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Migration of rhenium and osmium interstitials in tungsten

Tomoaki Suzudo [a, *], Masatake Yamaguchi [a], Akira Hasegawa [b]

[a] Center for Computational Science and e-Systems, Japan Atomic Energy Agency, 2-4 Shirane Shirakata, Tokai-mura, 319-1195, Japan
[b] Department of Quantum Science and Energy Engineering, Tohoku University, 6-6-01-2 Aramaki-aza-Aoba, Aoba-ku Sendai, 980-8579, Japan

HIGHLIGHTS

- We investigate the migration of rhenium and osmium in tungsten.
- The low rotation barrier of mixed dumbbells greatly influences their diffusivities.
- One cannot reduce their migration behavior to that of spherical objects.

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ABSTRACT

Tungsten is expected to be a promising plasma-facing material for future fusion devices, but radiation-induced precipitation (RIP), which leads the material to hardening, is a concern at their practical use. One of the keys to accurate prediction of the emergence of RIP is migration of solute atoms, rhenium and osmium, that are produced by nuclear transmutation through irradiation. We conduct a series of numerical simulations using an atomic kinetic Monte Carlo method and investigate the migration of these solute atoms in the form of tungsten–rhenium and tungsten–osmium mixed dumbbells, considered to be the most efficient "carriers" of the solute atoms. We find that the low rotation energy barrier of these mixed dumbbells leading to three-dimensional migration greatly influences their diffusivities. The result also suggests that, although these dumbbells have three-dimensional motion, one cannot simply reduce their migration behavior to that of vacancy-like spherical objects.

1. Introduction

Due to its high melting point and good resistance to sputtering, tungsten (W) is a promising candidate material for high-temperature and heavy irradiation applications, e.g. for the first wall and the diverter armor of nuclear fusion devices of the future. See, for example, Ref. [1] for information regarding recent developments of W materials for fusion applications.

Under neutron irradiation, transmutation elements are produced in W [2,3]. Among them, rhenium (Re) and osmium (Os) are the two main products; Os is produced via the production of Re. According to a predictive estimation [3], pure W used for the first wall armor of a nuclear fusion device is transmuted to an end-of-service alloy composition of approximately 91 at.% W, 6 at.% Re, and 3 at.% Os.

It is widely known that Re and Os precipitate under irradiation with solute concentrations below their solid solubility limits [4–10] which, at room temperature, are ~27 at.% and ~5 at.% for Re and Os, respectively [11,12]. This is a common radiation effect, observed in many alloys, and is known as radiation-induced precipitation (RIP). Because incoherent precipitates become obstacles for dislocation motions and are responsible for radiation hardening [9,13] that can lead to embrittlement of the materials, nucleation and growth of RIP under the fusion reactor environment must be accurately predicted.

As well as nuclear transmutations, direct effects of irradiation of materials are atomic displacements, i.e., vacancies, self-interstitial atoms (SIAs) and their clusters. The emergence of RIP is roughly explained as follows: an SIA is binding a solute atom; a vacancy is binding another solute atom; then SIA-vacancy recombination can cause aggregation of solute atoms [14–16]. See Fig. 1 for the schematic picture of the RIP nucleation mechanism. Obviously, point defects produced by atomic displacements (i.e., vacancy and SIA) and solute atoms (Re and Os) are all attractive in W crystals. As an aggregated solute atom cannot be dissociated from others without the actions of 'carriers' such as vacancies and interstitial atoms, the

* Corresponding author.
E-mail address: suzudo.tomoaki@jaea.go.jp (T. Suzudo).

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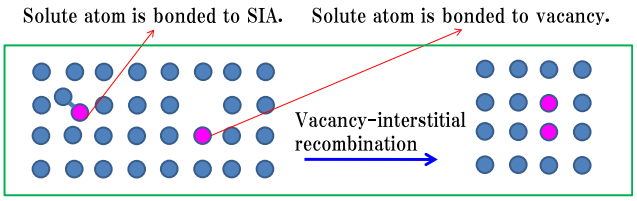

Fig. 1. RIP development mechanism; how solute atoms are aggregated [14].

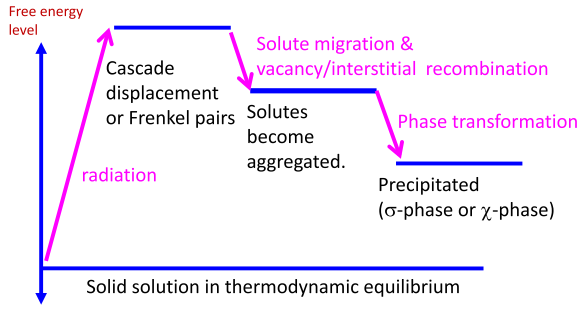

Fig. 2. Free energy changes over the process of RIP development.

clusters of solute atoms have long lives, giving them a chance to become large enough to be transformed into a different phase, such as σ-phase or χ-phase [8]. These precipitates are not in thermal equilibrium if the solute concentration is below the solid solubility limit. In other words, the free energy stored in a system having precipitates is larger than a system with Re, or Os, in solution. Because of this, the emergence of the RIP seems to be at odds with intuitive physical understanding, because the system seemingly gains the energy by itself. This is explained as follows: The free energy of the system is increased when the radiation produces atomic displacements; the system then loses the free energy when an interstitial atom and a vacancy recombine. The diagram in Fig. 2 illustrates this.

An additional atomistic phenomenon is necessary for the emergence of RIP, that is, migration of the solute atoms. The migration of Re and Os atoms in W plays a critical role in the development of RIP. The migration of solute atoms via the vacancy mode occurs through an exchange of a vacancy and an atom. In addition, a vacancy is able to drag a solute atom along with it if the solute atom and the vacancy are attractive to each other [17,18]. The migration style of solute atoms dragged by interstitials is more complicated, as there exist many types of interstitial atoms, located in tetrahedral or octahedral sites, or they exist as dumbbells with choices of various directions. As shown later in this paper, the migration of solutes via the interstitial mode are much faster than migration via the vacancy mode, and we believe that the interstitial migration mode would be the greater contributor to the aggregation of solute atoms.

In the current study, we aimed at investigating the migration of solute atoms via interstitial mode in W crystals by exploiting an atomic kinetic Monte Carlo method parameterized by the first principles studies [19]. We particularly focused upon the influence of the rotation energy barriers upon the migration of mixed dumbbells, as our previous study [19] suggests that their low rotation energy barriers leading to three-dimensional migration is key in the explanation of the radiation effect experimentally observed in W crystals.

Table 1
Formation energies of Re and Os interstitials in W (eV).

Interstitial-type	SIA	Re-interstitial	Os-interstitial
<111> crowdion	9.77	9.29	6.11
<111> dumbbell	9.78	9.17	5.84
<110> dumbbell	10.21	9.20	5.68
Tetrahedral interstitial	11.38	10.53	6.90
<100> dumbbell	11.86	10.20	7.79
Octahedral interstitial	11.97	11.32	8.11

2. Methodology

2.1. Energetics of point defects and solute atoms

The first principles studies necessary to the current study are conducted in the framework of generalized gradient approximation with projector-augmented wave pseudo-potentials [20] using the Vienna *ab initio* simulation package (VASP) [21]. The detail is reported in Ref. [19], and we use its results in the following. Table 1 and Fig. 3 summarize formation energy of SIA, Re interstitials, and Os interstitials in various forms. According to these results, the most energetically-favored forms for SIA, Re-interstitial, Os-interstitial are <111> dumbbell, <111> dumbbell and <110> dumbbell, respectively. Note that in the current paper, we use the expressions, <111> crowdion, and <111> dumbbell, interchangeably. For SIA and Re interstitials, some *ab initio* studies reported that asymmetric directions <11h> (h ~ 0.5) are slightly more favored than <111> dumbbells [19], but we do not know whether these asymmetric dumbbells are realistic. In addition, excess formation energies of <111> dumbbells from <11h> are not large, being equal to ~0.1 eV or less, so we consider <111> dumbbells as the most favored species for SIAs and Re interstitials in the current study.

Table 2 summarizes the binding energies between the solute atoms at a substitutional position and at point defects. Attractive relations are indicated for all the combinations. Note that in the current paper, it is assumed that a positive binding energy indicates attraction. This ensures that part of the conditions necessary for RIP development, described above, are satisfied. Since a vacancy and either of the Os and Re solute atoms are mutually attractive at the second nearest neighbor positions, vacancies are expected to drag the solute atoms.

Table 3 summarizes migration energy barriers of the solute atoms. It indicates that migrations via the vacancy mode are extremely slow compared with those via the interstitial mode. We believe that migrations via the interstitial mode carry the solute atoms much more effectively than do migrations via the vacancy mode and we, in the current paper, concentrate on the migration

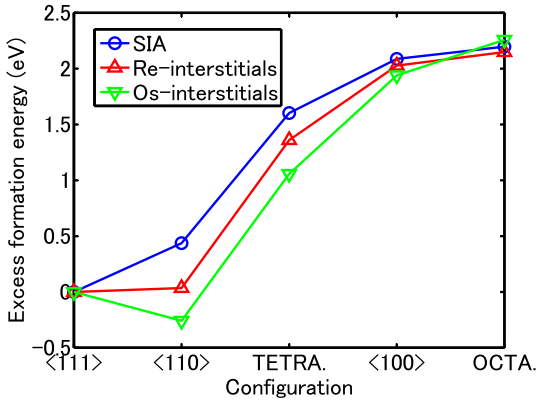

Fig. 3. Relative formation energy of SIAs and Re/Os interstitials in W crystals.