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Towards understanding the mechanism of rhenium and osmium precipitation in tungsten and its implication for tungsten-based alloys

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ABSTRACT

Using a first-principles method in combination with thermodynamic models, we investigate the interaction between rhenium/osmium (Re/Os) and defects to explore the mechanism of radiation-induced Re/Os precipitation in tungsten (W). We demonstrate that radiation-induced defects play a key role in the solute precipitation in W, especially for self-interstitial atoms (SIAs). The presence of SIAs can significantly reduce the total nucleation free energy change of Re/Os, and thus facilitate the nucleation of Re/Os in W. Further, SIA is shown to be easily trapped by Re/Os once overcoming a low energy barrier, forming a W-Re/Os mixed dumbbell. Such W-Re/Os dumbbell forms a high stable Re/Os-Re/Os dumbbell structure with the substitutional Re/Os atoms, which can serve as a trapping centre for subsequent interstitial-Re/Os, leading to the growth of Re/Os-rich clusters. Consequently, an interstitial-mediated migration and aggregation mechanism for Re/Os precipitation in W has been proposed. Our results reveale that the alloying elements-defects interaction has significantly effect on their behaviors under irradiation, which should be considered in the design of W-based alloys for future fusion devices.

1. Introduction

Both energy crisis and enormous potential of nuclear fusion energy drive us to make great efforts to develop nuclear fusion all over the world. The final application of nuclear fusion energy is mainly dependent on the development of key materials in the thermonuclear fusion device “Tokamak”, in which the choice of the plasma facing materials (PFMs) is one of the critical issues [1,2]. In future nuclear fusion reactors, PFMs must tolerate the irradiation of excessive heat flux, high fluxes of hydrogen (H) isotopes and helium (He) ions as well as deuterium-tritium (D-T) fusion neutrons [3,4]. Such extreme irradiation condition will significantly influence the properties of PFMs, which pose great challenges to materials research. Because of excellent thermal properties, high threshold for sputtering and low H/He retention, tungsten (W) and W-based alloys are considered to be the primary candidates for PFMs in future fusion reactors [5,6]. However, W also exhibits serious degradation under severe fusion irradiation environment. Especially, the irradiation of high energy (14 MeV) fusion neutrons not only results in the cascade damage but also causes pure W transmutation. Currently, since experimental measurement of W-PFM in a fully realistic fusion condition is not feasible, the concentrations of transmutation elements (TEs) are mainly obtained by calculations. Burn-up calculations show that a significant Rhenium (Re) and Osmium (Os) production is expected from pure W transmutation under future fusion power plant conditions [7–9]. Recent calculation [9] reports that the content of Re and Os after five years fusion power plant irradiation can reach up to 3.80 atomic% (at.%) and 1.38 at.%, respectively, under the condition of the first wall in mode B within European power plant conceptual study (PPCS) [10]. Despite the concentrations of Re and Os are low, their influences on the performance of W-PFM cannot be ignored. These transmutation elements (TEs) may form brittle precipitation in W, leading to the great concerns for the life-limiting of W-PFM in fusion devices.

Now, the effects of TEs on the properties of W and their behaviors in W are mainly investigated by studying W-based alloys with Re/Os alloying elements [11,12]. Re/Os will be uniformly dispersed in irradiation-free W, and has positive effects on the

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mechanical properties of W, such as reducing ductile-to-brittle transition temperature (DBTT) [13,14], strengthening recrystallization resistance [13,14] and improving both toughness and ductility [15,16]. Further, it is found that Re can inhibit the H bubbles and He 'fuzz' structure in W under H isotope ions and He ions irradiation [17–20]. Interestingly, the distribution of Re/Os in irradiated-W is significant different from that in irradiation-free W. Especially, under high irradiation level, high density of homogenous nanometric Re/Os-rich precipitate with acicular shape has been observed in W-Re, W-Os and W-Re-Os alloys [21–23]. Such precipitations substantially enhance the irradiation hardening and embrittlement of W [24–26], as well as other detrimental effects, which may severely reduce components lifetime. For example, using 2 MeV W^+ implantation and nanoindentation method, it is found that the hardness of W-5 wt.%Re alloy will increase by 2.88 GPa at 33 dpa with the formation of Re-rich precipitation, which is 213% larger than that of pure W under the same irradiation dose [24]. Apparently, the influence of Re/Os precipitation on W are remarkably different from the dispersed Re/Os. Therefore, exploring the mechanism for Re/Os precipitation under irradiation is very important to evaluate the influences of TEs on the performance of W-PFM.

It has been demonstrated that the structure of such precipitation is consistent with intermetallic phases (σ- and χ-phase) in W-Re, W-Os and W-Re-Os alloys [27]. Generally, Re/Os-rich phases are thermodynamically only expected for relative high Re/Os concentration (>28 at.% Re in W-Re and >6 at.% Os in W-Os at 1773 K) [27–30]. However, it is observed in nominally solid solution W-Re/Os alloys under high irradiation level [21,22,27,31]. For example, precipitation will be formed in W-3 wt.%Re alloy after neutron irradiation to ~1 dpa at 1073 K in high flux isotope reactor (HFIR) [21]. More recently, using atom probe tomography (APT) to examine clustering in W-2 at.% Re and W-1 at.% Re-1 at.% Os alloys induced by 2 MeV^+ W ion irradiation at 573 K and 773 K, Xu et al. [32,33] clearly observed the homogeneous aggregation of Re and Os atoms in bulk W. These suggest that irradiation should be responsible for the formation of Re/Os precipitation in W, which is the so-called radiation-induced precipitation (RIP) phenomenon. Obviously, the biggest difference between irradiated and irradiation-free W is whether there are radiation-induced defects or not. Therefore, the radiation-induced defects should play a key role in the precipitation of Re/Os in W [27,32–35].

Modelling and simulation are very useful to understand the behaviors of alloying elements/impurities and their interactions with defects in fusion materials [35,36]. Previous studies found that the interaction between Re/Os atoms in W is very weak [37,38], while there is strong attractive interaction between Re/Os and vacancy [38–41]. In W-Re alloys, it is also found that vacancy can contribute to the clustering of Re [41–43]. In our previous study [43], we have demonstrated that the presence of vacancy significantly reduces the total nucleation free energy change of Re/Os cluster in W, suggesting that vacancy can facilitate the nucleation of Re/Os in thermodynamics. However, due to high migration energy barrier [38–40,43], the diffusivity of Re/Os via vacancy-mediated path should be extremely low. On the other hand, it is found that there is also strong attractive interaction between Re/Os and self-interstitial atoms (SIAs) [38,40,43–49]. Meanwhile, SIAs and Re/Os-W mixed interstitials can readily diffuse in W with extreme low migration energy barriers [40,46–48]. These Re-W mixed interstitials are strongly attracted to each other, leading to the formation of interstitial pairs, which maybe contribute to the clustering of Re in W [48]. More recently, based on the energetics of defects in W from first-principles calculations, Huang et al. [50] conducted kinetic Monte Carlo simulation of Re cluster formation in irradiated W-Re alloys. They found that both vacancy and SIA can lead to Re agglomeration in W. Although many efforts have been made to investigate the behaviors of TEs and their interactions with radiation-induced defects in W, the energetic parameters on some key processes (the influence of SIAs on the thermodynamic stability of Re/Os clusters and the nucleation of Re/Os via defect-mediated paths) and the formation mechanism of Re/Os-rich clusters in W under irradiation are still unclear.

Here, to explore the physical origin for the formation of Re/Os clusters in W under irradiation, we systematically investigate the interactions between Re/Os atoms and SIAs using a first-principles method in combination with thermodynamic models. Thermodynamically and kinetically, we demonstrate that the interstitial-mediated mechanism could be mainly responsible for the radiation-induced Re/Os clustering in W, which can qualitatively well explain the recent experimental phenomena. More importantly, the interstitial-mediated mechanism is expected to be applied in some W-based alloys, leading to the clustering of alloying elements and resulting in the detrimental effects on the properties of materials, which must be considered in the design of W-PFM. The present results, together with previous calculations, suggest that Ta might be a suitable alloying element in W to suppress Re/Os precipitation without introducing the other clusters. Our calculations will provide a good reference for estimating the behaviors of alloying elements in W under irradiation and also have a guiding significance for developing W-based alloys for future fusion devices.

2. Computational method

Our first-principles calculations were performed within density functional theory (DFT) as implemented in the Vienna Ab initio Simulation Package (VASP) [51,52]. The projector augmented wave (PAW) potential [53] was used to describe the interaction between ions and electrons. Exchange and correlation functionals were taken in a form developed by Perdew and Wang [54] based on generalized gradient approximation (GGA). Unless otherwise stated, we mainly used the $4 \times 4 \times 4$ supercell containing 128 lattice points, the $5 \times 5 \times 5$ supercell containing 250 lattice points being used only when the interstitial atoms are more than one, and the $6 \times 6 \times 6$ supercell containing 432 lattice points being used to determine the interaction range between SIA and Re/Os. Through the convergence tests for the system, the cutoff energy was set to be 350 eV and a Monkhorst-Pack *k*-points grid density of $3 \times 3 \times 3$ was employed [55]. The structure optimization was continued until the forces on all atoms reached less than 10^{-3} eV/Å. Both atomic positions and supercell volume and shape were relaxed in all calculations. Migration energy barriers were obtained by the climbing image-nudged elastic band (CI-NEB) method [56,57].

3. Results and discussions

3.1. Thermodynamic stability of interstitial-mediated Re/Os cluster in W

3.1.1. Stability of a single SIA and Re/Os-SIA pair

Although a lot of studies have been made to investigate the stability of a single SIA and Re/Os-SIA pair (Re/Os-W mixed dumbbell) in W, we also revisited it here because the most stable configurations of a single SIA and Re-SIA pair are still under debate. To determine the stability of SIA and Re/Os-W mixed dumbbell in different directions, we calculate the formation energies of them.