Journal of Nuclear Materials 457 (2015) 80–87

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

The influence of high grain boundary density on helium retention in tungsten

G. Valles [a,*], C. González [b], I. Martin-Bragado [c], R. Iglesias [b], J.M. Perlado [a], A. Rivera [a]

[a] Instituto de Fusión Nuclear UPM, José Gutiérrez Abascal 2, 28006 Madrid, Spain
[b] Departamento de Física, Universidad de Oviedo, C/ Calvo Sotelo, s/n, Oviedo, Spain
[c] IMDEA Materials Institute, C/ Enric Kandel 2, 28906 Getafe, Madrid, Spain

HIGHLIGHTS

- Comparison between monocrystalline and nanostructured irradiated tungsten.
- OKMC parameterization published and new DFT data.
- Important role of grain boundary density on defect evolution.
- Cluster pressurization much lower in nanostructured tungsten.
- Promising expectations on nanocrystalline tungsten in view of results.

ARTICLE INFO

Article history:
Received 14 July 2014
Accepted 15 October 2014
Available online 29 October 2014

ABSTRACT

In order to study the influence of a high grain boundary density on the amount, size and distribution of defects produced by pulsed helium (625 keV) irradiation in tungsten, we have carried out Object Kinetic Monte Carlo (OKMC) simulations in both monocrystalline and nanocrystalline tungsten. The parameterization of the OKMC code (MMonCa) includes binding energies obtained with our in-house Density Functional Theory (DFT) calculations. In the interior of a grain in nanocrystalline tungsten the mixed He_nV_m clusters are larger and have a lower He/V ratio. Thus, they are less pressurized clusters. The total elastic strain energy remains almost constant with the increasing number of pulses, contrary to its increase in monocrystalline tungsten. A better response to helium irradiation is therefore expected in nanocrystalline tungsten, opening a new path to investigate these nanostructured materials for fusion purposes.

1. Introduction

Tungsten has been proposed as a convenient material for plasma-facing components (PFC) in future fusion reactors since it offers several advantages: high melting point, high thermal conductivity, low sputtering coefficient and low tritium retention. Owing to these properties, tungsten is expected to be suitable for the first-wall and the divertor in magnetic fusion reactors and the first-wall (armour) in inertial fusion confinement chambers [1–4]. In both technologies the most adverse irradiation events are of pulsed nature: in the case of IFE (Inertial Fusion Energy) due to the target explosions in which the technology is based and in the case of MFE (Magnetic Fusion Energy) because of the edge-localized modes [5], which up to now (e.g., ITER) are intrinsic to TOKAMAK operation in H-mode.

In direct-target IFE, the energy will be released as follows: ~1% in the form of X-rays with a pulse duration of a few ns, not considered to be a great threat to the wall; ~71% due to pulsed neutron irradiation, of which energy will be absorbed beyond the first-wall; and ~27% due to pulsed ion irradiation (burn products and debris ions) [6,7]. The resulting intense ion pulses will cause, on the one hand, very high thermal loads and on the other hand, damage due to Frenkel-Pair (FP) production and ion retention. Provided that the first wall is situated far enough from the target explosions, an acceptable thermo-mechanical response is feasible [8–10]. As regards the damage, different ions and burn products will reach the first wall, He ions among them [11]. The energy of He ions can be as high as 4 MeV with an average value of ~2.5 MeV after thermalization in the compressed target [12], more than enough to produce Frenkel Pairs. Since He is not soluble in tungsten, He atoms tend to nucleate inside vacancies, which could result under certain circumstances in dramatic He bubble formation [13,14]. In general, He_nV_m clusters (not only large bubbles) cause microstructural changes [15,16], that may develop into blistering, cracking

* Corresponding author.
E-mail address: gonzalovallesalberdi@hotmail.es (G. Valles).

http://dx.doi.org/10.1016/j.jnucmat.2014.10.038
0022-3115/

and exfoliation of the material [17,18], all of them detrimental to the armour [19]. Experimental results show that continuous He irradiation (typical flux 10^{10}–10^{14} cm^{-2} s^{-1}) leads to deleterious effects (adverse porosity) at fluences higher than 10^{17}–10^{18} He cm^{-2} [15,17]. However, Renk et al. [6] showed that pulsed He irradiation (with fluxes up to 2×10^{19} cm^{-2} s^{-1}) leads to detrimental effects (pore formation and protrusions) at fluences as low as 10^{15} He cm^{-2}.

Many efforts are being carried out in order to mitigate the effect of He retention. A possible solution would be the use of nanostructured tungsten due to its large grain-boundary density: As grain boundaries may act as defect sinks for vacancies, interstitials and He atoms, nanostructured tungsten is expected to present higher irradiation tolerance [20]. The defects are supposed to accumulate at grain boundaries, where vacancies and self interstitial atoms (SIAs) annihilate [21]. On the other hand, He atoms can become trapped if the binding energy is high enough and diffusivity along the grain boundaries is low. Nanocolumnar tungsten has recently been fabricated with a high grain boundary density [22] and it has been experimentally observed that a high grain boundary density has a direct influence in the retention of light species [23]. A point not elucidated yet is whether inter-grain He migration allows for efficient He release or if the distribution and size of defects in the interior of a grain is affected by the high density of grain boundaries.

In the present study, we investigate the influence of high grain-boundary density in damage production by pulsed He irradiation in tungsten, by comparing monocrystalline tungsten (MW) to nanocrystalline tungsten (NW). We have carried out OKMC (Object Kinetic Monte Carlo) simulations parameterized with the aid of DFT calculations to describe the damage distribution in both materials. MMonCa code was used for this purpose [24,25]. In order to compare the influence of high grain-boundary density in the damage distribution in both nanocrystalline and monocrystalline tungsten, we have carried out two different sets of simulations with the experimental conditions used by Renk et al. in Ref. [6].

2. Simulation methods

2.1. Density Functional Theory (DFT) calculations

Calculations based on DFT techniques were performed using the *Vienna Ab initio Simulation Package* (VASP) [26–28]. The PBE [29] parameterization of the Generalized Gradient Approximation (GGA) for the exchange and correlation functional was used as well as the Plane Augmented Wave pseudopotentials [30], provided by the code. Six valence electrons have been considered for W (4 3*d* and 2 4*s*) and two 1*s* valence electrons for He. Within these approximations, the lattice parameter was estimated to be 3.172 Å, using an energy cutoff for the plane waves of 479 eV. The $5 \times 5 \times 5$ cubic supercell (250 W atoms) used in this work is built repeating the unit cell five times along each direction while 64 *k*-points sampled the first Brillouin zone. Several He atoms will be placed inside the unit cell in combination with *n*-vacancies (for *n* up to 4). Each structure is fully relaxed until the forces on all atoms are smaller than 0.025 eV/Å. The resulting final total energy is used to obtain the formation and binding energies for the subsequent OKMC simulations. The general equation giving the formation energy for a system formed by N_W and N_{He} atoms can be written as:

$$E_f = E_{tot} - N_W E(W) - N_{He} E(He) \quad (1)$$

where E_{tot} is the final total energy obtained in the relaxation of each supercell, $E(W)$ is the atomic energy of one metal atom inside the pure bulk when a $5 \times 5 \times 5$ supercell is used and $E(He)$ corresponds to an isolated He atom located inside an otherwise empty $5 \times 5 \times 5$ box with sides equal to the equilibrium lattice parameter of the pure metal. On the other hand, the binding energy E_b corresponds to the energy released when two objects (in general more) merge to become a mixed one. It is defined as the formation energy difference between a system in which the objects are close together and a system in which the objects are far apart. Therefore, the formation energy of the configurations where the objects are separated is usually calculated individually in a supercell for each of the objects, as defined in Eq. (1). The general expression for E_b for a system with N_{def} defects (SIAs, n-vacancies and/or He atoms) can then be written as:

$$E_b(N_{def}) = \sum_{i_{def}=1}^{N_{def}} E_f(i_{def}) - E_f(N_{def}) \quad (2)$$

2.2. Object Kinetic Monte Carlo (OKMC)

OKMC simulations were carried out with our recently developed OKMC Open Source code MMonCa [24,25]. OKMC allows simulating large spatial regions (up to several μm^3) on large timescales (from minutes to hours). As described in Ref. [24] He atoms, SIAs, vacancies and their clusters are considered as objects, which can migrate, dissociate from or agglomerate into clusters, desorb from free surfaces or annihilate in the case of vacancies (V) and self interstitial atoms (SIAs), often abbreviated simply as interstitials (I), i.e., V + I → Ø. Cluster agglomeration, desorption and annihilation events take place when two objects interact geometrically (an interaction radius is defined for each object). Migration and dissociation probabilities are calculated via an Arrhenius law:

$$\nu = \nu_0 \cdot e^{-\left(\frac{E^{event}}{k_B T}\right)} \quad (3)$$

where ν_0 is the prefactor, k_B the Boltzmann constant, T the temperature and E^{event} is the corresponding activation barrier (migration or dissociation energy). The dissociation energy is the sum of the migration energy of the emitted object plus the binding energy of this object to the trapping cluster. Clusters can only emit single particles (a single vacancy, a SIA or a He atom in this work). The prefactor and the activation energy must be set for each migration or dissociation event.

We have parameterized object migration values in MMonCa following the scheme published by Becquart et al. [31,32]. The prefactors and migration energies for all mobile objects can be seen in Table 1. Mixed He_nV_m and He_nI_m clusters are considered immobile (with the exception of HeV, HeV_2 and HeV_3 [33]). Pure He cluster

Table 1
Prefactor and migration energy (eV) for mobile objects: He, single vacancy, SIA, pure He clusters, vacancy clusters and SIA clusters as published by Becquart et al. in Ref. [32]. The values of the constants are $\nu_0 = 6 \times 10^{12}$ s^{-1}, $q = 1000$ and $s = 0.5$. In the case of HeV_m (only HeV, HeV_2 and HeV_3), migration energies have been calculated by us in Ref. [33]. We have considered their prefactors as ν_0/m, in order to reproduce the movement of the whole cluster as the result of single jumps of the *m*-vacancies.

Object	Prefactor (s^{-1})	Migration energy (eV)
He	$10^{-2}\ \nu_0$	0.01
He_2	$10^{-2}\ \nu_0$	0.03
He_3	$10^{-2}\ \nu_0$	0.05
He_n, $n > 3$	$10^{-2}\ \nu_0$	$E_{mig}(He_{n-1}) + 0.01$
V	ν_0	1.66
V_m, $m > 1$	$\nu_0\ (q^{-1})^{m-1}$	1.66
I	ν_0	0.013
I_m, $m > 1$	$\nu_0\ m^{-s}$	0.013
HeV	ν_0	4.83
HeV_2	$\nu_0/2$	2.04
HeV_3	$\nu_0/3$	1.94