Development of machine learning potential and evaluation of material properties for UO_2-Cs based on the molecular dynamic simulations

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1. Introduction

With increased awareness due to accidents and recent global shifts of nuclear power policies, the importance of research for accident tolerant fuel (ATF) has highly increased. In ATF systems, the accident can be delayed by inhibiting the deformation, fracture, and release of nuclear fission materials even after the loss of emergency cooling system in nuclear power plants. Research for ATF concepts is broadly divided into the cladding material, fuel, and non-fuel components, and the improvement of UO_2 fuel is one of the main topics for developing ATF fuel.

The expected performance of enhanced UO_2 fuel is hindering the release of radiotoxic elements among fission products, and for this purpose, element diffusion in UO_2 is mostly analyzed. Element diffusion analysis is commonly performed using two approaches: experiment and simulation. However, conducting actual experiments using UO_2 is quite challenging due to the hazardous radioactive properties, resources scarcity, and high cost. Therefore, research using computer simulations is actively conducted within the field, which is cheaper, safer, and have lower barrier of entry than experiments.

We developed a machine learning potential to analyze Cs diffusion behavior in UO_2 system, with the aim of applying it to ATF fuel. Building potential file is the effective way to enable more realistic simulation with allowing large scale of calculations to mimic Cs diffusion in UO_2. As far as author's knowledge, the research about potential for simulation of ATF are scarce, even there are several UO_2 potentials that have been developed until now.

The machine learning potential developed in this study is based on DFT+U good to obtain oxide data with high accuracy, and optimized to get accurate thermal properties of UO_2 as well as Cs diffusion behavior. The simulation result of basic structural properties based on our potential showed a good agreement with ab initio simulation data and other references. This indicates the suitability of the potential to simulate UO_2 pellet deformation and Cs diffusion at high temperatures. Developed potential could be extensively applied to evaluate the potential of sintered UO_2 pellet to commercialization as ATF.

2. Methods and Results

In this section, the conditions utilized to develop moment tensor potential and simulate cesium diffusion behavior are indicated. We devided the used conditions into three main parts to describe.

2.1 Ab-initio molecular dynamics simulations

The calculated data of UO_2 fuel with cesium was obtained for making training set of moment tensor potential. Whole ab-initio molecular dynamics (AIMD) simulations are performed with Vienna ab initio simulation package (VASP). [1] We made total 37 initial structure models with switching the site of vacancy, substitution, and interstices, to consider various behavior of system in wide temperature range.

The initial structure models have 2×2×2 supercell structures, and these are utilized to AIMD simulation with Nose-Hoover thermostat in temperature conditions 500, 1500, 2500, and 3000 K. With setting the time step unit of 1 femtosecond (fs), the entire molecular dynamics (MD) simulation was completed within 100 steps. Therefore, the expected number of configurations to obtain was around 14800, but there are some steps not converged, so the final obtained configurations number is about 4700.

One of the important things to simulate UO_2 is the accurate description for the strong correlation between 5*f* electrons of U. The simulation of UO_2 shows low accuracy based on the local-density approximation (LDA) or the generalized gradient approximation (GGA); because it seriously underestimates the correlation between electrons. Therefore, in this research the reliable approximation named DFT+U employed to improve accuracy of data. [2] We used the 4.5 eV and 0.54 eV as the effective U and J values, particularly; which is confirmed to exhibit high accuracy based on the experimental data. Additionally, we set the cutoff energy value as 500 eV, with the electronic energy convergence value, 4.0×10^{-5} eV.

2.2 Developing moment tensor potential

The data obtained by AIMD simulations were whole utilized to prepare the training set and test set for the construction of moment tensor potential (MTP). The potential was developed using the Machine-learning interatomic potentials (MLIP) package, which is a highly reliable code for various predictions in multi-component systems. [3]

During the construction of MTP, the functional form called level plays a crucial role. It serves to control the accuracy and computational efficiency of MTP with setting hyperparameters. After the mean absolute error (MAE) and root-mean squared error (RMSE) convergence test for energy, we decided to use an untrained MTP of level 16 for the production of a more accurate UO_2 potential. Additionally, we selected configurations with interatomic distances ranging from 0.5 Å to 7 Å, to enhance accuracy with ensuring computational cost.

2.3 Simulation result of structural properties of UO_2

To verify the behavior of UO_2 lattice at high temperatures, we first examined the expansion of lattice parameter with temperature using molecular dynamics simulation. The simulation was conducted using a 2×2×2 size UO_2 supercell model and NPT ensemble, which was employed to fix the number of particles, pressure, and temperature. We also used a unit of 1 fs per time step, and run simulations for total 1,000,000 steps.

Figure 1 shows a graph comparing the lattice parameters obtained from actual experiments, the potentials developed in previous literature and this study. The results indicated that the thermal expansion coefficient depending on temperature is consistent with values reported in previous studies. Although there are some differences between the experimental and calculated data, these errors are negligible; since their overestimation tendency are resulted from the effective correction value U used in DFT+U.

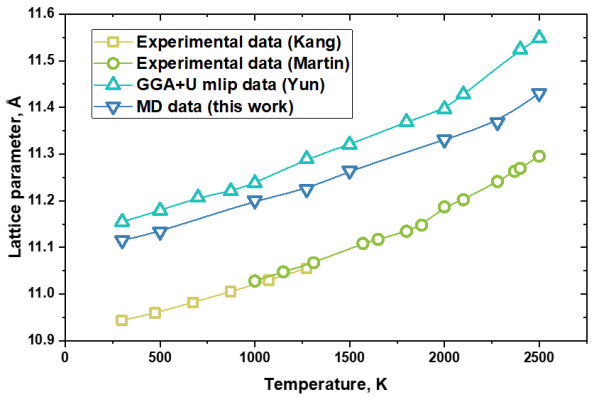

Fig. 1. Expansion behavior of lattice parameter depending on temperature.

3. Conclusions

We have developed a machine learning potential to investigate UO_2-Cs system, and confirmed that simulation results are similar to those obtained in DFT+U calculations and previous references. It indicates that this potential is suitable to simulate UO_2 expansion behavior and cesium diffusion in lattice at high temperatures. We may expect the wide application of this potential to simulations which aims to evaluate characteristics and commercialization potential of ATF pellet.

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