Nuclear Instruments and Methods in Physics Research B 352 (2015) 96–99

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

Nucleation and growth of hydrogen bubbles on dislocations in tungsten under high flux low energy plasma exposure

Petr Grigorev [a,b,c,*], Dmitry Terentyev [a], Vladimir Dubinko [d], Giovanni Bonny [a], Guido Van Oost [b], Jean-Marie Noterdaeme [b], Evgeny E. Zhurkin [c]

[a] SCK•CEN, Nuclear Materials Science Institute, Boeretang 200, Mol 2400, Belgium

[b] Ghent University, Department of Applied Physics EA17 FUSION-DC, St. Pietersnieuwstraat, 41 B4, B-9000 Gent, Belgium

[c] Department of Experimental Nuclear Physics K-89, Institute of Physics, Nanotechnology and Telecommunications, St. Petersburg State Polytechnical University, 29 Polytekhnicheskaya Str., 195251 St. Petersburg, Russia

[d] National Science Center, Kharkov Institute of Physics and Technology, Kharkov 61108, Ukraine

ARTICLE INFO

Article history:
Received 10 July 2014
Received in revised form 10 October 2014
Accepted 29 November 2014
Available online 26 December 2014

Keywords:
Hydrogen retention
Tungsten
Bubble

ABSTRACT

A new mechanism for the nucleation and growth of hydrogen (H) bubbles on dislocations under plasma exposure of tungsten was recently proposed on the basis of direct *ab initio* calculations. Density functional theory calculations demonstrated that H atoms are strongly bound to a screw dislocation core and exhibit fast one-dimensional migration along its line. Once the number of hydrogen atoms trapped on a dislocation segment exceeds eight, the emission of a jog occurs thereby converting a pure H_N cluster into a H_{N+1}-jog configuration. On the basis of these results a kinetic model was formulated to evaluate the conditions (i.e., range of temperature and flux exposure) for the transformation of pure H clusters into supercritical hydrogen–vacancy clusters attached to the dislocation line. In this work, a parametric study employing the kinetic nucleation model was performed to derive the hydrogen bubble formation energy function that offers the best agreement with available experimental results. The obtained results allow one to rationalize the depth and temperature dependence of the experimentally observed hydrogen deposition after high flux low energy plasma exposure for ITER relevant conditions.

1. Introduction

Tungsten is the main candidate material for plasma facing components in future plasma devices. It was chosen as the divertor armor in ITER and is planned to be used in DEMO [1]. Plasma facing materials in fusion devices will be exposed to severe conditions in terms of heat loads and particle flux. Moreover, strict safety limitations are imposed on the amount of tritium accumulated in the reactor's chamber, because it is a toxic, radioactive and expensive material. For ITER the limit was set to be 700 g [2]. From this point of view understanding the mechanism of hydrogen isotopes (HI) retention is important in order to predict the tritium accumulation in the reactor and define its operational regimes.

There is a large amount of experimental studies on HI retention in tungsten (W) available in the literature and an extensive review can be found in [3,4]. Experiments involving plasma exposures at ITER relevant temperatures (400–800 K [1]) showed that after exposure below 600 K, HI sub-surface bubbles and surface blisters are formed [5–9]. Such surface modifications can lead to the ejection of W atoms or flakes in case of a blister rupture, leading to plasma disruption and consequently to the reduction in the performance of the fusion device. Current models of bubble nucleation and growth [9,10] are based on the assumption that trapping of HI originates at vacancies created during implantation and followed by their subsequent growth by clustering HI and vacancies. Such approaches have shown a very good agreement with experiments involving ion beams exposure with an ion energy in the range 5–30 keV/ion [10]. The energy of such HI ions is above the threshold energy for the creation of displacement damage in W (threshold displacement energy for W is 45–90 eV [11]), and therefore it leads to the creation of stable Frenkel pairs, i.e., vacancy–interstitial defect pairs. More realistic conditions for the ITER environment are reached in experiments involving linear plasma generators where the energy of ions is around 50–100 eV [5–7]. This is significantly lower than the threshold energy for the generation of Frenkel pairs and such irradiation conditions will be referred to as "sub-thresholds conditions". Thus, during the sub-threshold plasma exposure no vacancies are created directly by plasma ions due to atomic displacement and the vacancy-trapping

* Corresponding author at: SCK•CEN, Nuclear Materials Science Institute, Boeretang 200, Mol 2400, Belgium.

http://dx.doi.org/10.1016/j.nimb.2014.11.103
0168-583X/© 2014 Elsevier B.V. All rights reserved.

nucleation mechanism imposed in the current numerical models is not relevant. However, intensive blistering is also observed under sub-threshold plasma exposures, for which the mechanisms of bubble nucleation and growth are not properly understood yet. Homogeneous nucleation of hydrogen clusters is highly unlikely due to a very low binding energy between two hydrogen atoms in the bulk ~0.01 eV [12], which does not allow reaching a cluster of critical size to punch out a self-interstitial atom and convert into a stable nucleus.

Recently, a new model of HI retention was proposed, based on the assumption that dislocation networks serve as nucleation sites for HI bubbles [13]. This assumption was confirmed by *ab initio* calculations addressing the interaction of hydrogen with a ½⟨1 1 1⟩ screw dislocation, which showed that the dislocation can attract up to six H atoms and that the migration barrier of a H atom along the dislocation line (computed to be 0.15 eV) is significantly lower than in the bulk, i.e., 0.24–0.27 eV [14–16]. Moreover, the calculations suggested that the so-called jog-punching mechanism of bubble growth is initiated as soon as eight hydrogen atoms group on a screw dislocation segment. The jog-punching implies a nucleation of two anti-jogs, so that hydrogen atoms occupy a vacancy-type jog, while an interstitial-type jog migrates away along the dislocation line. A rate theory model based on these computational results has shown good agreement with the experimental trends regarding the saturation of HI retention with increasing implantation dose [5,6]. In this work we implement the previously formulated rate theory model to perform a numerical integration and to investigate the exposure conditions (i.e., flux and temperature) resulting in the bubble growth and blister formation.

2. Model description

A schematic picture of the system used in simulations is shown in Fig. 1(a). This is a one-dimensional system where material is divided into slabs of equal width λ_d. Each slab corresponds to a single nucleation site and is fed with H by a source term defined from the steady state solution for the bulk concentration of H atoms discussed in [13]:

$$S(x) = \omega Z \rho R F \exp(-x\sqrt{Z\rho}), \tag{1}$$

with Z – geometrical factor ($Z = 1$), ρ – dislocation density, ω – atomic volume, F – particle flux, R – particle range.

H atoms accumulate into H_N clusters at dislocation trapping sites following a binding energy function, and H atom can be dissolved from a H_N clusters at a rate defined by the equation:

$$R^-(i) = \nu C_T(i) \exp(-E_{bin}(N)/k_B T), \tag{2}$$

with ν – the attempt frequency, $C_T(i)$ – the trap (or trapped cluster) concentration at slab number i, $E_{bin}(N)$ – the binding energy of a H atom to the trapped H_N cluster, k_B – Boltzmann constant. The amount of H atoms that escapes from the traps is redistributed between neighboring slabs. Thus, the amount of trapped H at each nucleation site is defined by the balance of trapping, dissolution and exchange between neighboring slabs (Numbers $i-1$ and $i+1$):

$$\frac{\partial C_H(i)}{\partial t} = S(x) + 0.5[R^-(i-1) + R^-(i+1)] - R^-(i), \tag{3}$$

The size of H_N clusters growing during the exposure is determined by a simulation that follows the number of vacant sites generated by the jog-punching mechanism. To introduce the jog punching mechanism that allows a bubble to grow, we must introduce a threshold value for the number of trapped H atoms per vacancy (N_H/N_{Vac}) beyond which the jog punching mechanism starts to operate. The main parameters of the model, which define the conditions for the bubble growth, is the binding energy of a H atom to a trapped H_N–V_M complex, and the threshold size for a H_N cluster beyond which it is converted into H_{N+1}-jog configuration.

3. Parametrization of the model

To parameterize the model, let us first consider the binding energy of H_N to a screw dislocation and to a vacancy jog on the screw dislocation. Fig. 1(b) compares the binding energy of a H to a H_{N-1} cluster trapped at a vacancy [17–19] and trapped at the dislocation vacancy jog [13], obtained by *ab initio* calculations from the cited works. As can be seen, there is no significant difference between the evolution of the binding energy at a vacancy and at the dislocation jog. The red dashed line is a fit of these *ab initio* results, which are valid for small H_N clusters forming as nuclei for H bubbles. For the well-developed H bubbles, one cannot apply the *ab initio* fitted function. As a limiting case we used the formula obtained within the so-called liquid tear drop (LTD) model, parameterized using a recently developed W–H–He potential in [20], and based on the balance of volume and surface energy, shown in Fig. 1(b).

To define the jog punching threshold parameter (N_H/N_{Vac}), let us consider the loop punching mechanism. Pressure is exerted on the bubble surface and the critical one needed to create a dislocation loop is defined as [21]:

$$P_{lp} = 2\gamma/R_b + \mu b/R_b \tag{4}$$

where $\gamma = 2.65$ N/m is the surface tension, $G = 158.6$ GPa is the shear modulus, $b = 2.7$ Å is the Burgers vector of the ½⟨1 1 1⟩ loop, and R_b is the bubble radius. The values for the surface tension (γ) and shear modulus (G) were taken from [22,23]. The variation of the critical pressure as a function of bubble size (expressed as a number of vacant sites) is plotted in Fig. 2(a) on the right-hand side Y axis. To estimate the critical concentration of hydrogen contained in

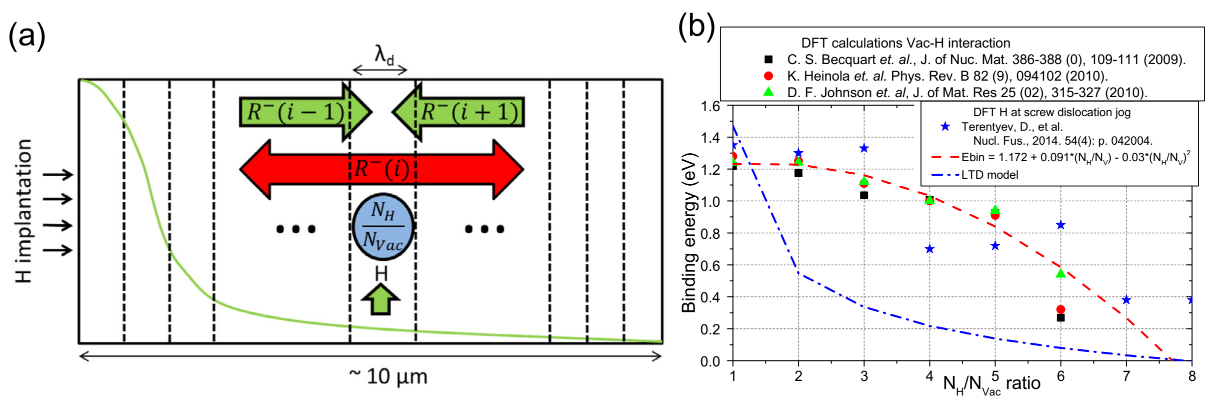

Fig. 1. (a) Schematic description of the model; (b) bubble-H binding energy data and approximations.