REVIEWER #1

1. In the Computational Methods section there is no mention whether or not the non-spherical contributions related to the gradient of the density was considered (i.e., LASPH). In addition, what value was considered for the LMAXMIX parameters?

The LASPH comment was addressed by adding “The non-spherical contributions within the PAW spheres were considered.” The LMAXMIX was addressed by adding “The LMAXMIX parameter in VASP was increased to 6 to account for the f-orbitals.”

2. The details related to the methodology are well presented. However, it would be more beneficial if the authors would consider specifying which systems have 64 and/or 72 atoms.

The number of atoms in the NVT simulations were added and are 200, 184, 198, 192, 190, 192, and 160 atoms for the NaCl, 15%, eutectic, 50, 67, 85, and PuCl3 systems, respectively. We also clarified the number of atoms in the NPT simulations by adding that NaCl, PuCl3, and 6.67%PuCl3 have 64 atoms, 28.57%PuCl3 had 72 atoms, and 35.71%PuCl3 had 76 atoms.

3. Following the previous note, how many atoms would the next bigger system contain had the authors considered doing calculations for? And, how much difference would they expect for the results in doing so?

This was somewhat addressed in the previous study which the authors performed and which is cited in the manuscript. The influence of size was evaluated by investigating a system with 64-72 atoms and a system with 216 atoms, which showed that there was little change in the radial distribution function, density, and heat capacity. This finding is now emphasized in the manuscript. To address the question more specifically, technically systems can be explored on an added formula unit basis, e.g., 4 additional atoms for a single additional PuCl3 unit. The larger system consisting of approximately 200 atoms is a significant number of additional molecules and is already a large number of atoms for AIMD molten salt studies, as many are conducted with less than 100 atoms. The maximum size which would still be feasible for a study of this scope is approximately 250 atoms, but it is expected that the results would minimally change and the extreme increase in computational cost would not be worth the very minor improvement in accuracy.

4. For the discussion related to Figure 3, how would your results compare to the NaCl-UCl3 from previous DFT study?

A comparison is now made in the text to the UCl3 RDF and bond lengths. “This is a comparable bond distance observed in UCl3 which has a U-Cl bond distance of 2.80 A. The PuCl3 system has longer bond distances than UCl3 for all bond types but they are comparable in length with a difference of 4.97%.”

5. In few parts of the discussion, the authors only discuss the results and the observation, and not necessarily the reasoning behind what they are observing. For instance, "While the vdW-DF2 functional has the lowest error from the experimental correlation, it does not capture the temperature dependence of the density as well as the dDsC, with the vdW-DF2 underestimating this dependence". In addition, what is the reason that the behavior between different functionals are different in Figure 2, and similar cases throughout the paper.

The manner in which the different vdW formalisms address the vdW forces fundamentally differs. The vdW-DF2 replaces the exchange-correlation functional while the vdW corrections (D3 and dDsC) add a correction to GGA-PBE. This can explain why the latter two have similar slopes as the main mechanism doesn’t change, while in the vdW-DF2 the different functional predicts a different slope. Refer to the answer to question 6 for additional comment.

6. In Table 1 results for vdW-DF2 is underestimating Cp for NaCl, but the opposite trend is seen for PuCl3, and there is no discussion regarding this in the text.

This is referring to the fact that vdW-DF2 has a larger predicted value than dDsC for PuCl3 but not NaCl, however, there is no experimental data, thus we do not know if one is over/under predicting, or if both vary in the same direction. However, the relative differences can be considered. There are many potential causes for this difference. One potential cause is how the vdW formalism handles electrons. specifically, f-electrons. In the example mentioned above NaCl does not have f-electrons while PuCl3 does. As the reasons for the differences between different vdW formalisms are not well understood, the exact reason behind the difference is not known. However, one should take into account the fact that certain vdW corrections may more appropriately handle certain systems and/or properties within those systems. This is similar to, but more dramatic than, how different GGA formulations (PW91, PBE, PBEsol, revPBE, etc.) can produce different results which compare better or worse with experiments depending upon the system.

7. It would be better to use less extreme wordings (i.e., excellent agreement), and instead, substitute with "good agreement" etc.

The extreme wording has been modified and excellent has been replaced with the word good. We now believe that the qualitative nature of the comparative descriptions accurately reflects the results.

8. Some convergence plots/data from AIMD calculations need to be included in Supplementary Materials/Supporting Information.

The convergence study for the DOS and the ENCUT parameter in VASP have been added to a supplementary materials document. Additionally, the total energies of different compositions as a function of temperature have also been included.

9. Typo in the abstract "For exist for"

The typo was corrected.

10. Consider removing URL from the the references as the might change in some years and will not be useful.

The URLs were removed from the references.

11. Consider forcing upper case in your bib file. lif instead of LiF, etc. This has been seen in some references such as 3, 4, 10, 26, 35, 36, 37, 38, etc.

The elements in the references have been forced to be uppercase.

REVIEWER #2

These comments do not apply to this manuscript, but instead refer to a different report. Thus, the comments have not been addressed.