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First-principles study of solvent-solute mixed dumbbells in body-centered-cubic tungsten crystals

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ABSTRACT

Tungsten (W) is considered as a promising candidate for plasma-facing materials for future nuclear fusion devices, and selecting optimal alloying constituents is a critical issue to improve radiation resistance of the W alloys as well as to improve their mechanical properties. We conducted in the current study a series of first-principles calculations for investigating solvent-solute mixed dumbbells in W crystals. The results suggested that titanium (Ti), vanadium (V), and chromium (Cr) are favorable as solutes for W alloys from irradiation-effect perspectives because these elements are expected to promote vacancy-interstitial recombination without causing radiation-induced precipitation that reduces ductility of irradiated materials.

1. Introduction

Tungsten (W) metals and alloys are considered as a promising candidate for plasma-facing materials for future nuclear fusion devices because of its high melting temperature, high resistance to sputtering, and high thermal conductivity. See related review articles [1,2] and the references therein for detail. However, radiation-induced effects, such as swelling [3] and increase in deuterium retention [4] seem to threaten its actual applicability. Generally speaking, primary damage of neutron and ion irradiation to crystalline materials is the displacement of countless atoms from their original lattice positions; this causes many interstitial atoms, which become important players of the microstructural evolution. So far, a lot of knowledge on interstitials not only in W crystals but also in other body-centered-cubic (BCC) transition metals have been accumulated. For instance, the most favored position of the interstitial atoms depends on element of interstitial: interstitials of light elements, such as hydrogen, helium, carbon, and oxygen, locate at octahedral site or tetrahedral site, on the other hands those of transition metals including self-interstitial atoms (SIAs) favor forming dumbbells, i.e., two atoms share a lattice site. Accordingly, heavy atoms in W crystals displaced by irradiation usually forms dumbbells. It had been conventionally assumed that such dumbbells would be configured in some symmetrical directions such as in $<111>$, $<110>$, or $<100>$ [5]. For example, $<111>$ SIA dumbbells in W crystals used to be recognized as the most energetically-favored configuration [6], and such SIAs would give one-dimensional (1D) motion: An atom in a dumbbell jumps to the first nearest neighboring (1NN) site, and it forms another dumbbell there; in the next step the atom initially located at the 1NN site jumps to the next leaving the atom that came to the site in the previous step. Currently, SIA dumbbells in a low-symmetrical direction between $<111>$ and $<110>$ are believed to be the most energetically-favored in W crystals [7]; they are called a $<11h>$ dumbbell [7] or a bridge dumbbell [8]. Note that the 1D migration described above is practically observable even for the low-symmetrical SIAs [9] (see also Fig. 1).

In the case of irradiation of W alloys, there are two possible consequences for displaced W atoms (i.e., an SIA): If the SIA and a solute substitutional are attractive, they form a stable solvent-solute dumbbell after the SIA migrates and reaches the vicinity of the solute within the reaction radius; on the other hand, if these are

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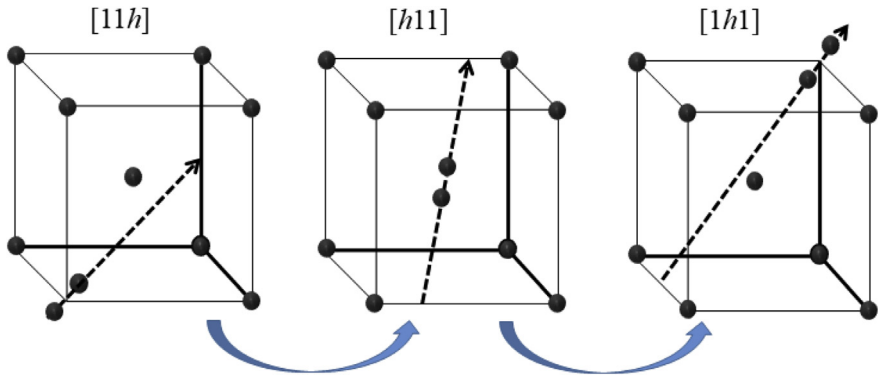

Fig. 1. An example of 1D migration path of bridge dumbbell SIA: Despite of the fluctuation of the dumbbell direction, the migration paths becomes linear [9] as observed in experiments [10].

repulsive to each other the displaced W atoms keep migrating without forming the mixed dumbbells. Rhenium (Re) and Osmium (Os) are produced from W through nuclear transmutation, and they would naturally be solute elements of W crystals under neutron irradiation. In such an alloy, such stable solvent-solute mixed dumbbells appear because the SIA and Re and Os substitutional atoms are attractive. Our previous study [7] analyzes the atomistic migration patterns of these two transmuted elements and indicates that the mixed dumbbells composed of Re (or Os) and W atoms are separated, and the Re (or Os) atom jumps to the 1NN site forming another mixed dumbbell. As discussed in the previous work [7,11], if only this kind of events is allowed, possible migration paths are extremely limited. For example, a mixed dumbbell in $<111>$ seemingly jumps back and forth between two neighboring sites. However, this is not the case because rotation of these mixed dumbbells is commonly easy; these solute elements can be carried for long distance by iteration of jumping and rotation events, letting them be capable of fast three-dimensional (3D) migration [11]. These findings significantly changed the landscape of the solute migration in W, because such SIA-Re complexes had been thought as an immobile defect [12]. Other solute interstitials such as vanadium (V) and titanium (Ti) interstitials in W crystals are also found to form stable mixed-dumbbells with a W atom [8] and to migrate easily keeping the form of mixed dumbbells [13] as described above.

Re and Os experimentally proved to suppress radiation-induced defects [3,14–19]. Through comprehensive theoretical discussions based on various pieces of experimental evidence [9], the 3D fast migration mentioned above seems to an origin of this phenomenon; this suggests that other solute elements may cause the similar effect. To optimize the property of W alloys as plasma-facing materials it is critical to explore the long-time kinetics of various solute elements under irradiation, but such a study is still halfway; kinetics of only Re and Os have been partly examined so far [9,11,13,20]. For this reason, parameters for various atomistic events related to many possible solute elements are indispensable.

In this paper, we examined stability and mobility of various solute interstitials in W crystals to search for candidates of solute elements of radiation-resistant W alloys. To be more specific, we intended to give comprehensive information necessary to the kinetic modeling of solute behavior under irradiation. In the current study, chromium (Cr) and molybdenum (Mo) interstitials were found to form stable mixed-dumbbells in addition to Re, Os, Ti, and V interstitials, which are previously known as solutes forming stable mixed-dumbbells [7,8]. The energetically-favored direction of the stable dumbbells was a low-symmetrical one between $<111>$ and $<110>$, except $<110>$ W-Os mixed dumbbells. Further detailed first-principles study suggested that Ti, V, and Cr would be favorable alloying elements for radiation-resistant W alloys.

2. Computational methodology

The density functional theory (DFT) is applied to conduct the first-principles calculations in the framework of generalized gradient approximation with projector-augmented wave (PAW) pseudopotentials [21] using the Vienna *ab initio* simulation package (VASP) [22]. We, within this framework, adopt Perdew-Burke-Ernzerhof pseudopotentials [23] from the VASP library. For all DFT calculations, we use a cut-off energy of 350 eV for the plane-wave basis with a first-order Methfessel-Paxton scheme employing a smearing parameter of 0.1 eV. Supercells applied are composed of 250 BCC lattice sites, having a dimension of $(5a \times 5a \times 5a)$, respectively, where a is the lattice constant obtained by the volume relaxation DFT calculation. For this supercell, the Monkhorst-Pack $3 \times 3 \times 3$ k-point meshes [24] is used to sample the Brillouin zone. The periodic boundary condition is applied to all three directions for each case. In each DFT calculation the total energy is obtained after relaxation of atomic positions without volume relaxation. Gharaee and Erhart [8] gave a systematic investigation how the formation energy of solute interstitial in W crystals changes in different size of super cells. Their results indicate that the estimated values increase as the system size increases, and that the increase rate does not vary with types of interstitials: the equation to calculate binding energy of an SIA to a solute atom from total energy values is

$$E_b^{SIA,\alpha} = E_f^{Sub-\alpha} + E_f^{SIA} - min\left(E_f^{SIA,\alpha}\right), \tag{1}$$

where $E_f^{Sub-\alpha}$ is the formation energy of α-element substitutional atom; $min(E_f^{SIA,\alpha})$ is the formation energy of the most energetically-favored SIA-solute complex, and we exclusively assumed that the most energetically-favored form of this complex is a solvent-solute mixed-dumbbell in the direction between $<111>$ and $<110>$ from our previous DFT study [7]. Note that the minimum formation energy of each mixed dumbbell is calculated not by nudged elastic band method explained below but by ordinary independent relaxation. Likewise, to calculate binding energy of a vacancy to a solute atom is given by:

$$E_b^{vacancy,\alpha} = E_f^{Sub-\alpha} + E_f^{vacancy} - E_f^{vacancy,\alpha}, \tag{2}$$

where $E_f^{vacancy}$ and $E_f^{vacancy-\alpha}$ are vacancy and vacancy-solute