Journal of Nuclear Materials 480 (2016) 202–206

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

First-principles study of the interaction between helium and the defects in tantalum

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ARTICLE INFO

Article history:
Received 11 December 2014
Received in revised form
18 July 2016
Accepted 2 August 2016
Available online 4 August 2016

Keywords:
Helium behaviors
Density functional theory
Tantalum

ABSTRACT

As a low power spallation target or target cladding material, tantalum has almost the same defects productions as tungsten under irradiation. In this paper, the detailed analysis of helium behavior in tantalum has been conducted based on density functional theory. The formation energy of tetrahedral interstitial configuration of helium is 0.3 eV lower than that of octahedral interstitial configuration, which means the tetrahedral interstitial configuration of helium is more stable than the octahedral configuration. Tetrahedral helium atoms in tantalum are predicted to have a very low migration, about 0.09 eV. A large binding energy between He and vacancy 2.03 eV indicates that He atom is easily trapped by vacancy, and a tetrahedral interstitial helium atom need overcome about 0.19 eV energy barriers when it directly jumps into the vacancy. Finally, the helium behavior in tantalum is compared with that in tungsten. It is obvious that the binding energy in tantalum is significantly different from those in tungsten. The interaction between two helium atoms in tantalum shows repulsive or weak repulsive other than attractive in tungsten, which suggests that helium atoms are easy to move other than to be a cluster in tantalum.

1. Introduction

Due to high neutron production and high melting point, tungsten is being considered for use as a target material in many spallation neutron sources such as CSNS [1], ISIS [2] and KENS [3]. Another refractory metal tantalum also can yield many neutrons under spallation conditions that are comparable to tungsten. Although tantalum has much higher afterheat and activity than tungsten when it is bombarded by high-energy protons or neutrons, it surpasses tungsten in ductility, workability and weld ability [4,5]. Furthermore, it forms highly stable anodic films on its surface, which increases tantalum chemical stability largely. Thus tantalum is a candidate target or target cladding material for low power spallation neutron sources [6,7].

The spallation neutron sources are usually propelled by protons whose energies range from 800 MeV to 3 GeV. At such high energies, the spallation neutrons lead to the production of large amounts of helium, hydrogen and new heavier species in forms of transmutation products. The tungsten and tantalum have similar helium production cross sections under spallation reaction [8]. These large amounts of helium, hydrogen and heavier transmutation products may exacerbate the displacement radiation damage [9]. Recent experiments led to the conclusion that helium caused a larger increase in yield stress than that due to displacement damage and any irradiation-induced precipitation hardening. An increase in the ductile-brittle transition temperature (DBTT) had been found experimentally due to helium and there is a linear relationship between ΔDBTT and helium concentration for steel irradiated over the range 2.5–18.4 dpa and 85–1530 appm He [10,11].

Much attention has been paid in helium production and much is still devoted to the study of the behavior of helium in metals such as Helium migration, diffusion, trapping, dissolution as well as their relation with the change in mechanical properties [12–15].

Because of the highly important role of helium in irradiated metals, especially for spallation neutron source and fusion materials, there are extensive works on tungsten, iron or zirconium

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http://dx.doi.org/10.1016/j.jnucmat.2016.08.003
0022-3115/

alloys [12–18]. Thus this work aims at investigating the formation energy and migration behavior of helium in tantalum. Firstly, the favorite positions of the spallation defects in tantalum are investigated by determining the formation energy of these defects. Then the interaction between He and other spallation defects is considered, and there exists a large binding energy between He and vacancy. Finally the helium-helium binding energies in tantalum is compared with those in tungsten. We surmise due to the sharp drop of the *5d* projected density of states around Fermi level, the interactions between helium atoms in tantalum are obviously different from those in tungsten.

2. Defect formation energy in tantalum

Although helium, hydrogen and self-interstitial atoms (SIA) are relatively rare in metals compared with vacancies under normal condition, they are plentiful and of prime importance in high-energy radiation environment. For example, under spallation helium can be produced in target by (n,α) transmutation reactions and the ratio of helium production to displacement (He/dpa ratio) is much higher than fission neutrons or even fusion neutrons [19]. For tantalum, helium forms by a two-step reaction of ^{181}Ta. Step one, ^{181}Ta transmutes to ^{182}Ta with thermal neutrons by (n, gamma) radiative capture; step two, helium forms in ^{182}Ta with fast neutrons by (n, a) reaction. We firstly calculate the helium, hydrogen, self-interstitial atoms (SIA), and vacancy formation energies for tantalum.

In body centered cubic (bcc) metal all substitution positions are equivalent and there are two interstitial positions: the octahedral and the tetrahedral positions. The location of helium in metals influences its migration; the more favored interstitial position in tantalum is calculated first. The following calculation had been performed on the basis of the frozen-core projected augmented wave (PAW) method within the Vienna *ab initio* Simulation Package VASP [20], where the exchange and correlation effects are described by the density functional theory (DFT) within the generalized gradient approximation (GGA) of Perdew and Wang [21]. For the plane-wave set, a cut off energy of 350 eV is used [12]. Except when otherwise mentioned, all the results presented below are obtained using 128 atoms and a $3 \times 3 \times 3$ k-points grid. The super-cell contains 128 atoms are fully relaxed until the Hellmann-Feynman forces become less then 0.02 eV/Å. The equilibrium lattice parameters obtained for tantalum is 3.3094 Å, which is a little higher than the experimental data 3.3013 Å [22].

The helium, self-interstitial atoms and vacancy formation energies in tantalum were calculated by

$$E^f_{He} = E(NTa + He) - E(N) - E(He_{isolated}), \tag{1}$$

$$E^f_I = E_{total}(N+1) - \frac{N+1}{N}E_{tot}(N), \tag{2}$$

$$E^f_V = E_{total}(N-1) - \frac{N-1}{N}E_{tot}(N). \tag{3}$$

where $E(NT_a + He)$ is is the energy of the super-cell containing N Ta atoms and 1 He atom, and $E(N)$ is the reference energy of Ta in a perfect bcc structure, the $E(He_{isolated})$ is an isolated He atom, $E_{toal}(N+1)$, $E_{total}(N-1)$ are the total energy of the supercell with a self-interstitial and with a vacancy, respectively.

Table 1 gives the formation energies of the defects in tantalum. As a consequence the predicted favored interstitial site of helium in tantalum is tetrahedral site and the energy difference between octahedral and tetrahedral is 0.31 eV, as shown in Table 1. The results shows that the formation energy of hydrogen in tantalum is negative, which means tantalum can absorb hydrogen atoms. The <111> dumbbell is the most stable configuration in tantalum. In this paper, we investigate the kinetics of helium in tantalum which is very important for the understanding the He migration, bubble nucleation or dissociation.

3. The interaction between helium and the other defects in tantalum

The Fundamental properties of helium in metals including the equilibrium structure and the migration barrier are usually inferred from direct measurements on irradiated samples. There exist some experiments on investigating the mechanical properties after spallation irradiation; however the migration, clustering or dissolution of helium at the high defect densities in tantalum may obscure the interpretation of experimental data.

Firstly He migration is examined. Here the climbing image nudged elastic band (CINEB) method is employed to find the minimum energy path (MEP) of the He atom migration when the initial and final configurations are known. Migration energy barrier is defined as the energy difference between an initial configuration and the saddle point.

A tetrahedral He will migrate between two equivalent sites without passing through an octahedral site. Very low migration energy, 0.09 eV, is found for He in tantalum, as shown in Fig. 1. Such a low migration energy means that the migration of interstitial He is almost athermal.

Since it is easy that He atom mobiles in tantalum, it will encounter other defects such as another He atom, Hydrogen atom, vacancy or SIA, even in very low He concentration. We now turn to the interaction between the He atom and the other defects. Here we assumed that the defects are located their most stable positions. The binding energy for helium-helium atoms is obtained as

$$E^b(He_1, He_2) = E^f(He_1) + E^f(He_2) - E^f(He_1, He_2), \tag{4}$$

and the binding energy for helium and other defects is obtained as

$$E^b(He, Vac.) = E^f(He) + E^f(Vac.) - E^f(He, Vac.), \tag{5}$$

$$E^b(He, H) = E^f(He) + E^f(H) - E^f(He, H), \tag{6}$$

$$E^b(He, SIA) = E^f(He) + E^f(SIA) - E^f(He, SIA), \tag{7}$$

where $E^f(He)$, $E^f(Vac.)$, $E^f(H)$, $E^f(SLA)$ are the formation energies for a tetrahedral helium atom, a vacancy, a tetrahedral hydrogen atom, and SIA, respectively. $E^f(He/Vac./H/SLA)$ is the formation energy that super-cell contains a helium atom and a nearest vacancy, or a tetrahedral hydrogen atom, or SIA atoms with the <111> configuration. Table 2 shows that the interactions in tantalum between two helium atoms, between a helium atom and a hydrogen atom, and between a helium atom and SIA atoms are repulsive. But the interaction between a helium atom and a vacancy is attractive. Furthermore, the attraction between a vacancy and a helium atom is so strong that the helium atom will be trapped by the vacancy and we do find the helium atom relaxes a lot towards to the vacancy position. It thus explains why small diffusion constants of helium in tantalum are obtained in the previous experiments, in which the authors thought the small value hinted to strong helium trapping by vacancies and He-V cluster formation [23].

Since there exist some interactions between helium and the other defects, the diffusion of a tetrahedral helium atom is considered, in the presence of another a hydrogen atom, or a