Diffusion Coefficient Calculation of ^{110m}Ag in ZrC at Very High Temperature Using Machine Learning Potential

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1. Introduction

In the context of Nuclear Thermal Propulsion (NTP), the requisites for nuclear fuel are intricate, given its operation under the demanding conditions of a peak temperature of approximately 2800 K. The key attributes mandated are high fission product retention, unwavering thermal stability, an elevated melting point, and robust thermal shock resistance. In this pursuit, the CERCER (CERamic-matrix Composite fuel with Embedded Reactor fuel) nuclear fuel concept has garnered attention. It manifests as a configuration wherein ceramic nuclear fuel particles are meticulously dispersed within a ceramic matrix. ZrC-coated Tri-structural isotropic (TRISO) or bi-structural isotropic (BISO) particle fuel dispersed in ZrC matrix hold promise as viable candidates for fulfilling the demanding requirements of NTP fuel, effectively addressing the multifaceted challenges posed by the arduous operating conditions.

Silver-110m is a highly radioactive fission product element which is a γ-ray emitter with a half-life of 249.8 days. The release of silver-110m from nuclear fuel can occur not only the surface contamination of the nuclear reactor of NTP but also in the event of an accident where it may leak outside the reactor. Therefore, the assessment of the diffusion and release behavior of silver-110m is necessary for the safety evaluation and commercial authorization of NTP. However, there is a notable absence of research concerning the diffusion behavior of silver-110m within the exceedingly high operational temperature range of NTP.

In this study, silver-110m diffusion constants within ZrC were calculated using several computational tools: Vienna ab initio simulation package (VASP) as a density functional theory (DFT) simulation tool, machine learning interatomic potential (MLIP) which functions as a machine learning potential generator, and Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) as a molecular dynamics (MD) simulation tool. [1-2]

2. Methods

To create a diverse training set, VASP-AIMD simulations were conducted. At 2800K, simulations were carried out for various structures including a 2×2×2 ZrC cubic lattice. Additionally, structures were generated with point defects in the cubic lattice, such as Zr vacancy (V_{Zr}), C vacancy (V_C), ZrC vacancy (V_{ZrC}), CC vacancy (V_{CC}), and ZrZr vacancy (V_{ZrZr}). Moreover, structures were generated with the introduction of Ag atoms into these defect sites which are Ag_{Zr}, Ag_C, Ag_{ZrC}, Ag_{CC}, and Ag_{ZrZr}. Each AIMD simulation was run for a duration of 700-time steps in each structure, with a time step of 1 fs in NVT ensemble. Out of the 700 configuration sets in each structure, the initial 200 sets were discarded. From the remaining 500 sets, a selection was made every 5-time steps to create the training set. This resulted in a total of 1100 training sets, each composed of 100 sets from 11 distinct structures. Each training set contains information about the positions of atoms, the direction and magnitude of forces acting on them at those positions, as well as the total energy of the system. All of this information is used to create a new interatomic potential using regression methods with machine learning.

The 1100 training sets were employed to generate a machine learning potential using the MLIP (Machine Learning Interatomic Potential) program. An MTP-level of 12 was utilized, and the maximum distance cut between atoms was set at 7 Å.

The generated potential was named MTP-1. LAMMPS simulations were conducted using MTP-1. The system was stabilized at 2800 K and 0 Pa over 2000-time steps with a time interval of 1 fs. In other words, LAMMPS simulations were performed for a total of 22,000 sets, each consisting of 2000 steps, across 11 distinct structure. Among these, structures significantly different from those used to create the MTP-1 potential were selected, resulting in a total of 149 steps out of the 22,000 steps. These selected structures were then subjected to single-point stabilization using VASP. The 149 new configurations that underwent VASP calculations were combined with the training set that was used to create MTP-1. This combined training set was used to create a new potential

using MLIP, named MTP-2. Using MTP-2, another MD simulation was conducted. This time, an NPT ensemble was employed at 2800 K and 0 Pa for 20,000-time steps in each structure. The reason for using a longer time step is that MTP-2 was generated from a more extensive and diverse training set compared to MTP-1. To capture structures with significant differences from those used to create MTP-2's training set through LAMMPS simulations, a longer time step was necessary. In the second LAMMPS simulation, 77 new structures were selected. These newly obtained configurations were then subjected to VASP single-point calculations, and the resulting data was added back to the training set to create a new potential, MTP-3. Using MTP-3, another LAMMPS simulation was conducted at 2800 K and 0 Pa in an NTP ensemble, spanning a total of 100,000-time steps in each structure. However, throughout this simulation, we were unable to identify configurations that displayed significant differences from the training set used to construct MTP-3. As a result, the active learning iteration aimed at potential refinement concluded at this point.

The diffusion coefficient was determined by utilizing the MTP-3 potential. LAMMPS simulations were performed in an NPT ensemble at 2800 K and 0 Pa for a duration of 1,000,000-time steps (equivalent to 1 ns), during which the mean square displacement of silver atoms was calculated with below equation.

$$6D_{Ag}t = (r_{Ag}(t)-r_{Ag}(0))^2 \qquad (1)$$

D_{Ag} = Diffusion coefficient of silver in ZrC [m^2/s]
t = time [s]
$r_{Ag}(t)$ = The displacement of silver over time t [m]
$r_{Ag}(0)$ = The position of silver at time t = 0s [m]

While validating the accuracy of the potential, it would be ideal to compare the diffusion coefficient of silver derived from the MTP-3 potential with experimental values. However, due to the lack of both experimental and simulated diffusion data for silver at the elevated temperature of 2800 K in ZrC, the author resorted to assessing the precision of MTP-3 potential by comparing the thermal expansion values of ZrC calculated using MTP-3 with experimental data

3. Results

Figure 1 demonstrates an excellent correlation between the experimentally reported thermal expansion data of ZrC and the thermal expansion data of ZrC measured using the newly developed machine learning potential in this study. These results provide confidence in the credibility of the constructed machine learning potential.

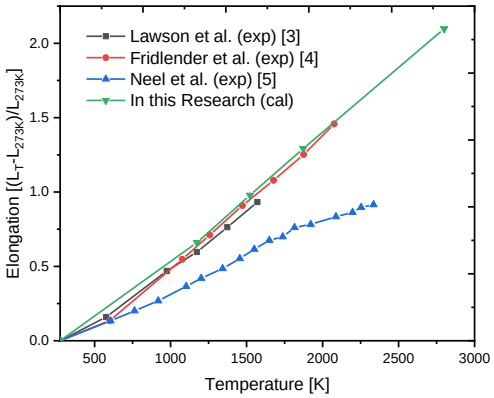

Figure 1. Thermal expansion behavior of ZrC calculated in this research and other experimental results. [3-5]

It has been reported that, among the point defects containing silver atoms in ZrC, the most stable configuration occurs when a silver atom occupies a vacancy at a carbon site. Furthermore, due to the lowest binding energy observed between the carbon vacancy and Ag substitution at a carbon site, it can be inferred that silver atoms in ZrC predominantly undergo diffusion driven by carbon vacancy. [6]

Figure 2 illustrates the graph of the Mean Squared Displacement (MSD) of a silver atom when substituting one carbon atom with a silver atom in a 4×4×4 $ZrC_{0.97}$ cubic structure. The diffusion was simulated using the LAMMPS software with the MTP-3 potential model under NPT ensemble at a temperature of 2800 K and pressure of 0 Pa. The graph depicts the MSD of the silver atom as a function of simulation time. In this graph, the slope divided by the geometric factor of 6 corresponds to the diffusion constant of the silver atom within the ZrC lattice at 2800 K. The value was determined to be 7.2×10^{-11} m^2/s. In the future, the diffusion constants will also be measured using the same method at temperatures of 2400 K and 3200 K. The calculated diffusion constants will provide valuable insights for empirical validation and safety analysis for NTP (Nuclear Thermal Propulsion) applications.

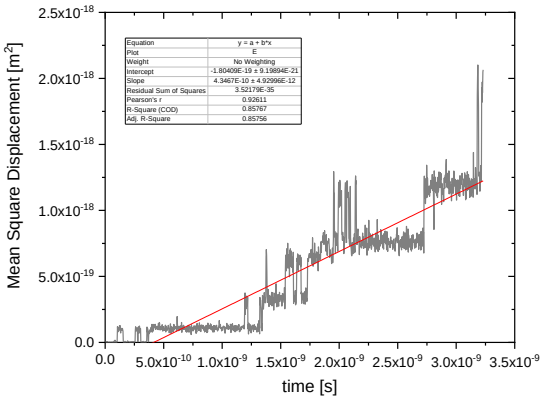

Figure 2. MSD along the time of silver atom in $ZrC_{0.97}$

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