Development of machine learning potential and evaluation of diffusion behavior in UO_2 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. The final object of the ATF concept is the implementation the system, which can delay the accident even if the loss of the emergency cooling system happens. Research for ATF concepts is broadly divided into the cladding material, fuel, and non-fuel components, and the improvement of UO_2 fuel with doping is one of the main topics for developing ATF fuel.

The primary goal of UO_2 fuel improvement is to enhance binding between fuel and fission products, preventing their release even after an accident. Therefore, to check the capturing effect of improved UO_2 fuel, the diffusion behavior of fission product elements in UO_2 should be studied at first. The diffusion behavior of fission products such as xenon and krypton has been extensively studied experimentally because of their insolubility and inert gaseous properties. However, the diffusion of cesium has not been sufficiently investigated due to its high boiling point, volatility, and hazard to human bodies, which make experimental implementation challenging. Even though first-principles calculations can predict its behavior, developing a machine learning potential of UO_2-Cs system and performing molecular dynamics simulations are necessary to efficiently obtain results with considering various conditions.

We developed a machine learning potential to analyze Cs diffusion behavior in UO_2 system, with the aim of applying result data to ATF fuel research. The machine learning potential developed in this study is based on DFT+U to obtain data of oxide 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

2.1 Ab-initio molecular dynamics simulations

The calculated data of UO_2-Cs crystal system with ab initio molecular dynamics (AIMD) simulations was obtained for making training set of moment tensor potential. Whole calculations are performed with Vienna ab initio simulation package (VASP), [1] and 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. The energy difference between MTP and DFT+U calculation was compared, as shown in Figure 1. Additionally, we selected configurations with interatomic distances ranging from 0.5 Å to 7 Å, to enhance accuracy with ensuring computational cost.

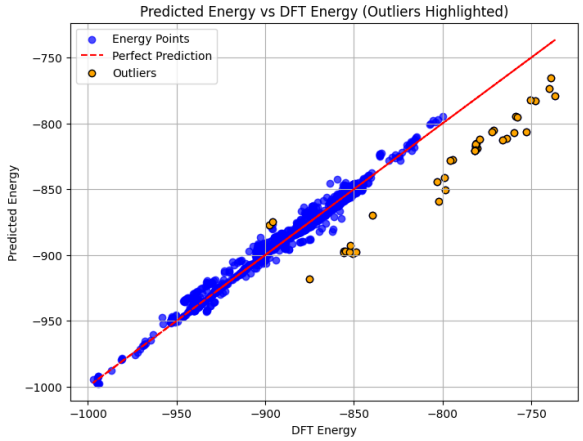

Fig. 1. Graph for comparison between predicted energy and DFT+U calculation energy.

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 (MD) 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 2 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.

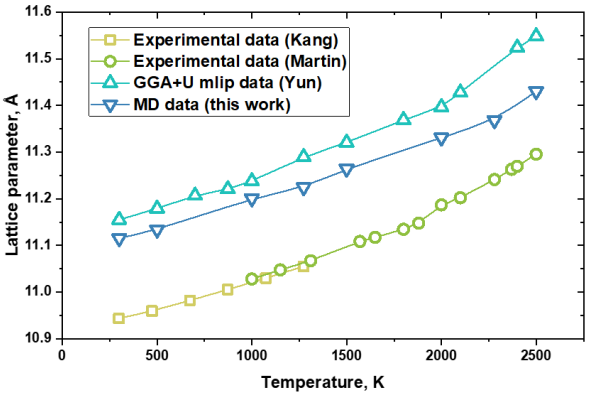

Fig. 2. Expansion behavior of lattice parameter depending on temperature.

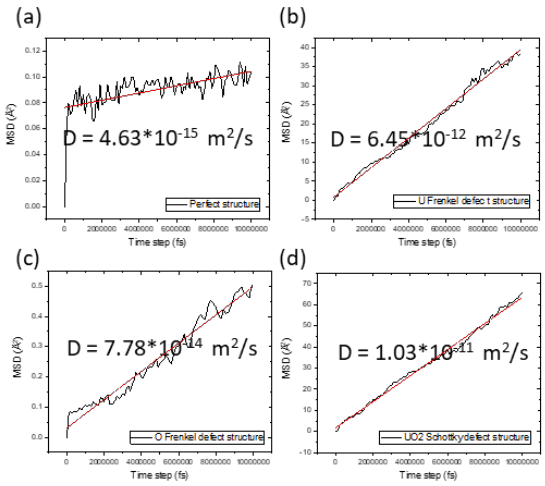

Fig. 3. Mean square displacement (MSD) of uranium in (a) perfect structure, (b) uranium Frenkel vacancy structure, (c) oxygen Frenkel vacancy structure, and (d) UO_2 Schottky vacancy structure.

Diffusion behavior of cesium and uranium was also simulated in 2000 K, based on MD calculation. Diffusion coefficient of element can be obtained with mean square displacement (MSD) data, using the following equation:

$$D(t) = D_0 exp\left(-\frac{E_a}{RT}\right) = \frac{<|S(t)|^2>}{6t}$$

where $<|S(t)|^2>$ is mean square displacement and t is time.

The diffusion of uranium is simulated in various structure model, to observe the effect of the concentration of defects on diffusion behavior at first. As shown in figure 3, the diffusion coefficient of uranium increases when uranium vacancies exist, whereas oxygen vacancies cause slight increase diffusion of uranium. These results correspond to the uranium diffusion mechanism reported in previous researches, where uranium mainly undergoes self-diffusion. [4] Therefore, the developed MTP can be concluded to be a reliable potential capable of

producing accurate results for high-temperature diffusion simulations in UO_2

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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