Machine-Learning-Accelerated Molecular Dynamics Approaches for Molten Salts

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ABSTRACT:

The proposed work will develop new machine learning potential (MLP) approaches and new MLPs to enable rapid prediction of molten salt properties with near ab initio quantum mechanical accuracy. Molten salts have potential use as both coolants and fissile material solvent in the molten salt reactor advanced reactor concept, considered among the most promising designs for future reactor technologies. In this project we focus on FLiBe (LiF-BeF2) and NaCl-MgCl2 salts, which are leading candidates for fissile material solvent in molten salt fast and breeder reactors. Flouride and chloride salts also represent the two most widely explored molten salt reactor salt families, providing opportunities for validation against extensive existing data and opportunities for significant impact on molten salt reactor development. Some of the properties of fluoride and chloride salts that are critical to the safe and practical operation of a nuclear reactor include viscosity and thermal conductivity, as they control how the salt flows and how it transports heat when supporting fissile materials or cooling the reactor, melting temperature, as it is essential to assure the salt remains liquid under operating conditions and understand its freezing behavior under accident conditions, and impurity redox potentials, as these play a dominant role in corrosion, one of the major challenges with molten salt technologies. These properties are often quite difficult to measure, particularly their coupling to impurity content, as controlling salt impurities is extremely challenging. For this reason there is strong interest in using molecular simulations to predict these and similar thermophysical properties. Traditional approaches include ab initio molecular dynamics (MD), which can treat all properties in theory but often cannot practically model the above properties due to computational limitations, and interatomic-potential MD, which has the necessary computational speed for the above properties but cannot be practically fit to accurately model the complex physics involved. MLPs can potentially overcome these limitations by providing a practical approach to achieve orders of magnitude speedup over ab initio MD with comparable accuracy. The objective of this work is to develop MLPs and use them in MD to calculate thermal conductivity, viscosity, standard potentials, and melting temperatures for FLiBe (LiF-BeF2) and NaCl-MgCl2 salts, focusing on modeling pure salts and effects of Cr, C, U, and O impurities. Since MLPs have multiple forms and the best approach for molten salts is not yet clear, we will explore the leading methods to determine which is most accurate and efficient to train and use for simulation. Furthermore, present MLPs require very large ab initio calculation training databases and treatment of many-component salts will be quite challenging. We therefore propose to develop and refine existing uncertainty quantification methods and integrate them with active learning and on-the-fly training approaches to greatly reduce the amount of human and computing time needed to train the MLPs. Finally, we will also develop a new MLP approach that allows sub-alloys to be combined without their refitting, a key capability to accelerate modeling of salts with many components and impurities. We will then use the optimized approaches we develop to study the above properties, providing valuable thermophysical data, particularly on the effects of impurities and their couplings with each other (e.g., how redox potentials depend on oxygen content). The impact of this work will be to (i) establish the most effective present MLP technique for molten salt property simulation, (ii) augment this MLP technique with accelerated fitting through uncertainty quantification, active learning, and on-the-fly fitting, enabling practical modeling of many-component systems such as salts with impurities, (iii) establish a new MLP that can efficiently tackle many component systems and provide very fast simulation, both critical needs for molten salts studies, and (iv) develop a database of important thermophysical properties of FLiBe (LiF-BeF2) and NaCl-MgCl2 salts, focusing on properties and effects of impurities. Together these impacts will enhance the present understanding of impurity effects in FLiBe (LiF-BeF2) and NaCl-MgCl2 salts, and greatly expand the range of salt properties that can be practically simulated from molecular modeling to support dramatically more extensive data generation and understanding in the near future.