Investigation of Diffusion Behavior of ^{110m}Ag in ZrC at Ultra High Temperature Using Machine Learning Interatomic Potential

Jae Joon Kim[a*], Eung-Seon Kim[a], Hyun Woo Seong[b], Ho Jin Ryu[b]

[a] Advanced Nuclear Fuel Technology Development Division, Korea Atomic Energy Research Institute, Daejeon 34057, Republic of Korea.
[b] Department of Nuclear and Quantum Engineering, Korea Advanced Institute of Science and Technology, Daejeon 34141, Republic of Korea.

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1. Introduction

Nuclear thermal propulsion (NTP) utilizes ultrahigh temperature nuclear fission reactors for rocket propulsion, offering higher flight speed and fuel efficiency compared to conventional chemical propulsion systems. In the NTP, the criteria for nuclear fuel are complex due to the need to withstand extreme conditions, including peak temperatures around 2800 K [1]. Such fuel must exhibit excellent fission product retention, consistent thermal stability, a high melting point, and strong resistance to thermal shock. The CERCER (CERamic-matrix Composite fuel with Embedded Reactor fuel) nuclear fuel concept has emerged as a noteworthy approach in this area. It features a design where ceramic nuclear fuel particles are carefully embedded within a ceramic matrix. The use of ZrC-coated Tri-structural isotropic (TRISO) or bi-structural isotropic (BISO) fuel particles within a ZrC matrix is seen as a promising solution to meet the stringent requirements of NTP fuel, effectively tackling the complex challenges presented by the harsh operational conditions.

^{110m}Ag, a significantly radioactive fission product element that emits γ-rays with a half-life of 249.8 days, poses a safety concern. The escape of ^{110m}Ag from the nuclear fuel could lead to surface contamination of the NTP reactor and, in case of an accident, its release into the environment. Hence, understanding the diffusion and release patterns of ^{110m}Ag is crucial for the safety evaluation and commercial approval of NTP technologies. Yet, research on the diffusion behaviors of ^{110m}Ag at the exceptionally high temperatures relevant to NTP operations is markedly lacking.

To address this gap, the diffusion constants of ^{110m}Ag in ZrC were determined through the application of various computational methods: the Vienna ab initio simulation package (VASP) for density functional theory (DFT) simulations, a machine learning interatomic potential (MLIP) for generating machine learning potentials, and the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) for molecular dynamics (MD) simulations.

2. Methods and Results

To construct a varied dataset for analysis, simulations using VASP-AIMD were performed at a temperature of 2800 K, focusing on a variety of structures including a 2x2x2 cubic lattice of ZrC. In addition to the pristine lattice, models were introduced with specific point defects within the lattice, such as vacancies of Zr (V_{Zr}), C (V_C), combined ZrC (V_{ZrC}), double C (V_{CC}), and double Zr (V_{ZrZr}). Furthermore, these defect sites were populated with silver atoms to create Ag_{Zr}, Ag_C, Ag_{ZrC}, Ag_{CC}, Ag_{ZrZr}, and interstitial Ag inserted configurations. For each structure, AIMD simulations were conducted over 700 time steps with a time step duration of 1 fs in the NVT ensemble. The first 200 configurations from each set were disregarded, and from the remaining 500, configurations were chosen 30 configurations randomly to compile the training dataset.

Using training sets, a machine learning interatomic potential was developed using the MLIP software, adopting a moment tensor potential (MTP) level of 16 and setting the atom cut-off distance at 7 Å. In order to increase diversity of training sets, Active learning method was used.

Developed machine learning interatomic potential was implied to (LAMMPS) to calculate diffusion coefficient of Ag in ZrC at temperature range of 2800-3200 K. The diffusion coefficient of silve in ZrC was calculated based on the slope of the mean square displacement (MSD) with following equation.

$$\mathrm{D} = \frac{1}{6}\lim_{t\to\infty}\left[\frac{|\vec{r}(t)-\vec{r}(0)|^2}{t}\right] \qquad (1)$$

Where $\vec{r}(t)$ is the position of the atom at a given time t, $\vec{r}(0)$ is the original position of the atom, corresponding to its position at the initial time $t = 0$.

3. Results

Figure 1 depicts the mean squared displacement (MSD) over time, derived through LAMMPS

simulations utilizing the developed potential. It is observed that the slope of MSD increase over time is greater at higher temperatures. Furthermore, it can be noted that the slope of MSD increase at a higher C vacancy concentration within ZrC, such as in $ZrC_{0.94}$, is greater compared to that of $ZrC_{0.97}$. Table 1 presents the pre-exponential factor and activation energy of diffusion coefficient of Ag within ZrC expressed in equation (1), calculated based on the slopes of Figure 2. It was confirmed that the results from the Arrhenius plot are similar to the experimental values at low temperatures as shown in Figure 3.

$$D_{Ag}[m^2/s] = D_0 \exp\left(-\frac{E_a}{k_B T}\right) \quad (1)$$

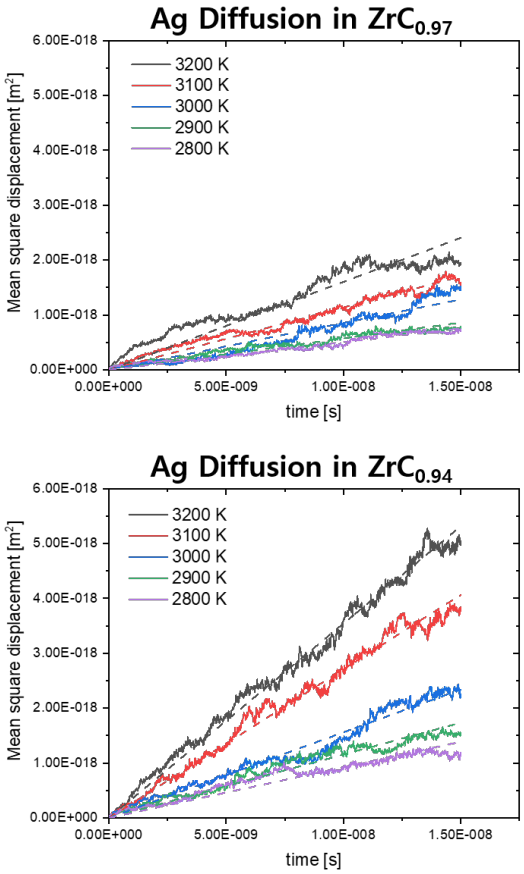

Figure 1. The calculated MSD values of Ag over simulation time at various temperatures and stoichiometries.

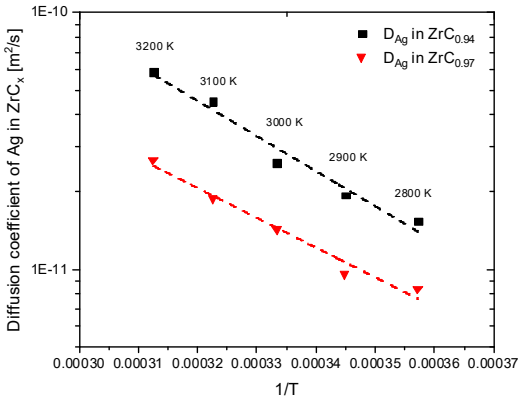

Figure 2. The calculated diffusion coefficients of Ag in Arrhenius plot

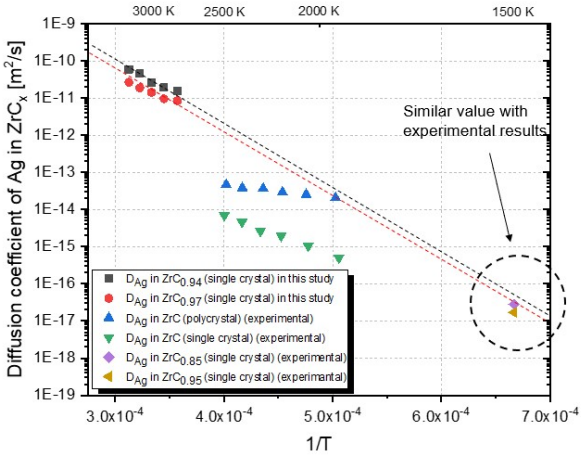

Figure 3. Comparison between simulation and experimental data.

Table 1. Pre-exponential factor and activation energy of diffusion coefficients of Ag in ZrC

	D_0 [m^2/s]	E_a [eV]
$ZrC_{0.94}$	1.095×10^{-6}	2.718
$ZrC_{0.97}$	1.067×10^{-7}	2.302

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References

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