Validation of Cs diffusion behavior in UO2 for accident tolerant fuels with moment tensor potentials

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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 UO2 fuel is one of the main topics for developing ATF fuel.

The expected performance of enhanced UO2 fuel is hindering the release of radiotoxic elements among fission products, and for this purpose, element diffusion in UO2 is mostly analyzed. Element diffusion analysis is commonly performed using two approaches: experiment and simulation. However, conducting actual experiments using UO2 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 UO2 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 UO2. As far as author's knowledge, the research about potential for simulation of ATF are scarce, even there are several UO2 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 UO2 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 UO2 pellet deformation and Cs diffusion at high temperatures. Developed potential could be extensively applied to evaluate the potential of sintered UO2 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 21 initial structure models with switching the site of vacancy, substitution, and interstices, to consider various cases of cesium diffusion.

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, and 2500 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 6300, but there are some steps not converged, so the final obtained configurations number is about 4600.

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] 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. In this research, we employed an untrained MTP of level 16 for the production of a more accurate UO2 potential. Additionally, we selected configurations with interatomic distances ranging from 1 Å to 7 Å, to enhance accuracy with ensuring computational cost.

2.3 Molecular dynamics simulations

Cesium is known to be the most stable when it occupies uranium sites within the UO2 pellet. Additionally, the diffusion behavior of cesium mainly caused when the cesium locates to near on uranium vacancies. During radiation exposure while in operation, radiation-induced point defects are formed within UO2 pellets, and these defects may form defect clusters depending on conditions such as operating temperature.

Therefore, total eight models were constructed to test based on molecular dynamics (MD) simulations, depending on the type of vacancies and the inclusion of cesium. The package we used to simulate was large-scale atomic / molecular massively parallel simulator (LAMMPS). Models containing a single type of vacancy were created with a supercell size of 2×2×2, while models containing all types of vacancies were constructed with a supercell size as 3×3×3. All simulations employed a unit of 1 fs per time step at a temperature of 2400 K, and also utilized the NVT ensemble to fix the number of particles, volume, and temperature.

2.4 Simulation result of cesium diffusion

In this study, the diffusion coefficient (D) of uranium and cesium in models is calculated using a coefficient named the mean square displacement (MSD). The value of MSD is usually utilized to analyze the diffusion behavior of materials.

The average MSD and D values obtained by all supercell models can be observed through Fig. 1 and 2. The simulation results indicate when cesium is included within the UO2 structure, the D of cesium usually exhibits negative values. This implies that the neighboring uranium to vacancy migrate faster to vacancy compared to cesium, rather than cesium diffusion coefficient being negative.

Even if vacancies gather together, the similar trend is observed. At first, the probability of forming structures

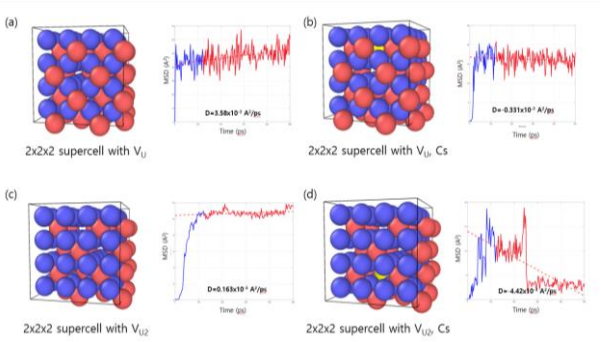

Fig. 1. (a) Simulation results of UO2 supercell containing uranium vacancies (V_U) without a uranium atom, (b) and including additional one cesium atom in structure (a). (c) Simulation results of UO2 supercell containing uranium di-vacancies (V_{U2}) without two uranium atoms, (d) and including additional one cesium atom in structure (c).

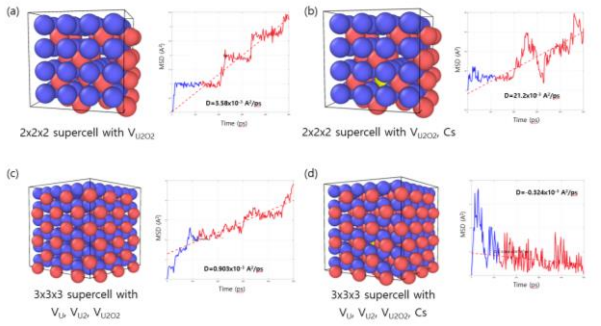

Fig. 2. (a) Simulation results of UO2 supercell containing uranium-oxygen di-vacancies (V_{U2O2}), which lack two neighboring uranium atoms and two oxygen atoms, (b) and including additional one cesium atom in structure (a). (c) Simulation results of UO2 supercell containing vacancy cluster (V_U, V_{U2}, V_{U2O2}) (d) and including additional one cesium atom in structure (c).

similar to those shown in Fig. 2 (c) or (d) is extremely low, because the actual vacancies generated during nuclear fuel operation occur at a low ratio. It is also predicted that a significant increase in the D of cesium within the structure would not occur if the majority of neighboring vacancies are uranium vacancies rather than oxygen vacancies. This is because uranium atoms have a heavier mass, resulting in lower self-diffusion, and cesium is also affected by this phenomenon.

3. Conclusions

We have developed a potential file to investigate cesium diffusion behavior, and confirmed that simulation results are similar to those obtained in previous references. In this study, we only examined cesium diffusion within pure UO2, but it is anticipated that the potential could also be applied to simulations aiming to explore various characteristics of ATF. Therefore, it is expected that this research could be utilized as one of the reliable research techniques in simulation studies for ATF.

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REFERENCES

[1] HAFNER, Jürgen. Ab-initio simulations of materials using VASP: Density-functional theory and beyond. Journal of computational chemistry, Vol. 29, 13, p.2044-2078, 2008.
[2] ANISIMOV, Vladimir I.; ZAANEN, Jan; ANDERSEN, Ole K. Band theory and Mott insulators: Hubbard U instead of Stoner I. Physical Review B, Vol. 44, 3, p.943, 1991.
[3] NOVIKOV, Ivan S., et al. The MLIP package: moment tensor potentials with MPI and active learning. Machine Learning: Science and Technology, Vol. 2, 2, p.025002, 2020.