Supplementary Materials for: 'First-principles investigation of the thermophysical properties of NaCl, $PuCl_3$, and NaCl-$PuCl_3$ molten salts'

Kai Duemmler[a], David Andersson[c], Benjamin Beeler[a,b,*]

[a] *Department of Nuclear Engineering, North Carolina State University, Raleigh, NC 27695*
[b] *Idaho National Laboratory, Idaho Falls, ID 83415*
[c] *Materials Science and Technology Division, Los Alamos National Laboratory, P.O. Box 1663. Los Alamos, NM, 87545, USA*

1. ENCUT optimization

The selection of the cutoff for the plane-wave basis set was optimized. The default value is around 260 eV and larger values were tested including 400 and 500 eV. The pressure as a function of the volume curve can be seen in Fig. 1 and shows that even 400 eV is insufficient and there is a difference between 400 and 500 eV pressure curves. This can also be seen in the densities as they do not fall into each other error bars with 400 eV having a density of 4.26 ± 0.041 g/cm^3 and 500 eV having a density of 4.17 ± 0.059 g/cm^3.

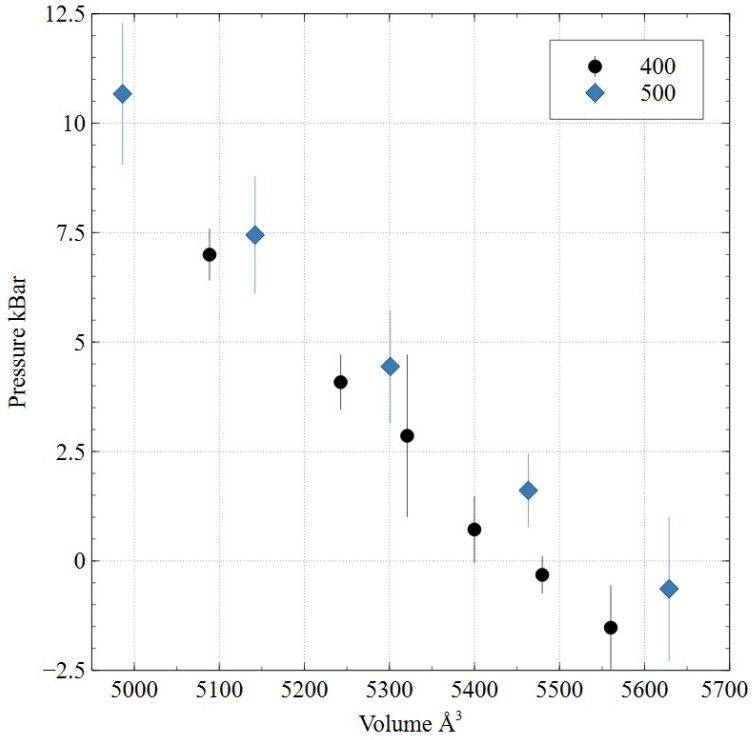

Figure 1: The pressure as a function of volume curve for different cutoff values for the plane-wave basis set.

*Corresponding author
Email address: bwbeeler@ncsu.edu (Benjamin Beeler)

2. Density of States

The density of states (DOS) is shown in Fig. 2 for $PuCl_3$ for a Hubbard U value of 0 eV and 5 eV. For a value of 0 eV, the Fermi level is occupied and a conduction band is present, while there exist additional states available at higher energies. As the U value is increased on the f electrons, peak shifting occurs, driving electronic states to lower energies, eventually opening up a bandgap. The bandgap opens and stabilizes at a U value of 5 eV, indicating a bandgap of 1.4 eV. While the actual bandgap of $PuCl_3$ is unknown, it is known that this system should exhibit a non-zero bandgap. Similar behavior was observed for the eutectic NaCl-$PuCl_3$ salt. Additional experimental information on the electronic structure of these molten salts would allow for a refinement of the Hubbard U value. Minor fluctuations in the DOS curves are due to the reduced accuracy imposed on these convergence tests, and minor changes in the bandgap may be observed with higher accuracy DFT simulations.

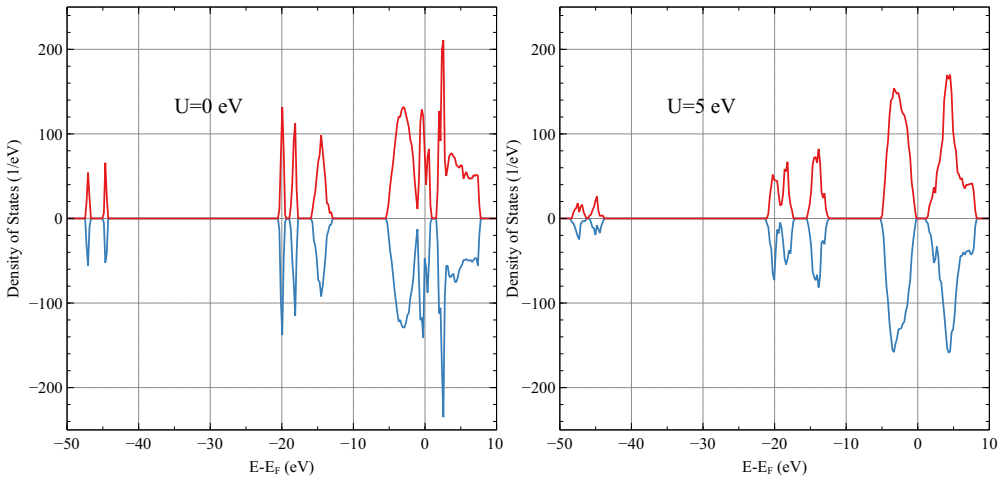

Figure 2: The electronic density of states of $PuCl_3$ for a Hubbard U value of 0 and 5 eV.

3. Total Energies

The total energies of the NVT simulations are shown in eV/mol in Fig. 3. They are fit to a line and all follow a nearly perfect linear behavior. This allows for the heat capacity to be calculated over this temperature range as a constant value.

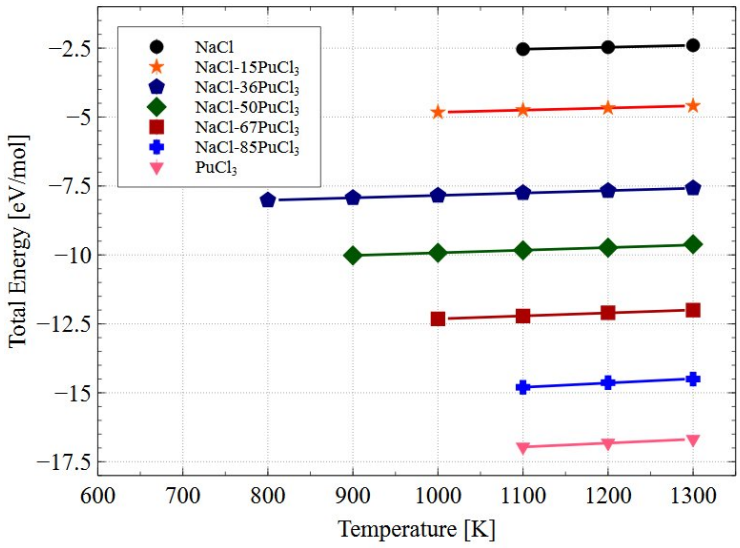

Figure 3: Total Energy of the nvt simulations. Lines are linear fits to the energy.