DOE-ID NEPA CX DETERMINATION
Idaho National Laboratory

CX Posting No.: DOE-ID-INL-21-011

SECTION A. Project Title: Machine Learning Interatomic Potentials for Radiation Damage and Physical Properties in Model Fluorite Systems

SECTION B. Project Description and Purpose:

Irradiation-induced defect production and subsequent microstructure evolution can significantly influence key properties of nuclear fuel, such as fission gas diffusivity, chemical redistribution, and thermal conductivity. Unfortunately, atom-scale computational studies of nuclear fuels containing correlated 5f electrons remain challenging due to the high computational cost of accurate density functional theory (DFT) calculations and the poor reliability of classical molecular dynamics (MD) simulations using empirical potentials (EPs). Recently, machine learning interatomic potentials (MLIP) have emerged as a powerful paradigm-shifting tool for materials modeling. By directly learning from big DFT datasets, MLIP can reach new levels of realism in terms of accuracy and transferability, far surpassing those of EPs, and at the same time remain orders of magnitude faster than DFT. The objective of this project is to harvest the recent theoretical advancements in MLIP and apply them to develop high-accuracy, highly-transferable interatomic potentials for studying the influence of radiation damage on physical properties of calcium fluoride (CaF2) and uranium dioxide (ThO2). CaF2 is a well characterized fluorite system that does not contain correlated electrons and will serve as a baseline, while ThO2, also a fluorite system, exhibits strong correlation effects requiring costly DFT approaches and will be used to demonstrate the efficacy of our approach. For both materials, electron irradiation experiments and thermal conductivity measurements will be performed to validate the effectiveness of developed potentials (thermal conductivity provides one of the most stringent tests for prediction of physical properties using MD simulations). This project fits nicely with Idaho National Laboratory (INL)'s Basic Energy Sciences (BES) engagement strategy and will provide critical modeling tools to help retain and to continue to grow the BES-materials program at INL. Furthermore, the high throughput capability of MD simulations using MLIP will become an important combinatorial materials science tool for developing and qualifying new nuclear fuels.

Significance: Fundamental understanding of radiation damage and its effects on materials properties is critically needed for rational design of advanced fuels and accurate prediction of their long-term in-reactor performance. Conventionally, DFT, the workhorse for materials discovery, has been quite successful in predicting thermal conductivities of perfectly ordered bulk crystals through the calculation of their second-order (harmonic) and third-order (anharmonic) force constants, which are then used as inputs to computer codes such as ShengBTE that solve the Boltzmann transport equation for thermal conductivity. However, such calculations quickly become extremely inefficient and nearly impossible for defective and disordered structures due to significantly reduced symmetry and the necessity to employ larger simulation cells. On the other hand, while classical MD simulations have the capability to simulate the aggregated impact of defects on phonon transport in complex systems using both equilibrium (Green-Kubo) and non-equilibrium methods, their predictive power is severely limited by the inability of empirical interatomic potentials with fixed analytical forms to describe complicated potential energy surfaces (PESs). Moreover, EPs are not structure generic and require new development for different crystal structures.

To bridge the gap between DFT and classical MD, this project plans to adopt the state-of-the-art data-driven machine learning (ML) techniques to develop next-generation interatomic potentials for model fluorite systems such as CaF2 and ThO2. Since a MLIP is not limited to any fixed functional forms, it can reproduce the complex DFT PES far better than any EP, and its predicting power can be systematically improved by increasing the size of the DFT training database. Furthermore, since a MLIP does not treat electrons explicitly, it remains orders of magnitudes faster than DFT. The quasi-DFT accuracy and computational efficiency of MLIP will enable MD simulations to quantify the effects of irradiation-induced point defects, defect clusters, and possible formation of correlated defect structures on thermal conductivity in these systems with unprecedented accuracy. The modeling efforts will be complemented and validated by electron irradiation experiments and thermal transport measurements.

This work will be among the pioneering attempts at INL to combine big data generated from high-throughput DFT computations with artificial intelligence to drive atomistic modeling of materials under irradiation. One-to-one comparison of predicted microstructure and thermal conductivity will be performed using state-of-the-art experimental tools located at INL. This project will have a broad impact on the design of advanced fuels, relevant to INL's science and technology (S&T) initiative "Nuclear reactor sustainment and expanded deployment" and Nuclear Materials Discovery and Qualification Initiative (NMDQi). More importantly, the development of computer codes implementing the MLIP methods in this project will directly support INL's S&T initiative "Advanced materials and manufacturing for extreme environments" by enabling realistic MD simulations of far-from-equilibrium processes such as collision cascades, shock loading, and crack propagation in a wide range of materials, all with near-DFT accuracy. Such simulations are particularly challenging for EPs due to their poor transferability among different atomic environments that will be encountered under extreme conditions of radiation, temperature, and pressure. Furthermore, the scientific knowledge and MLIP modeling capability established in this project will provide key enabling capability to solidify renewal proposals for two Energy Frontier Research Centers (Center for Thermal Energy Transport and Molten Salts in Extreme Environments) and a BES Materials core program (The Role of Anisotropy on the Self-Organization of Gas Bubble Superlattices) at INL.

Research Plan: The proposed research work can be divided into four main tasks and eight subtasks. All tasks can be accomplished by utilizing the supercomputing resources at INL and experimental facilities at INL and Rensselaer Polytechnic Institute (for electron irradiation experiments). The technical details are given below.

Task 1: Database construction

Subtask 1.1. Training structures: For constructing MLIP that can accurately describe both radiation damage and thermal conductivity, a sufficiently large database containing various atomic structures needs to be built to extensively sample many different local atomic environments. The database should incorporate the effects of temperature, deformation, defects, and other possible irradiation-induced structural changes, e.g., amorphization and melting. Trajectories of ab initio molecular dynamics (AIMD) simulations, which explore the part of the PES near equilibrium accessible by finite temperature lattice vibrations, will be included in the database in order to accurately model thermal conductivity. Snapshots of liquid structures will also be added to capture localized melting during cascades. In this stage, MD simulations using conventional EPs can be utilized to expedite the structure generation process.

Subtask 1.2. DFT dataset: Perform high-throughput DFT calculations using the Vienna Ab initio Simulation Package (VASP) code and INL's supercomputers (e.g., Sawtooth, Falcon) to generate large DFT training dataset containing information such as lattice vectors, atom positions, total energies, atomic forces, and stress tensors.

Task 2. Potential development

Subtask 2.1. Atomic environment descriptor: Based on conventional wisdom associated with the development of state-of-the-art ML potentials, efficient descriptors for local atomic environments will be surveyed and investigated. This fingerprinting process involves transforming complex atomic configurations into matrix forms and/or parameter sets that can capture the rotation, permutation, and translational symmetry, and is a key to determining the speed and accuracy of MLIP.

Subtask 2.2. MLIP fitting and testing: Existing methods based on artificial neural network (ANN), Gaussian approximation potential (GAP), spectral neighbor analysis potential (SNAP), and moment tensor potential (MTP) will be implemented using C++/Python codes, which will be employed to train MLIP for CaF2 and ThO2 using DFT datasets constructed in Subtask 1.2 as inputs. The performance of various ML techniques will be benchmarked in terms of accuracy and computational efficiency. Optimization of parameters such as the cut-off distance for defining the range of local environments perceivable by atoms and the architecture of neutral networks will be performed to fully unleash the potentials of the ML techniques.

Task 3. MLIP applications

Subtask 3.1. Interface to MD software: After developing MLIP to achieve the best trade-off between speed and accuracy, an efficient C++ interface to the most commonly used MD code, i.e., the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS), is planned. The code will take advantage of the neighbor lists and data structures in LAMMPS and provides the forces and energy of each atom based on the trained ML models.

Subtask 3.2. MD simulations: High-fidelity MD simulations on the primary radiation damage in CaF2 and ThO2 will be performed in LAMMPS using the MLIP developed in Subtask 2.2. The radiation induced defect structures will be analyzed and their impact on thermal transport will be directly calculated using equilibrium and non-equilibrium MD methods.

Task 4. Experimental validation

Subtask 4.1. Irradiation experiments: High-energy electron irradiation of CaF2 and ThO2 will be carried out at Gaerttner Electron Linear Accelerator (LINAC) center at Rensselaer Polytechnic Institute (RPI). Since electron beam irradiation rarely introduces displacement cascades, the primary defects being generated are point defects in the form of Frenkel pairs and their clusters depending on irradiation dose and temperature. CaF2 crystal substrate will be purchased from top chemical materials suppliers. INL will provide CaF2 to RPI. Large grain sized ThO2 samples will be fabricated by spark plasma sintering (SPS) by RPI to facilitate thermal conductivity measurement. Samples will be returned to INL for baseline characterization at the INL Research Center (IRC), Irradiated Materials Chacterization Laboratory (IMCL) and/ or the Electron Microscope Laboratory (EML). The samples will be returned to RPI to be irradiated. After irradiation, focused ion beam (FIB) technique will be employed to prepare lift out samples suitable for transmission electron microscopy (TEM) characterization of their radiation damage. Meanwhile, areas that are suitable for thermal properties measurement will be marked by FIB for following measurement of thermal conductivity (see subtask 4.2).

Subtask 4.2. Thermal conductivity measurements: Samples will be returned to INL for post-irradiation examination. Charactization will occur at IRC, IMCL and/or EML. Localized thermal conductivity measurements will be performed on both pristine and irradiated CaF2 and ThO2 samples by using the Thermal Conductivity Microscope (TCM). This state-of-the-art instrument is capable of mapping thermal conductivity and thermal diffusivity of the target material in a three-dimensional manner with micrometer spatial resolution. Specifically, the sensing depth can be adjusted within a range from a few to tens of micrometers, making the TCM an ideal tool to measure degradation of thermal conductivity caused by electron irradiation. Low temperature (cryogenic temperature) measurements will be made to identify thermal energy scattering mechanisms that are directly related to defect size, density and scattering cross section. High temperature measurements will be used to measure the recovery of thermal conductivity due to defect annealing. The results will be compared directly with MD thermal conductivity calculations in Subtask 3.2. Thermal conductivity measurements performed in this manner provide one of the most stringent tests for atomistic models of physical properties.

INL will provide CaF2 to RPI for preparation of the samples. RPI will fabricate ThO2 for the sample. Since the last step of the project is characterization of the samples at INL, the samples will be managed as low-level waste at INL at the end of the project

SECTION C. Environmental Aspects or Potential Sources of Impact:

Air Emissions

N/A

Discharging to Surface-, Storm-, or Ground Water

N/A

Disturbing Cultural or Biological Resources

N/A

Generating and Managing Waste

The project will result in the generation of some low-level waste. This is in the form of samples. The samples will be disposed at the INL following characterization. Industrial waste in the form of PPE and wipes may also be generated.

Releasing Contaminants

When chemicals are used, there is the potential the chemicals could be spilled to air, water, or soil.

Using, Reusing, and Conserving Natural Resources

All materials will be reused and recycled where economically practicable. All applicable waste will be diverted from disposal in the landfill where conditions allow. Project description indicates materials will need to be purchased or used that require sourcing materials from the environment. Being conscientious about the types of materials used could reduce the impact to our natural resources Project activities may release known greenhouse gases (GHGs) to the atmosphere and increase INL's energy use.

SECTION D. Determine Recommended Level of Environmental Review, Identify Reference(s), and State Justification: Identify the applicable categorical exclusion from 10 Code of Federal Regulation (CFR) 1021, Appendix B, give the appropriate justification, and the approval date.

For Categorical Exclusions (CXs), the proposed action must not: (1) threaten a violation of applicable statutory, regulatory, or permit requirements for environmental, safety, and health, or similar requirements of Department of Energy (DOE) or Executive Orders; (2) require siting and construction or major expansion of waste storage, disposal, recovery, or treatment or facilities; (3) disturb hazardous substances, pollutants, contaminants, or Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA)-excluded petroleum and natural gas products that pre-exist in the environment such that there would be uncontrolled or unpermitted releases; (4) have the potential to cause significant impacts on environmentally sensitive resources (see 10 CFR 1021). In addition, no extraordinary circumstances related to the proposal exist that would affect the significance of the action. In addition, the action is not "connected" to other action actions (40 CFR 1508.25(a)(1) and is not related to other actions with individually insignificant but cumulatively significant impacts (40 CFR 1608.27(b)(7)).

References: 10 CFR 1021 Appendix B to subpart D, Item B3.6, "Small-scale research and development, laboratory operations, and pilot projects" and B1.24 "Property Transfers"

Justification: The proposed R&D activities are consistent with CX B3.6 "Siting, construction, modification, operation, and decommissioning of facilities for small-scale research and development projects; conventional laboratory operations (such as preparation of chemical standards and sample analysis); small-scale pilot projects (generally less than 2 years) frequently conducted to verify a concept before demonstration actions, provided that construction or modification would be within or contiguous to a previously disturbed area (where active utilities and currently used roads are readily accessible). Not included in this category are demonstration actions, meaning actions that are undertaken at a scale to show whether a technology would be viable on a larger scale and suitable for commercial deployment.",

B1.24, "Transfer, lease, disposition, or acquisition of interests in personal property (including, but not limited to, equipment and materials) or real property (including, but not limited to, permanent structures and land), provided that under reasonably foreseeable uses (1) there would be no potential for release of substances at a level, or in a form, that could pose a threat to public health or the environment and (2) the covered actions would not have the potential to cause a significant change in impacts from before the transfer, lease, disposition, or acquisition of interests."

Is the project funded by the American Recovery and Reinvestment Act of 2009 (Recovery Act) ☐ Yes ☒ No

Approved by Jason Sturm, DOE-ID NEPA Compliance Officer on:02/4/2021