Machine learning interatomic potential for hydrogen and helium solutes in nickel for an atomistic investigation on embrittlement

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1. Introduction

Nickel-based alloys are considered as promising structural materials in nuclear reactors, such as molten salt reactor (MSR), due to their advantages in mechanical properties and corrosion resistance over a wide temperature range. Despite these advantages, an embrittlement issue is one of the factors that limit the service life of an alloy. Hydrogen and helium are impurities known to cause embrittlement when incorporated into metals. Tritium is generated by nuclear reactions between neutron and lithium in a lithium-containing salt [1] and then it penetrates into the structural metal. Helium is generated by the transmutation of ^{58}Ni to ^{59}Ni by neutron irradiation and then the sequential neutron reaction with ^{59}Ni [2]. The generated tritium and helium segregate to grain boundaries (GBs) and cause significant degradation of the mechanical properties of the nickel-based alloys. However, due to the structural complexity of GBs, it is difficult to find out how hydrogen/helium segregate at GBs and how the mechanical properties are degraded. In addition, the synergistic effect of hydrogen and helium has not been understood clearly.

Atomistic simulation can be a powerful tool for detailed analysis of the embrittlement caused by hydrogen and helium. First-principles calculation, which is based on quantum mechanics, has high accuracy but its application has a limitation in the system size and simulation length due to the high computational cost. Thus, to conduct simulations to analyze embrittlement, such as tensile test simulations, molecular dynamics (MD) simulations are usually used. For MD simulations, an interatomic potential model needs to be prepared first.

Therefore, we aim to construct a potential model that can accurately describe grain boundary properties and the interaction between nickel, hydrogen, and helium. For this purpose, with reference to first-principles calculations based on density functional theory (DFT), a moment tensor potential (MTP) [3] was developed, which is a kind of machine-learning potentials and has a good balance between accuracy and computational cost. In this presentation, we will report performance test results of developed MTPs, and preliminary calculation results on hydrogen/helium embrittlement analysis.

2. Methods

2.1. Moment tensor potential (MTP)

In a potential model, the potential energy of interatomic interaction is expressed by an atomic environment of each atom, and the atomic environment function is described by a combination of basis functions. In MTP, the moment tensor descriptors, which consists of radial and angular functional term, are used to define the basis functions. The number of basis functions, which affects the accuracy and the computational cost, is controlled by the so-called level of MTP, and level 18 was adopted for constructing Ni-H-He interatomic potential in this study. The minimum and maximum cutoff radii for the radial basis functions were set as 0.2 Å and 6.0 Å, respectively. The coefficients of basis functions were optimized by fitting to a set of energy, force, and stress data obtained by first-principles calculations.

2.2. Density functional theory (DFT) calculation

First-principles calculations were conducted by means of DFT using the Vienna ab-initio simulation package (VASP). The Perdew-Burke-Ernzerhof (PBE) functional of a generalized gradient approximation (GGA) was used for exchange-correlation energy. The projected augmented wave (PAW) method was employed for the core-valence electron interaction. An automatic scheme with a length parameter 20 was used to define a k-point grid and the cutoff energy for the plane wave basis set was specified as 500 eV. All calculations were performed under spin-polarized conditions.

2.3. Tensile test simulation

After the construction of MTP, tensile test MD simulations were performed using LAMMPS code [4] to investigate the effect of hydrogen and helium on the mechanical properties of nickel with GBs. A bicrystal system composed of 12600 Ni atoms having Σ5[100](021) GB, which can be regarded as a representative of the high angle GBs, under periodic boundary conditions was prepared. When constructing the GB structure, the coincident site lattice (CSL) method [5] was used. A small system of Σ5[100](021) GB structure, which is composed of 80 Ni atoms, is shown in Fig. 1. After the geometry optimization, an equilibration run was performed at a target temperature for 20 ps with the Nose-Hoover thermostat [6][7] and the Parrinello-Rahman barostat [8]. Then, a uniaxial strain was applied to the x direction, which corresponds to the perpendicular direction to the GB plane, with a constant engineering strain rate of 10^{10} s^{-1}. The stresses in the y

and z directions were kept nearly zero during the deformation using an NPT ensemble.

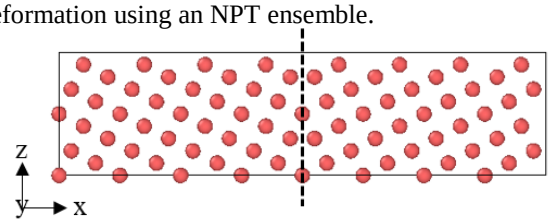

Fig. 1. The structure of the Σ5[100](021) GB with 80 Ni atoms. The black dashed line indicates the GB plane at the center.

3. Results and discussions

3.1. Validation of MTP

First, the fitting quality of the MTP was estimated by the root mean square error (RMSE) of energy, force, and stress from the DFT calculations, which were 5.3 meV/atom, 0.084 eV/Å, and 0.39 GPa, respectively. Next, basic material properties of fcc-Ni, such as (i) lattice constant and elastic constants, (ii) the solution energy of H and He, and (iii) vacancy formation energy, calculated using this MTP and DFT are compared in Table 1. Except for the solution energy of H in octahedral site, MTP showed good performance in reproducing the properties.

Table 1. The comparison of the lattice constant (a), elastic constants (C_{11}, C_{12}, C_{44}), vacancy formation energy (E_{f_vac}), the solution energy of H located at the octahedral site ($E_{s\_O,H}$) and at the tetrahedral site ($E_{s\_T,H}$), and solution energy of He located at octahedral site ($E_{s\_O,He}$) and at the tetrahedral site ($E_{s\_T,He}$) in fcc-Ni obtained from DFT, the constructed MTP, and previous studies by experiment or DFT.

	DFT	MTP	Ref.	Ref. type
a (Å)	3.517	3.517	3.524	Exp. [9]
C_{11} (GPa)	272.6	288.5	253	Exp. [9]
C_{12} (GPa)	157.5	159.7	152	Exp. [9]
C_{44} (GPa)	128.8	113.8	124	Exp. [9]
E_{f_vac} (eV)	1.526	1.543	1.54-1.80	Exp. [10]
$E_{s\_O,H}$ (eV)	0.049	0.142	0.088	DFT [11]
$E_{s\_T,H}$ (eV)	0.322	0.294	0.311	DFT [11]
$E_{s\_O,He}$ (eV)	4.561	4.602	4.735	DFT [12]
$E_{s\_T,He}$ (eV)	4.440	4.468	4.577	DFT [12]

When embrittlement occurs, the intergranular fracture is generally observed through GB or interface. Thus, we also validated the MTP with GB properties, especially the segregation energy (E_{seg}) of H at GB sites. E_{seg} values at several GB sites were obtained by DFT and MTP as shown in Fig. 2. Although the relative stability of H at different GB sites was reasonably reproduced, the present MTP underestimated the segregation energy by 0.12 ~ 0.19 eV. This underestimation error is largely due to the overestimation error of the solution energy of H because the segregation energy is calculated from the energy difference between the solute H and the segregated H. Therefore, more training data related to H at octahedral sites should be included in the construction of MTP if we need to more accurately determine the segregation energy.

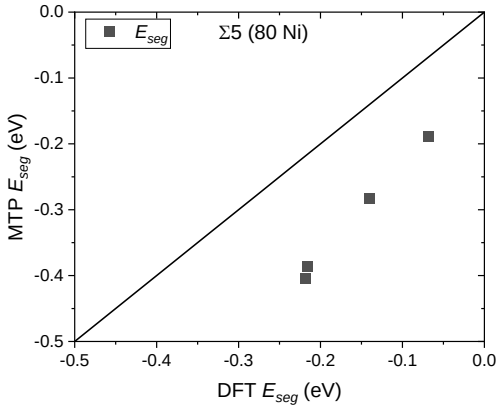

Fig. 2. The segregation energy of hydrogen at Σ5 GB sites calculated by DFT and MTP. The small system composed of 80 Ni atoms was used in this calculation. The reference line indicates $y = x$.

3.2. Application of MTP to tensile test simulations

As a preliminary test using the constructed MTP for MD simulations of embrittlement analysis, we performed a tensile test with the Σ5 GB system composed of 12600 Ni atoms. To investigate the effect of hydrogen and helium on the embrittlement, we inserted 650 hydrogen or helium randomly in the system, which corresponds to approximately 5 % of the atomic ratio of H or He to Ni. Higher concentration of solutes than the experimental conditions was adopted as an example to address the embrittlement effect due to the limitation in the system size and the simulation time. Calculated stress-strain curves are shown in Fig. 3. Helium has more influence on the degradation of the mechanical strength than hydrogen, which is consistent with previous experimental studies [13].

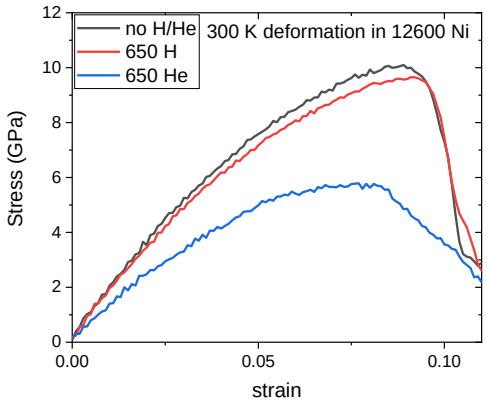

Fig. 3. The stress-strain curves obtained from the MD simulations using the constructed MTP for Ni-H-He. Black,

red, and blue line indicates the tensile test results of 12600 Ni, 12600Ni with 650 H, and 12600 Ni with 650 He system with Σ5 GB, respectively.

4. Conclusion

A MTP for Ni-H-He ternary system was constructed by training with energy, force and stress data obtained by first-principles calculations. We validated our MTP with lattice constant, elastic constants, vacancy formation energy, and solution energy of hydrogen and helium in fcc-Ni, which showed reasonable agreement with the DFT calculation results and available experimental data. We also compared the segregation energy of hydrogen at GB sites, because our main purpose of constructing this MTP is to investigate embrittlement phenomena, which mainly happen along GBs. Although the segregation energy was systematically underestimated compared to DFT results, the relative stability at several GB sites was reasonably reproduced. In addition, tensile test MD simulations were successfully performed using the constructed MTP, and helium in nickel appeared to have a greater impact on the embrittlement. We plan to perform a series of dynamic simulations to understand the embrittlement mechanism in future studies.

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