Molecular Dynamics Simulation of Helium Diffusion in Nickel

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1. Introduction

It is well known that neutron irradiation significantly affects the performance of structural materials in nuclear reactors. Ni-based alloys have been proposed for in-core applications in molten salt reactor (MSR) because of their superior high-temperature strength, toughness and creep. However, intergranular attack by the fission product Te and irradiation embrittlement due to He atoms are potentially limiting factors for the application. Wright and Sham characterized the behavior of Hastelloy N, which was obtained from the MSRE (Molten Salt Reactor Experiment) program, and found that the most significant challenges for MSR structural materials are embrittlement from He produced by Ni(n,α) reactions [1]. Despite the detrimental effect of He on the mechanical properties of Ni-based alloys, the fundamental mechanisms behind the embrittlement are little understood.

An understanding of He behavior is important to estimate the materials property. We are interested in He embrittlement in Ni-based alloys under neutron irradiation. The first step is to evaluate the production of He by the nuclear transmutation reactions, which was carried out in the previous study [2]. In this paper, the migration of He atoms in Ni is described by using a molecular dynamics (MD) program, LAMMPS 'Large-scale Atomic/Molecular Massively Parallel Simulator' [3]. The evaluated He diffusivity will be used for the subsequent study in the future, which will cover the intergranular embrittlement by He bubble formation

2. Methods

In this section, we deal with He diffusion in the grains mathematically. In determining the He diffusivity in the model, we applied the MD simulation by using the LAMMPS code.

2.1 Diffusivity & Activation Energy

In general, the Einstein equation connects the macroscopic diffusivity D with the atomic properties of a particle motion, including the jump frequency and distance. The diffusivity D represents how fast a particle diffuses through a medium, which has such a relation as:

$$|\Delta r^2(t)| = 6Dt \quad (1)$$

where $|\Delta r^2(t)|$ is the mean squared distance (MSD) of a particle, which describes how far, on average, a particle migrates from the initial position after time t elapsed. Since the MSD is linearly proportional to the diffusivity D and the time, we can readily estimate the D from Eq. (1). In LAMMPS, the MSD of a group of atoms is computed with proper options [3]. We computed the MSD of He in Ni, from which the He diffusivity D_{He} was derived.

Diffusion is a random process, which follows an Arrhenius-type temperature dependence, implying the diffusivity increases exponentially with temperature such as:

$$\mathrm{D}(T) = D_o exp\left(-\frac{E}{RT}\right) \quad (2)$$

where D_o is pre-exponential factor, E is the activation energy for diffusion, R is the gas constant and T is temperature.

2.2 Ni-He Interatomic Potential

In order to produce reliable results from the MD simulations, it is critical to use the appropriate interaction potential. Although the Ni-He potential is not listed in the IPR (Interatomic Potential Repository) site [4], we obtained the potential from W. Zhang, who developed Ni-He potential by using the first principle calculation [5]. This potential is known to give a good description of stability and migration of He defects in bulk Ni.

3. Results

In LAMMPS, there is a built-in function of evaluating the MSD, from which we determine the diffusivity and activation energy. For a given temperature, the MSD was computed over sufficiently large samples and its average was taken. Fig. 1 shows temporal changes of MSD at 923K, where the sold line represents the average. The displacement of He atoms in Ni increases with time.

From the relation between D and MSD in Eq. (1), the linear regression model was used for predicting the behavior. The changes in MSD and fitting equations for given temperatures are expressed in Fig. 2. In this application, the linear portion of the curves was selected,

roughly corresponding to elapsed time of more than 20 ps. The fitting equations with the standard error of the slopes are listed in Table I

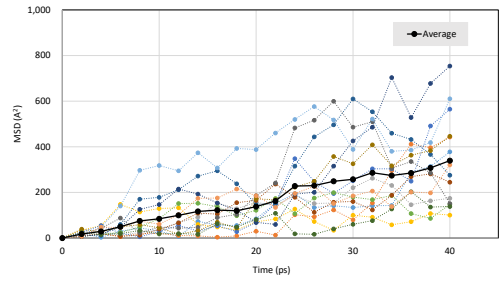

Fig. 1. Temporal changes of mean squared distance (MSD) of He atoms at 923K. (Sold line – average)

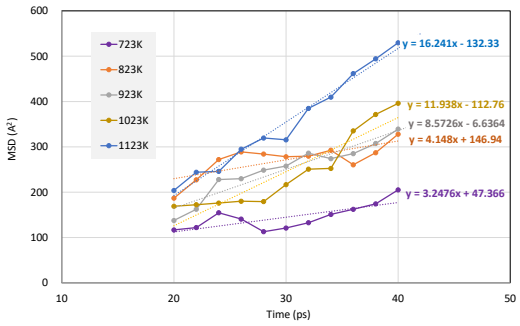

Fig. 2. MSD vs. observation time from LAMMPS simulation. The linear regressions are fit after 20 ps of the simulation time.

Table I: Linear regression statistics of MSD

Temperature (K)	Slope	R-square
723	3.248±0.923	0.579
823	4.148±1.242	0.553
923	8.573±0.915	0.907
1023	11.938±1.550	0.868
1123	16.241±0.817	0.978

The slope in the linear regression model represents the diffusivity D. By plotting ln (D) as a function of (1/T), called Arrhenius plot, we can determine the activation energy of E in Eq. (2). The Arrhenius plot is displayed in Fig. 3, from which the activation energy for He diffusion in Ni was found to be 0.13±0.02 eV.

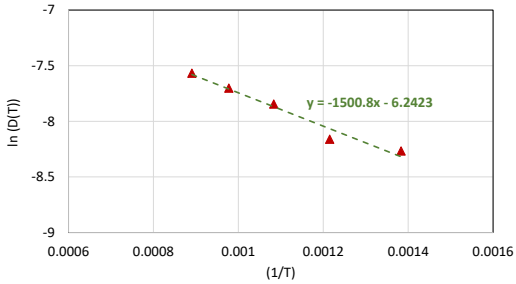

Fig. 3. Arrhenius plot of ln (D) vs. (1/T) for deriving the activation energy

4. Discussion

The presence of He in Ni-based alloys significantly affects their performance in nuclear applications. Due to the low solubility of He, its precipitation and bubble formation are inevitable. He bubbles along the grain boundaries are known to lead to embrittlement even at low He concentrations. Hence, we investigated the diffusive behavior of He in pure Ni at elevated temperatures (> 723K) by using the MD simulations.

The activation energy of He diffusion was estimated to be 0.13±0.02 eV from the simulation, which is very close to the measured value of 0.14±0.03 eV [6]. While such an agreement is encouraging, it is necessary to look into the microscopic diffusion mechanism. He atoms are known to be strongly bound to vacancies in metals. Hence, He atoms tend to occupy a substitutional site when they encounter a vacancy. In this process, the activation energy lies between 2.1 and 2.9 eV [7]. Other mechanism is an interstitial diffusion, in which He atoms in an interstitial position perform a series of jump without the interaction with vacancies. The calculated activation energy in this study corresponds to the interstitial diffusion of He. This diffusion mechanism plays an important role in fast migration of He to grain boundaries and enhances He embrittlement in Ni-based alloys.

Acknowledgements

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