Materials Science and Technology Division
MST-8, Materials Science in Radiation and Dynamics Extremes
Los Alamos National Laboratory
PO Box 1663, MS G755
Los Alamos, NM 87545
505-665-8621, email: andersson@lanl.gov

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Dear Editor:

We hereby submit our manuscript "Ab initio molecular dynamics (AIMD) simulations of NaCl, UCl_3 and NaCl-UCl_3 molten salts" for publication in Journal of Nuclear Materials.

Molten salt reactors (MSRs) are among the advanced concepts currently pursued by industry, e.g. TerraPower, Moltex and Terrestrial, and supported by research and development activities at national laboratories and in academia. However, the basic concept is not new and was first developed as part of the effort to power aircrafts with nuclear energy in the 1950's. Later in the 1960's, Oak Ridge National Laboratory (ORNL) built and operated the Molten-Salt Reactor Experiment (MSRE). This reactor used a fluoride salt with uranium as fuel. Fluoride salts are still highly relevant and proposed in several designs. In addition, chloride salts are being considered for MSRs operating in the fast neutron spectrum. Our manuscript focuses on chloride salts, in particular the NaCl-UCl_3 system, which is one of the primary fuel salt candidates. To re-emphasize the last point, this paper targets a uranium bearing fuel salt meant to provide the fissile material in a molten salt reactor, which we believe falls into the scope of Journal of Nuclear Materials.

Property data for chloride salts are in many cases limited and sometimes of unknown accuracy. This is especially true as actinides (U, Pu), impurities, and corrosion and fission products are added. Using atomic scale simulations to fill these data gaps and to provide mechanistic understanding of property relations would facilitate more accurate evaluation of various concepts by reactor designers, developers and other interested parties. Modeling and simulations have an important role to play in reducing data gaps, because the compositional space of interest is extensive and difficult to cover with experiments alone, especially since some of the salts are also highly toxic and radioactive. Further, modeling enables a deeper understanding of the chemistry underlying the salt properties.

In the present study, Ab Initio Molecular dynamics (AIMD) simulations relying on different models for the Van der Waals interactions were used to predict temperature dependent thermophysical (density, compressibility, and diffusivity) and thermodynamic (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. This extension has not previously been utilized for studying actinide salts. The purpose of the study was first to determine with what accuracy fundamental properties can be predicted by 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. Our study extends AIMD simulations to predict advanced temperature dependent properties of actinide containing molten salts for MSR applications. The corresponding knowledge and understanding are important for a range of scientists and engineers involved in developing MSRs as well as the supporting salt technology and structural materials. For these reasons, we believe that our manuscript will be of high interest to the readers of Journal of Nuclear Materials.

Thank you for considering our manuscript for publication.

Sincerely yours,

David Andersson

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