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Automatic kinetic Monte-Carlo modeling for impurity atom diffusion in grain boundary structure of tungsten material

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ABSTRACT

The diffusion process of hydrogen and helium in plasma-facing material depends on the grain boundary structures. Whether a grain boundary accelerates or limits the diffusion speed of these impurity atoms is not well understood. In the present work, we proposed the automatic modeling of a kinetic Monte-Carlo (KMC) simulation to treat an asymmetric grain boundary structure that corresponds to target samples used in fusion material experiments for retention and permeation. In this method, local minimum energy sites and migration paths for impurity atoms in the grain boundary structure are automatically found using localized molecular dynamics. The grain boundary structure was generated with the Voronoi diagram. Consequently, we demonstrate that the KMC simulation for the diffusion process of impurity atoms in the generated grain boundary structure of tungsten material can be performed.

1. Introduction

The diffusion processes of hydrogen and helium in tungsten material are important because of their retention problem in fusion reactors. In particular, hydrogen isotope retention in plasma-facing material (PFM) is a key factor in determining the recycling of fuel in a vacuum vessel. Helium retention causes the formation of helium nanobubbles [1] and fuzzy nanostructures [2,3] on the surface of plasma-facing tungsten material. Their nanostructure causes a drastic change in the plasma wall interaction in fusion reactors [4,5]. From the viewpoint of tritium handling, the diffusion process of hydrogen isotopes continues to pose a problem for tritium permeation.

One problem in the research on impurity diffusion in reactor materials is the difference of diffusivities of hydrogen and helium in experimental measurements. One reason of the difference in impurity diffusion is the grain boundary structures. The grain boundary structures differ in experimental tungsten sample materials. To begin with, the experimental measurement of diffusivity in tungsten material is difficult. Whether the grain boundary accelerates or limits the speed of impurity diffusion is not well understood.

We aim to clarify the effect of the grain boundary structure on impurity diffusion using simulation research. To simulate the impurity diffusion in PFM, molecular dynamics (MD) and kinetic Monte-Carlo (KMC) simulations are often employed. In terms of hydrogen isotopes, the speed of diffusion has been reported to both increase and decrease in the grain boundary structure based on simulation studies [6,7]. In terms of helium, the anisotropy of the speed of diffusion on grain boundaries was investigated using MD with high-performance computing [8]. However, most simulation studies treat a symmetric grain boundary structures. Therefore, our purpose is to investigate the impurity diffusion in asymmetric grain boundary structures comparable to those in the target materials used in experiments, which indicates that the microscopic structure in the grain boundary has rough atomic configurations.

The time scale of MD reached by current computer systems is insufficient for comparison with experimental studies. KMC, which has a higher speed and consists of a simplified model, is often used to treat impurity diffusion in/on PFM. For modeling, KMC requires information on the location of local minimum energy sites for impurity atoms and on the migration paths with their barrier energies. Recently, this information has been estimated using

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density functional theory (DFT) calculations. In fact, the existing KMC models and codes [7,9,10] were constructed based on the estimated values from DFT. In terms of symmetric grain boundaries such as the Σ5 structure [11,12], the local minimum energy sites were also investigated using DFT. However, if the grain boundary has an asymmetric structure and is distributed in a larger material, the calculation speed of DFT on a current computer system is too slow to estimate all the local minimum energy sites and migration paths. In this situation, KMC modeling for hydrogen transport in a large material with a grain boundary structure has been proposed [13], and that work is the motivation for the present study.

In the present work, to treat impurity diffusion in realistic and rough grain boundary structures, we propose automatic KMC modeling for an arbitrary structure. In the present method, a realistic and rough grain boundary structure of tungsten material is generated using the Voronoi diagram. Moreover, all the local minimum energy sites, migration paths, and migration barrier energies for impurity atoms in the grain boundary structure are automatically found using localized MD.

The method of automatic KMC modeling is described in Section 2. We discuss the present method with actual KMC simulation on the generated migration paths in Section 3. We discuss the effectiveness of problems associated with the automatic KMC modeling with our future plans in Section 4, and we provide conclusions in Section 5.

2. Automatic KMC modeling

The details of the method for the automatic KMC modeling for a grain boundary structure are described in this section. Fig. 1(a)–(d) shows the work flow of the present method. The method consists of the following six steps. Steps 1 to 4 are the processes to generate a grain boundary structure using Voronoi diagram, which is similar to the previous works [14,15]. Main methods in the present paper is the process to find local minimum energy sites in step 5 and migration paths in the generated grain boundary structure in step 6.

In step 1, the center positions of grains in a simulation box under periodic boundary conditions are determined, as shown in Fig. 1(a). The number of grains and size of the simulation box are parameters for this step. The mean grain radii, which are calculated using these parameters, correspond to those of the sample materials in actual experiments. Here, we used coarse-grained MD to determine the center positions of the grains. In the coarse-grained MD, the structure of the center positions of the grains is relaxed on the Lennard–Jones potential, which acts as simple two body forces on all the pairs of center positions of grains. This approach prevents the appearance of much closer center positions of grains.

In step 2, the simulation box is divided into grain regions according to the Voronoi diagram, as shown in Fig. 1(b). The center positions of the grains determined in step 1 are regarded as the points called seeds, sites, or generators in mathematics to generate Voronoi cells. A Voronoi cell including a seed is the region whose distance to the seed is smaller than distances to the other seeds. Each Voronoi cell is regarded as a grain having a single crystal structure.

In step 3, tungsten atoms having a body-centered cubic (BCC) crystal structure are arranged in each Voronoi cell, as shown in Fig. 1(c). The directions of the BCC crystal axes are randomly determined cell by cell. As a numerical problem of this method, the distance between the tungsten atoms disposed on both sides sandwiching the Voronoi boundary often becomes much short. To prevent this problem, we add the width of the gap between grains, g, as the third parameter. Before a tungsten atom is placed in a Voronoi cell, the distances from tungsten atoms already positioned in the other Voronoi cells are calculated. If the minimum distance is smaller than g, the allocation of the tungsten atom is skipped. Through this verification, the mass density of the material is controlled by the gap in the grain boundary. In addition, unstable allocation of tungsten atoms that are much closer is prevented.

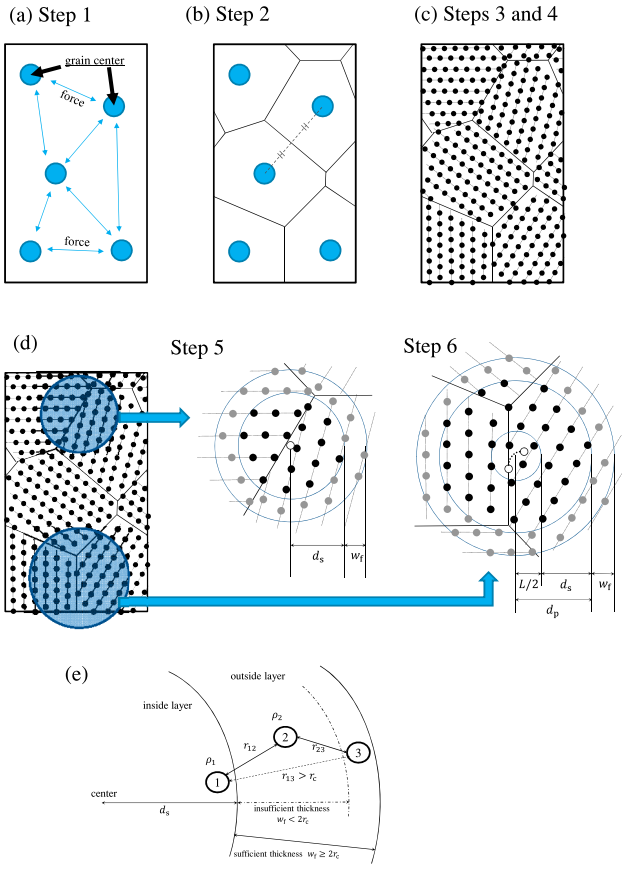

Fig. 1. Work flow of the automatic KMC modeling method. (a) Step 1: the center positions of the grains are determined by coarse-grained MD. (b) Step 2: the grain regions are defined as Voronoi cells. (c) Steps 3 and 4: the tungsten atoms are allocated along the crystal axes, varying randomly grain by grain, and then, the structure of all the atoms is relaxed by MD in the entire simulation box. (d) Steps 5 and 6: the local minimum energy sites and migration barrier energy are evaluated by the localized MD in a small area. The white spheres are impurity atoms for the structure relaxation in the step 5 and as the initial and final state of the NEB calculation in the step 6. The black spheres in the inside layer of the small area are movable atoms during structure relaxation and the NEB calculation, whereas the gray spheres in the outside layer of the small area are fixed atoms during relaxation. (e) Example of atomic location in the inside layer and the outside layer of a small area. The spheres numbered 1 to 3 indicate the positions of the atoms 1 to 3. The curved lines are the boundaries of the inside layer and outside layer. The curved dashed-dotted line is the boundary in the case that the thickness of outside layer w_f is smaller than double of cutoff length, $2r_c$.

In step 4, structure relaxation and annealing process for all the allocated tungsten atoms are performed using MD simulation as follows: First, we performed the structure relaxation, in which all atoms moved along with the direction of the forces on the interatomic potential. The structure relaxation before annealing was important because the atomic structure just generated by Voronoi diagram was a very unstable structure. Next, in the annealing process, all atoms were simulated under the NPT conditions that constant temperature is at 2000 K and constant pressure was at 10^5 Pa. The Nosé–Poincaré thermostat [16] and the Andersen method [17] were used to control temperature and pressure, respectively. By the Andersen method, the size of the simulation

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