Development of Machine Learning Potential and Investigation of Diffusion Behavior in UO_2 via Molecular Dynamics Simulations

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**Keywords : uranium dioxide, ab-initio molecular dynamics, machine learning potential, diffusion, bulk modulus*

1. Introduction

Recent shifts in global nuclear energy policys and heightened safety awareness have renewed attention on accident tolerant fuels (ATFs). Among diverse ATF strategies, performance enhancement of UO_2 remains central, as improved fuel–fission-product interactions can delay release under severe conditions.

While the transport of inert fission gases such as Xe and Kr is comparatively well documented, cesium remains less characterized due to its volatility and handling hazards. Computational approaches can bridge this gap by providing systematic insights under conditions that are not easily accessible experimentally.

To this end, we constructed a machine-learning interatomic potential for the UO_2–Cs system based on DFT+U, and used molecular dynamics to examine structural response and diffusion at elevated temperatures. The potential reproduces key thermophysical trends, indicating suitability for high-temperature defect and diffusion studies.

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. Additionally, we selected configurations with interatomic distances ranging from 0.5 Å to 7 Å, to enhance accuracy with ensuring computational cost.

2.3 Elastic property evaluation (ongoing)

In prior work, elastic constants were inferred from energy–volume curves by fixing the cell volume across discrete sampling points and fitting a quadratic to estimate the bulk modulus. We observed that the inferred modulus can vary with the chosen volume spacing, and extrapolation to higher temperatures becomes less consistant. As a remedy, we are testing dedicated bulk-modulus workflows within the

MD/analysis stack to improve robustness and temperature fidelity, as shown in Figure 1.

$$E(V) = E_0 + \frac{9V_0B_0}{16}\left\{\left[\left(\frac{V_0}{V}\right)^{2/3} - 1\right]^3 B_0' + \left[\left(\frac{V_0}{V}\right)^{2/3} - 1\right]^2 \left[6 - 4\left(\frac{V_0}{V}\right)^{2/3}\right]\right\}$$

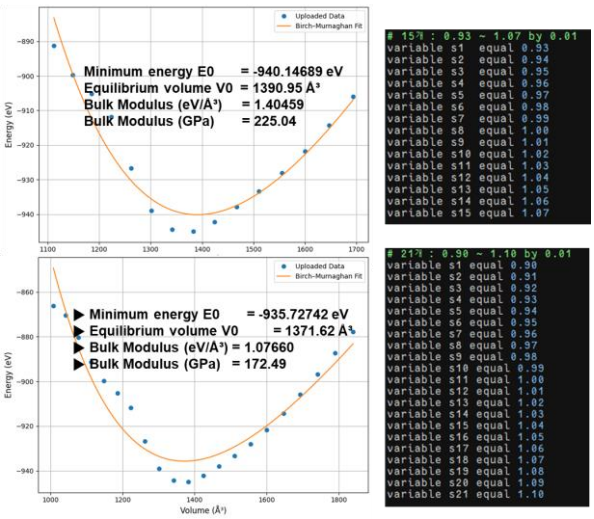

Fig. 1 Energy–volume relationship highlighting the limitations of quadratic fitting for bulk modulus estimation.

2.4 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.

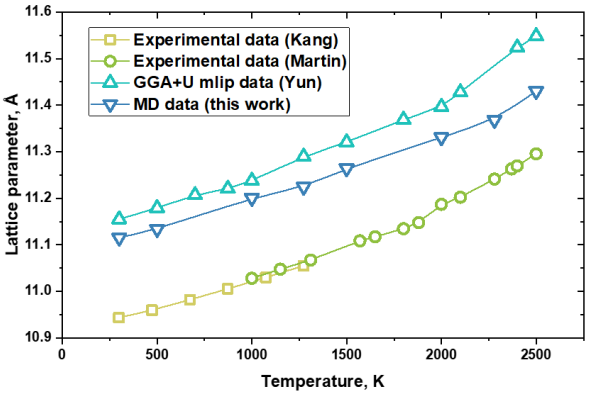

Fig. 2. Expansion behavior of lattice parameter depending on temperature.

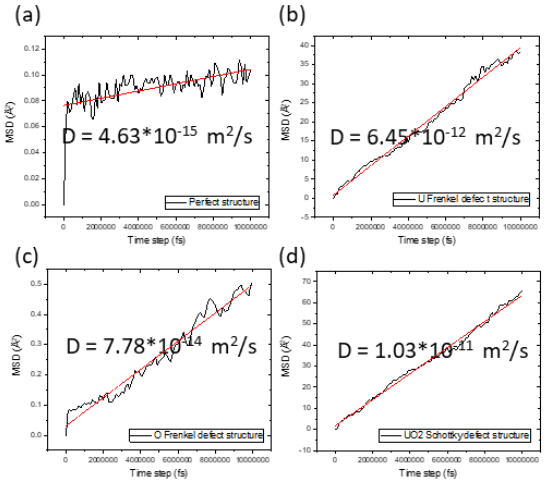

Fig. 3. Mean square displacement (MSD) of uranium in (a) perfect structure, (b) uranium Frenkel vacancy structure, (c) oxygen Frenkel vacancy structure, and (d) UO_2 Schottky vacancy structure.

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 developed and validated a machine-learning potential for UO_2–Cs that captures structural expansion and diffusion behavior at high temperatures. Building on this, an improved bulk-modulus evaluation strategy is being integrated to resolve inaccuracies in volume-fit-based estimates, supporting more reliable predictions for ATF-relevant conditions.

ACKNOWLEDGMENT

This work was supported by the National Research Foundation of Korea (NRF, No. 2022M2E9A304619011) grant funded by the Korea government (MSIT)

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