First, we would like to thank the reviewers for their insightful feedback and comments. We have addressed them in the revised version of the manuscript and we believe that they have led to substantial improvements of the manuscript. Below, we summarize our response to each question/comment. We mark the reviewers’ questions in blue and our responses in red. Changes to the manuscript have been highlighted in red in the marked-up version of the resubmission.

Reviewer #1: This is a quite interesting work given how difficult DFT calculations of uranium in liquids are. However, there are issues. I believe that this study can be a good addition to Journal of Nuclear Materials once the my comments listed below are addressed.

We appreciate the reviewer’s feedback. The questions are answered below, which we hope satisfactorily addresses the concerns raised.

A. Methodology

1. I would move most of the results in this section to a supporting information file or to the result section of the main text.

This is a good suggestion. In the revised manuscript we have moved the “Supercell Size” and “Application of the Hubbard U term” subsections of the Methodology to the Results section, with exception for roughly the first paragraph in each of the sections, which is kept to define the methodology used in production runs. The headings of the subsections were updated/removed to conform with the new structure. The changes to the Methodology and Results sections are marked in red in the revised manuscript. However, note that only the location of the paragraphs has changed and the text remains essentially identical.

See manuscript for detailed changes.

2. My biggest concern is that, the threshold for the convergence electronic minimization of 10^-3 eV is too large.

The reviewer correctly identified that the convergence criterion for the total energy in each self-consistency loop is on the high-end, however, we performed several tests for this setting and the results indicate that the properties derived from the simulations do not change in a meaningful way by increasing the criterion to, e.g., 10-4 eV. As an example, the density of NaCl at T=1250 K was converged to within <0.5% when comparing results for 10-3 eV and 10-4 eV. Similar results were obtained for other samples and properties. Based on our tests, the uncertainty introduced by sampling of the configuration space is larger than that introduced by the convergence criterion currently used for the self-consistency cycles, in particular for the constant pressure simulations. For these reasons, we are confident that the current simulation settings are sufficiently converged for the properties investigated. As a note, the NVT vdW-DF2 simulations already use a tighter convergence criterion, but tests for that methodology also came to the conclusion that going from 10^-3 to 10^-4 or higher does not significantly change the predicted properties of interest. To conclude, our tests show that the simulation settings currently use give sufficiently converged results, but at the same time we agree with the reviewer that future studies should consider using a tighter setting to be on the safe side. As is often the case, this choice is a matter of balancing computational efficiency with accuracy. In the process of verifying the simulation settings, some data points were slightly refined based on sampling range from the original submission, but these changes are all very minor and well-within the uncertainty already specified. Figures 2, 4, 6, 8, 9, as well as Table 1 and 2 were updated accordingly.

3. Please specify the valence electrons (for example, Na 2s^2 2p^6 3s^1 )

This information was provided in written form in the original version of the manuscript, but it has now been updated to include the explicit valence electron configuration.

Sentences changed (original text in black and changes in red, see manuscript for exact typesetting): For Na, the pseudopotential version that only includes the s electron(s) in the valence shell (3s1) was used with the dDsC and DFT-D3 methodology, while the version that also includes the semi-core p electrons (2p63s1) was utilized for the vdW-DF2 simulations. Although different choices were made for the simulations with regards to the Na potential, the results are not sensitive to this choice at a scale that impacts the conclusions of the present study. The PAW potential for Cl also included p electrons (3s23p5) and for U it included the outer s, p, d, and f electrons (6s26p65f36d17s2) in the valance shell.

4. Fig 1:  Usually the range of r in the g(r) plot is smaller than 1/2 of the size of unit cell vectors. It seems that a larger range is used in the case of the 64-atom unit cell.

The reviewer is correct that a larger range than half of the unit cell vectors were was used for the small-cell plots. While the g(r) plot was generated with periodic boundaries, removing the tail to zero, it is more appropriate to only utilize the single supercell to construct the g(r). In order to satisfy the requirement expressed by the reviewer, the range has been reduced to 7 Å for all plots (Figs. 1 and 13).

5. I am not sure if I understood the effect of the +U correction on the density and other properties of the molten salts. Moreover, for a given value of +U, how did the authors decide to use AFM or FM? How was +U chosen for each system? Please specify +U for each system and each dispersion correction approach. Are results in Fig 3 calculated using 1 frame or multiple frames (ensemble average)?

Adding the U parameter very slightly increases the volume of the salt and reduces the density, which was already mentioned in the manuscript. More specifically, the difference in the density between the case with U= 0 eV (FM) and the optimized value of U=4 eV (AFM) is approximately 0.5%. For clarity, this information has been added to the revised manuscript. The impact of the U parameter on, for example, mixing energies and heat capacities has not been investigated. Both AFM and FM orderings of the magnetic moments were used for each U value in the targeted investigation. As specified in the manuscript, the lowest energy was obtained for the AFM ordering when the salt was predicted to be an insulator (high U values) and for the FM ordering when the U value was sufficiently low to predict metallic behavior. If the AFM state is assumed to represent a random distribution of spins, that state would be expected to be favored for all U values at high temperature due to entropy effects. After identifying the optimal/preferred U value of 4 eV and AFM magnetic ordering, these choices were used in all simulations, regardless of the approach applied for the dispersion correction. We have added a statement to clarify this assumption. The density of states in figure 3 are ensemble averages. A clarifying statement was added to the figure caption.

Changes to the manuscript: 1 (in reference to the choice of Hubbard U parameter): These settings were used for all systems, except where explicitly stated otherwise. 2 (in reference to the density of states): The density of states are ensemble averages over 200 steps.

6. In the heat capacity calculation the authors assumed that the heat capacity is temperature independent. Please justify.

The total energy as function of temperature, from which the heat capacity was calculated for each system, was predicted to follow a linear relation within the accuracy of the simulations. This implies that the heat capacity is approximately constant. Identifying small deviations from a linear relation would require a denser sampling as function of temperature and likely stricter convergence criteria than used in our simulations.

Changes to the manuscript (original text in black and changes in red, see manuscript for exact typesetting): The simulation results indicate a constant heat capacity (the total energy depends linearly on temperature within the accuracy of the simulation results), though in order to identify small deviations from this behavior a denser temperature mesh would be required.

B. Results

1.  Table1. Please specify the temperature range (for both theory and experiment)

This information has been added for all cases where it is available to us, see manuscript for updates. The caption was modified accordingly: Where known, the temperatures within parenthesis indicate the range of data used for fitting the models from either AIMD simulations or experiments.

2. Section 3.2.2. How well the heat capacity data agree with results reported by Li et al. (<https://doi.org/10.1016/j.molliq.2019.112184>)?

In the composition range where Li et al. performed simulations of the heat capacity (x(UCl3)<0.55) the agreement is very good. The corresponding data points, calculated from the expression derived by Li et al., have been added to Figure 8 and the caption has been modified accordingly: Data points calculated from the heat capacity relation derived by Li et al. \cite{Li2020} from semi-empirical MD simulations are included for comparison, but only for the composition range where the relation was fitted (x(UCl3)<0.55). A similar statement was included in the text: Data points calculated from the heat capacity relation as function of UCl3 composition derived by Li et al. \cite{Li2020} from semi-empirical MD simulations are included for comparison, but only for the composition range where the relation was fitted (x(UCl3)<0.55). The agreement between the two simulation data sets is very good.

Note that the expression due to Li et al. does not capture the behavior beyond x(UCl3)=0.55 very well, which is probably to be expected as the expression was not fitted to any data points in that range. For this reason, data points for x(UCl3)>0.55 were not used for the comparison in Figure 8.

3.  For transport properties of NaCl-UCl3, given the lack of experimental data, the agreement between computational studies is important. How well the computed results in this work agree with results from the machine learing potential MD study by Nguyen et al. (<https://doi.org/10.1021/acsami.1c11358>) and polarizable ion model potential MD study by Li et al. (<https://doi.org/10.1016/j.molliq.2019.112184>)? The diffusion coefficient of ~ 10^-6 (m^2/s) is too large (Fig 12). Fig 11 shows that for a range of UCl3 mol fraction, the diffusion coefficient of U at 1400K is lower than that at 1100K, I believe that error bars should be added to the transport data.

You are correct that the diffusion coefficient in Fig 12 was too large. This was a total diffusion coefficient of the supercell, and not scaled per molecule/species. This has been rectified. A comparison between the work of Li and Nguyen has been included for comparison, as stated below:

In the absence of such experimental data, comparisons must be made to existing computational studies. While there are no AIMD investigations into diffusion in the NaCl-UCl$\_3$ system, two prior studies utilizing classical molecular dynamics with a polarizable ion model interatomic potential (Li) \cite{Li} and a machine learning interatomic potential (Nguyen) \cite{nguyen\_ML} have studied diffusion in the NaCl-UCl$\_3$ system. While neither of these studies included the composition and temperature ranges explored within this work, there are general comparisons that can be made. Li showed the same relative diffusion pattern (D$\_{Na}$ $>$ D$\_{Cl}$ $>$ D$\_U$), while Nguyen predicted Cl diffusion to proceed more quickly than Na diffusion. Li provided diffusion as a function of temperature at two compositions (5 mol\% and 50 mol\% UCl$\_3$), showing that diffusion decreases and the activation energy increases with increasing UCl$\_3$ content, in agreement with this work. However, Li did not provide pre-factors or units in their Arrhenius plots, making direct comparisons difficult. However, all of the qualitative trends in this work are consistent with that of Li. Nguyen only studied the eutectic composition, and only at temperatures below 1100 K. Comparing the data at their maximum temperature (1070 K) and our minimum temperature (1100 K), all of the diffusion coefficients, including individual species and total diffusion, are within a factor of four, indicating excellent agreement. Additionally, Nguyen determined the activation energy of the eutectic composition to be 0.39 eV, which is similar to our calculation of 0.3 eV. While the qualitative trends in species diffusion differ between this work and that of Nguyen, the quantitative values generally agree quite well.

Finally, while there are no compositions at which diffusion at 1100 K is larger than 1400 K, there are compositions at which diffusion at 1250 K is slightly larger than that at 1400 K. This is largely due to the limited diffusion of U in these systems, yielding higher variability in the diffusion data. Error bars have now been included which indicate the twice the standard error of the mean.

Reviewer #2: The paper describes ab initio molecular dynamics (AIMD) simulations of NaCl, UCl3 and NaCl-UCl3 molten salts. The introduction reviews previous MD simulations and experimental results of the same systems. It seems that the new results presented in this paper are in agreement with previous simulations and mostly in agreement with experiments. This work seems to differ in the details of the method, and so the work is new and worthy of publication. The connection of the MD simulation and physical properties is important (density, heat capacity, eutectic point) and seems well argued.

We appreciate the reviewer’s positive assessment.

Another interesting observation and discussion is related to possible phase separation between NaCl and UCl3 systems. If correct, I'd be very interested in consequences of  such phase separation. I am not yet fully convinced, though, given exceedingly small number of ions in the simulations.

The reviewer brings up an important limitation of our simulations. We have added a caveat stating that the limited size of the simulation supercells prevents us drawing strong conclusion regarding the partial phase separation. Note that we still believe that the observation is real, but more expansive simulations would be required to confirm the observation. Those are beyond the scope of the present study.

Changes to the manuscript: However, it is important to acknowledge that the small size of the simulation cells limits the ability to draw strong conclusions. More expansive simulations would be required for that purpose, which is beyond the scope of the present study.

The paper is written very well. I don't have any edits besides trivial things such as missing

"to" in the abstract "...in order assess..."

The missing “to” has been added.

I wish the calculated coordination numbers U-Cl were also summarized in the description of results and not just in the bulk of the text.

It is true that the coordination numbers could have been presented in the Results section, however we prefer to keep them in the Discussion section. If the reviewer feels like this is a poor approach, we would be happy to go back and update the manuscript. No changes were made to the manuscript.