Accelerating the Development of Reliable and Robust Machine Learning-Based Interatomic Potentials

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ABSTRACT:

To develop advanced molten salt technologies, the physical properties and thermochemical behavior of molten salts must be known over a wide range of chemical environments. However, experimentation with molten salt is often complex due to the handling of toxic and radioactive substances, which greatly increases the cost and time for data acquisition. Thus, there is a need to develop predictive models to radically advance our understanding of molten salt chemistry. Most classical atomistic models today are incapable of capturing charge ordering, charge transfer and are inaccurate for multi-element systems. Ab initio molecular dynamics (AIMD) simulations can generate chemical data from first principles but are limited to short simulation times and small systems, which are insufficient for obtaining statistically converged properties. Recent breakthroughs in representing the interatomic interactions with neural networks provide a means to overcome these size and accuracy limitations. However, the development of neural network-based interatomic potentials (NNIPs) can be a lengthy process, potentially requiring up to hundreds of thousands to millions of ab initio calculations to train a robust NNIP for a single system. Further, while neural networks interpolate well between data, extrapolating beyond atomic configurations that differ from the training set can cause unexpected failures in neural network models.

Studies in the last couple of years have used NNIPs to reproduce properties for simple molten salts under limited conditions. However, the transferability of NNIPs has not been demonstrated for broad application or optimizing salt compositions. The objective of this proposal is to accelerate the development of reliable NNIPs by addressing the key challenge of transferability. In our recent works, we showed that robust NNIPs can be developed using highly diverse datasets for various chloride and fluoride salts. Here, we will extend this work by efficiently generating representative datasets using active learning to rapidly explore areas of chemical space while limiting redundancies in ab initio calculation. A model will be developed for predicting NNIP uncertainty, which will identify potential inaccuracies during simulation. This will greatly enhance the applicability of NNIPs to new systems with many components. Further, the uncertainty model will be able to relate real atomic features (e.g., local density, species coordination, etc.) to NNIP failure modes, which will provide guidance for generating smart training datasets and enable a physically-informed understanding of NNIP uncertainties. This will improve their versatility, which will allow accelerated chemical exploration. Further, these methods can be applied to other types of machine learning algorithms, providing method-agnostic benefits. NNIPs will be generated to fill important gaps in molten salt property databases for relevant salts such as NaCl-$MgCl_2$-UCl_3-$PuCl_3$ (fast reactor fuel salt), LiF-BeF_2-ThF_4-UF_4 (thermal reactor fuel salt), and ZrF_4-based salts. Using NNIPs, important properties such viscosity, heat capacity, thermal conductivity, and volatility will be calculated over a range of compositions and thermodynamic conditions, enabling the validation and development of new models for predicting properties in multi-component mixtures.