth order polynomial. b) Heat capacity as function of the UCl_3 fraction. The line represents a least-squares fit of a linear correlation.

Mixing energy and heat capacity

The mixing energies of NaCl-UCl_3 at temperatures ranging from 1100 K to 1500 K are plotted in Figure 2.9a), with the NaCl and UCl_3 end members as reference points. The mixtures exhibit a negative deviation from ideal solution behavior, which implies that the solution phase is favored over a two-phase mixture of the end-points. Addition of entropy further stabilizes the mixed solution phase, as shown in Figure 2.9b) by adding a simple ideal solution model to the potential energy in Figure 2.9a). The minimum (most negative) mixing energy is between $x_{\text{UCl}_3}=0.35$ and $x_{\text{UCl}_3}=0.5$, which qualitatively mimics the results for the density.

The mixing energy was measured at 1100 K by Matsuura et al. [3] The results are also shown in 2.9a) and indicate very good agreement with the simulations across the full temperature range. In addition to the experimental data points, there are two sets of thermodynamic models for the solution energy [4, 5], see Figure 2.9a) for the correlation from Ref. [5]. Both thermodynamic models assume the solution energy to be independent of temperature, which is in good agreement with the simulations. The model by Yin et al. [5] was derived from the experimental measurements by Matsuura et al. [3] and consequently agree similarly well with our simulation results. The model by Benes et al. [4] exhibits a smaller mixing energy than both the experimental data points and our simulations (not shown).

The total energy for each NaCl-UCl_3 composition is plotted as function of temperature in Figure 2.7b), from which heat capacity can be calculated as the slope, similar to the pure NaCl and UCl_3 systems. The heat capacity is a linear function of the salt composition, which is expected based on the lack of temperature dependence for the salt mixing energies. The negative deviation from an ideal solution seen for the mixing energy and density is not present for the heat capacity.

2.4 Discussion

This discussion section will focus on the behavior of mixed salts, in particular how the mixing properties (energy and density) relate to the evolution of the pair distribution functions

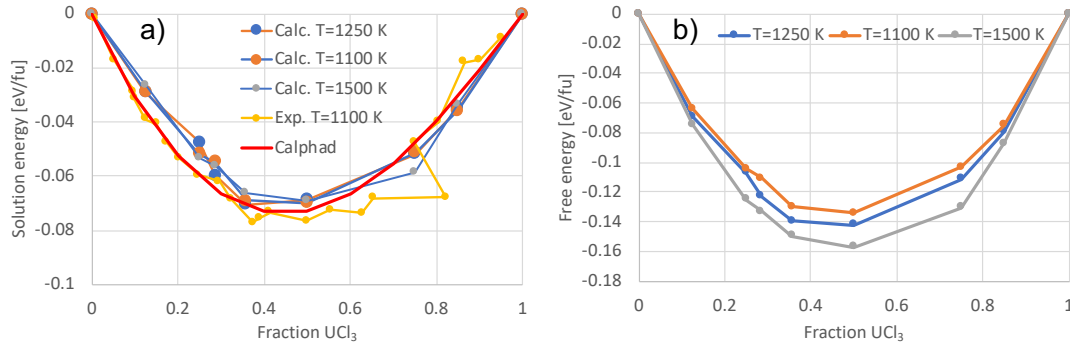


Figure 2.9: a) Mixing energies for NaCl-UCl₃ at 1100, 1250 and 1500 K. Pure NaCl and UCl₃ are used as references. The calculated results are compared to experiments [3] and a thermodynamic assessment [5]. Results are shown for the best converged supercells. b) The free energy of mixing at 1100 1250 and 1500 K assuming an ideal solution.

in the mixed salts. Figure 2.10 plots the pair distribution functions at four salt compositions ($x_{UCl_3} = 0.25$, $x_{UCl_3} = 0.29$, $x_{UCl_3} = 0.36$ and $x_{UCl_3} = 0.50$) and compares them to the reference pair distribution functions for UCl₃. Ref. [6] already showed that as NaCl is added to pure UCl₃, the number of Cl ions in the first neighbor shell around U ions increases, from 6 in UCl₃ to almost 8 close to NaCl, with a corresponding decrease around Na ions. This is also confirmed in the present study (see the increase of the first peak in Figure 2.10 as the NaCl fraction increases). This redistribution of Cl ions clearly represents a favorable interaction as the mixing energy is negative across the full composition range. The same study also identified networks of UCl₃ units above a fractional UCl₃ concentration of 0.30, and more isolated units below this concentration. This behavior is also visible in the pair distribution functions. The U-U pair distribution function for mixtures with a composition equal to or above a fractional UCl₃ concentration of 0.36 maintain the same shape and height as in pure UCl₃ (they essentially overlap), which emphasizes the importance of network formation in the mixtures. Below a fractional concentration of 0.36 the U-U pair distribution function rapidly deviates from the pure UCl₃, indicating an inability to maintain the favorable network structure. This behavior is visible in the third coordination shell in Figure 2.10. Related changes may be observed in other distribution functions. The location of this transition coincides with the minimum in the mixing energy and the maximum deviation of the density from an ideal solution (compare Figures 2.10, 2.6 and 2.9). In turn, these coincide with the eutectic composition according to the experimental phase diagram [5]. Combined with the evolution of the U-Cl and U-U pair distribution functions in the mixtures, this suggests that the negative mixing energy is driven by increases in the Cl coordination around U ions, but if the fractional UCl₃ concentration is below ≈ 0.36 , the gain from increasing this U-Cl coordination is countered by not being able to maintain the favorable U-U coordination seen in UCl₃, as evidenced by the break-up of the network structure. The balance of the increase in the U-Cl coordination and decrease in U-U coordination as function of the composition of NaCl-UCl₃ salts is responsible for the minimum in the mixing energy and by extension the location of the eutectic point in the phase diagram.

The negative deviation from an ideal solution exhibited by the density implies that the vol-

ume increases. Often, a negative mixing energy is associated with a stronger bonding and lower volume, clearly that is opposite to what is observed for NaCl- UCl_3 . The reason for increased volume and decreased density is again related to the evolution of the pair distribution function. The increased coordination number of Cl around U ions means that the bonding environment starts to resemble that of U^{4+} ions in UCl_4 , which may also be what drives the favorable interaction. The density of UCl_4 is noticeably lower (and the molar volume higher) than that of UCl_3 , which we believe correlates with the negative deviation from an ideal mixture for the density of NaCl- UCl_3 mixtures. Note that this analogy does not imply presence of formal U^{4+} ions in NaCl- UCl_3 , but a partial transition that drives the evolution of both the mixing energy and density. Additional simulations and experiments would be required to prove this hypothesis.

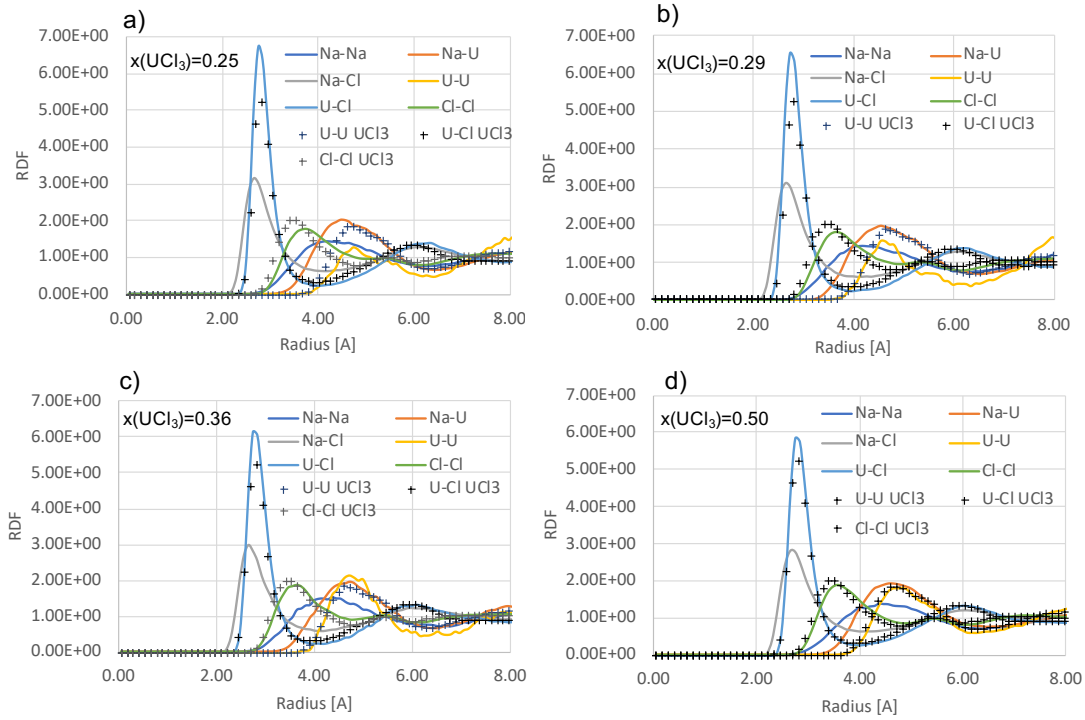


Figure 2.10: Pair distribution functions for NaCl- UCl_3 mixtures as function of composition ($x_{\text{UCl}_3} = 0.25$, $x_{\text{UCl}_3} = 0.29$, $x_{\text{UCl}_3} = 0.36$ and $x_{\text{UCl}_3} = 0.50$). The reference U-Cl and U-U pair distribution functions for UCl_3 are plotted in each figure for comparison.

3 Summary and conclusions

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