# Development of machine learning potential and evaluation of diffusion behavior in $UO_2$ based on the molecular dynamic simulations

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

## 1. Introduction

Due to nuclear accidents and recent changes in nuclear energy policies around the world, research on accident tolerant fuel (ATF) has become more important. The main goal of ATF is to develop a fuel system that can delay the progress of an accident, even when the emergency cooling system fails. ATF research generally focuses on three parts: cladding materials, fuel, and other non-fuel components. Among these, improving $UO_2$ fuel by adding dopants is one of the key approaches.

The main purpose of improving $UO_2$ fuel is to strengthen the interaction between the fuel and fission products, so that the fission products are not easily released during an accident. To evaluate this effect, it is necessary to first understand how fission products diffuse in $UO_2$. The diffusion behavior of gaseous fission products such as xenon and krypton has been widely studied because they are inert and do not dissolve well in the fuel. However, cesium diffusion has not been studied as much. This is because cesium has high volatility and is hazardous, which makes experiments difficult. Although first-principles calculations can provide useful information, molecular dynamics simulations using a machine learning potential are needed to study various conditions more efficiently.

In this study, we developed a machine learning potential to investigate the diffusion behavior of Cs in the $UO_2$ system, with the goal of applying the results to ATF research. The potential was trained using DFT+U data to ensure reliable accuracy for oxide materials. It was also optimized to reproduce the thermal properties of $UO_2$ and the diffusion behavior of Cs. The calculated structural properties showed good agreement with ab initio results and previous studies. These results suggest that the developed potential can be used to simulate $UO_2$ pellet behavior and Cs diffusion at high temperatures. The potential can also be applied to evaluate the feasibility of sintered $UO_2$ pellets for 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, $4.0\times10^{-5}$ 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.

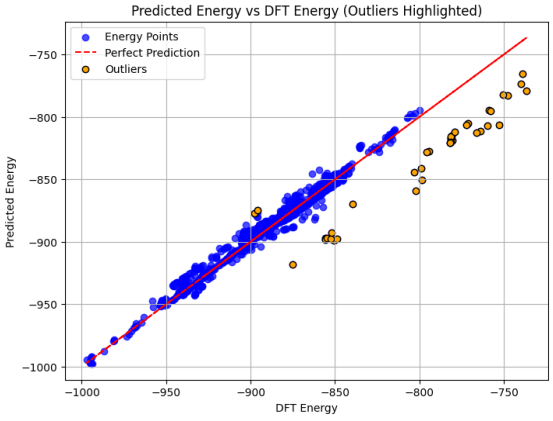


Fig. 1. Graph for comparison between predicted energy and DFT+U calculation energy.

To reduce bias caused by overrepresented data in the train and test sets, we checked how the data are distributed with respect to energy per atom (EPA), volume, and maximum force (max|F|). For each property, we divided the data and examined the data density in each range. When too many data points were concentrated in a specific region, part of them were removed to balance the dataset.

The sampling results and data density depending on criteria are shown in the Figure 2. The red dots indicate regions where similar data points were highly concentrated, and these points were partially trimmed. The histograms in the lower panels compare the original dataset (gray bars) and the filtered dataset (colored bars), showing how the distribution changed after removing the overrepresented data.

Fig. 2. Distribution of train and test data based on energy per atom (EPA), volume per atom, and maximum force.

### *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.

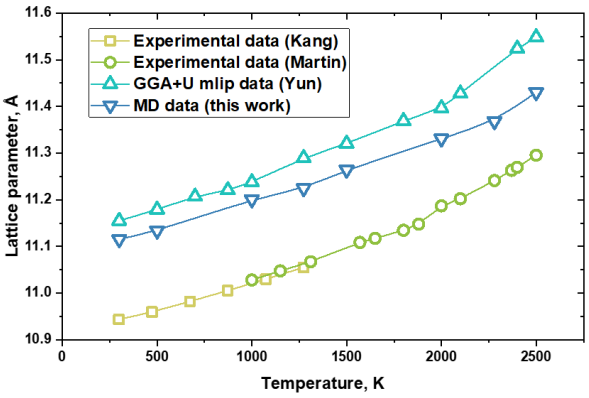


Fig. 2. Expansion behavior of lattice parameter depending on temperature.

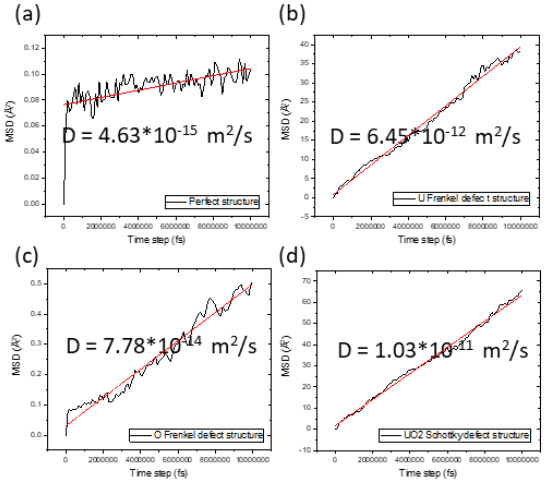


Fig. 3. Mean square displacement (MSD) of uranium in (a) perfect structure, (b) uranium Frenkel vacancy structure, (c) oxygen Frenkel vacancy structure, and (d) $UO_2$ Schottky

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.

## ACKNOWLEDGMENT

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Ministry of Science and ICT (MSIT) of the Republic of Korea (RS-2024-00420956, RS-2025-02353161), Korea Institute of Energy Technology Evaluation and Planning (KETEP) and the Ministry of Trade, Industry & Energy (MOTIE) of the Republic of Korea (RS-2025-02633904).

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