We thank the reviewer for their time and effort in reviewing this manuscript. The feedback and comments we received allowed us to make this manuscript into a more robust and clear work. We also appreciate the different view on the topic that a fresh set of eyes allows and were glad to consider those comments and make changes.

Other major edits.

In figure 5, the data is no longer compared to the experimental data from Wang et al. as this data was from another paper and the AIMD method is no longer compared to the CMD method used in Wang et al. With these changes, their corresponding portions of the manuscript have been removed. “by Wang et al., but are slightly below the values calculated” and “Finally, a comparison can be made to the Born-Mayer-Huggins interatomic potential employed in the classical molecular simulations by Wang et al., which significantly underestimates the equilibrium density across the entire compositional range. This validates the use of DFT with vdW-DF2 van der Waals functional for the calculation of density in molten salt systems.”

*Reviewer #1: The experimental work included in this report is insufficiently detailed and has neglected to accurately represent the uncertainties in the method. I would recommend divorcing the experimental work from the computational work which is exemplary. When comparing the computational work to literature data it is essential to include the published uncertainties of those literature values, which may improve the perceived performance of your models. Detailed comments are included in an attached PDF as edits to the text and comments.*

*How is standard error calculated for a simulation that does not depend on experimental values? Is this analogous to a convergence coefficient of some kind?*

The standard error is the standard error of the sample mean of the calculated samples. Each individual simulation yields a unique result, and data analysis is performed on the produced data set. The following equation was used.

Standard error =

Where n is the sample size, is the sample mean and x is the sample. This method does not require experimental values and shows the precision of the data and does not show the convergence of coefficients. This was addressed in the paper by making it clear that it the standard error of sample mean by referring to it as so in the manuscript. “An example of the volume versus pressure relationship is illustrated in Fig. 2 for the 70% KCl system at 900 K with the standard error of the sample mean pressure shown as the error bars for each unique volume. Which shows the precision of the 5 unique simulations.”

*Were any of the materials cleaned before use to remove oxide layers which might dissolve into the molten salt, altering the composition? Furthermore, how was the salt composition measured and confirmed?*

We have added a clarification regarding material cleaning. “All materials were cleaned used DI water and isopropanol and baked at 393 K for 1 hour to remove any residual moisture. Additionally, the glassy carbon crucible was baked at 773 K for 4 hours to remove any adsorbed organic matter.”

*This equation (Eq. 7) neglects the effect of surface tension which has an appreciable effect of the measured density due to the wire transversing the salt surface. If you are going to neglect this effect you should at least state that, so that you can note that any values you measure are likely lower than reality. Ideally, you would measure the surface tension and correct for it.*

We have added the effect of surface tension into the paper’s data. These corrections were on the order of 0.1% of the density value, which is quite less than the error of the density value. Nevertheless, the effect of surface tension of the wire has been included.

*Was only one weight measurement made? Or did you make a series of measurements over time to ensure the system was at equilibrium without volatilization or other thermally induced motion in the system.*

We have added clarification regarding the multiple measurements that were made. “This tare and measure method was repeated for a total of three weight measurements at each temperature and the average of these measurements was taken for further density calculation.”

*Using a figure (fig 4) to compare to one data point from literature seems questionable. It would seem more useful to compare the model data to the literature data at the pressure at which it was measured and give a % difference value for each model method.*

The table has been added and the plots removed. The corresponding text has been adjusted to account for this modification.

*It seems important to note what the uncertainty is in the literature data (fig 5) to which you are comparing. If the uncertainty is larger than your deviation from it, your models may be better than previously thought. Show uncertainty for Janz data.*

Janz experimental data does not provide experimental error for all systems. In some cases, Janz states an error of 1%. For these systems, it is unclear if experimental error has been reported. A statement regarding this information has been included as follows: “ The density data from Janz does not provide the experimental error.”

*For Fig 7, why linear fits? the data clearly is not linear in many of the temperatures.*

A linear fit was used to show the overall trends between the different temperatures and is addressed in the manuscript by adding “The linear fits were only intended to be used to look at overall trends of the data and not to suggest that a linear fit suits the data the best.”

*For KCl at 1000 K: Do you mean you assumed for the sake of the model that it was a liquid, because in actuality it would clearly be a solid. The statement here seems to suggest that you determined (aka measured?) that it was still a liquid below its freezing point. If that is the case, you need to provide evidence.*

Given the initialization as a liquid, the small amount of subcooling, and the short time scale of picoseconds, the system would remain a liquid and can be treated as a liquid in a metastable state. This radial distribution function was analyzed to confirm the retention of a liquid structure. This was addressed in the manuscript by adding “This is due to the low level of subcooling and the short time period. Radial distribution function was analyzed to confirm the retention of a liquid structure”

*For Table 1: Was there no analysis of fit with these equations? Uncertainty analysis is crucial to any experimental effort.*

We have added the r2 values for these lines of fit.

*Notably absent is any independent measure of concentration of these salt mixtures, discussion of salt preparation procedures, and trace impurity analysis. How can you be sure you are measuring what you think you are measuring if you don't verify your salt compositions.*

Response…

*Reviewer #2: This paper reports a combined experimental and theoretical study of the thermophysical properties of LiCl-KCl molten salt system. The authors have performed ab initio molecular dynamics simulations with different methods to incorporate van der Waals interactions, and the results compare favorably with their new experimental measurements. I recommend this paper for publication in Journal of Nuclear Materials after some minor revisions, see comments below:  
  
1) In Fig. 1, the partial radial distribution function plot for K-K pair exhibits multiple maxima. It is not clear if they are all physical or due to the small size of the computational cell used in AIMD simulations.*

This is caused by a combination of the small supercell and the short time period as this is only a snapshot and not a time-averaged structure. This issue was muted by smoothing the trajectories over 200 time periods to generate something closer to a time-averaged structure. The figure and text have been updated to reflect this change.

*2) In Fig. 4, AIMD simulations using SCAN functional suggest that the pressure of liquid KCl is almost independent of density, which is clearly unphysical. Such discussions of the origin of this anomaly will be useful.*

The SCAN functional was predicting a significantly higher density than the other dispersion options, and thus was displaying a negative pressure for all densities explored. This manifested as a seemingly independent behavior with density, but this is simply due to the fact that it is a liquid system in a large volume, leading to negative pressures. If smaller volumes (higher densities) were explored, the SCAN functional would indeed show a density-dependent pressure. Since this illustration is misleading of the true behavior, and per the suggestion of reviewer #1 to translate the figure into a table, this data point is removed and addressed by adding, “SCAN is excluded from the KCl data as the predictions were far outside the expected bounds and not pursued further.” *3) The definition of Cp in Eq. 3 is incorrect. Since Cp is the first derivative of enthalpy with respect to temperature, it is a generally temperature-dependent property. The limit of Cp as temperature tends to zero Kelvin will be different from Cp values at high temperatures.*

The equation was corrected from saying temperature approaches zero to incremental temperature approaches zero (ΔT). We apologize for the error.