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An object Kinetic Monte Carlo Simulation of the dynamics of helium and point defects in tungsten

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ABSTRACT

In the near surface of plasma facing materials, high concentrations of hydrogen and helium isotopes can build up, which will interact with the point defects resulting from the bombardment of the surface as well as with the impurities of the materials. It is important to develop an understanding of the evolution of W microstructure in such conditions and to be able to model this evolution. The task is very complex, as many elements have to be included in the model which must be all parameterized correctly. Isochronal annealings experiments are simple experiments which can help in the making of more complicated models. In this work, an object Kinetic Monte Carlo technique parameterized on ab initio calculations as been used to model He desorption in W. The He atoms and the self interstitial atoms have been found to be very mobile but they can bind quite strongly with impurities such as carbon or molybdenum atoms. The evolution of the number of defects in the Kinetic Monte Carlo simulation was found to be in good agreement with the resistivity changes observed during an He desorption experiment of above threshold He implantation in a thin wire of tungsten.

1. Introduction

One of the promising candidates for the divertor plate in International Thermonuclear Experimental Reactor (ITER) is tungsten, because of its high melting temperature, high thermal conductivity and low sputtering erosion. During its lifetime, the divertor surface will be impinged by high energy particles: helium, hydrogen isotopes and neutrons. These isotopes will interact with the point defects resulting from the displacement cascades induced by the high energy particles as well as with the impurities of the materials. These interactions will induce changes in the microstructure and thus in the mechanical properties. The overall objective of this work is to simulate radiation damage in tungsten with He production in order to predict the evolution of the microstructure and the possibility of swelling. The object kinetic Monte Carlo (OKMC) method is one of the tools which can be used to model the microstructure evolution under such conditions. However, for this modeling, the elementary physical phenomena associated with the point defects created and their interaction with the He produced have to be determined. As these phenomena take place at the atomic level, this has to be done with the help of ab initio calculations. Ab initio calculations have thus been used to parameterize an object Kinetic Monte Carlo code and simulate He desorption in W.

In a first part the OKMC model which has been used is presented. In a second part the most important parameters of the OKMC are described and compared with experimental results when possible. The OKMC code is then used in the third part to simulate an isochronal annealing experiment of He desorption in W.

2. Methodology

The OKMC code LAKIMOCA developed at EDF has been modified to take into account He.

The general features of the LAKIMOCA code have been extensively described in a previous publication [1]. Briefly, the model treats radiation produced defects (vacancies, self interstitial atoms (SIA) and their clusters) as well as foreign interstitial atoms (FIA – He in the present work) as objects with specific positions in a simulation box and with associated reaction volumes. The different events are presented in Fig. 1. Each object can migrate or emit single entities (a vacancy, a SIA, a FIA) and participate in a series of predefined reactions. The probabilities for physical transition mechanisms Γ_i, which are basically migration jumps and emission from larger defects or from traps, are calculated in terms of Arrhenius frequencies for thermally activated events,

$$\Gamma_i = \nu_i \exp\left(\frac{E_{a,i}}{k_B T}\right), \tag{1}$$

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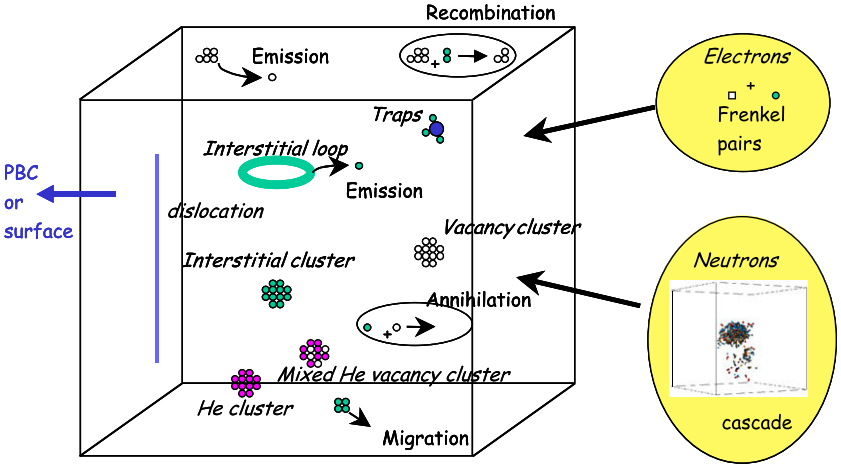

Fig. 1. Summary of the different events: migration, recombination, emission of single entities or trapping as well as electron or neutron irradiations, taking place in an object KMC simulation using the LAKIMOCA code. The dimensions of the box are $199a_0 \times 200a_0 \times 201a_0$, where a_0 is the lattice parameter of tungsten.The white spheres are the vacancies, the green ones, the SIAs; the pink ones represent the helium atoms (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

where ν_i is the attempt frequency (prefactor) for event i, $E_{a,i}$ is the corresponding activation energy, k_B is Boltzmann's constant and T is the absolute temperature. After a certain event is chosen, time is increased according to the residence time algorithm [2],

$$\Delta\tau = 1 \Big/ \sum_{i=1}^{N_e^{th}} \Gamma_i + \sum_{j=1}^{N_e^{ext}} P_j \tag{2}$$

where P_j are the probabilities of external events, such as the appearance of a cascade, of isolated Frenkel pairs produced by impinging particles, or of implanted FIAs. In addition, the model includes non-thermally activated events, such as the annihilation of a defect after encountering either a defect of opposite nature (i.e. a SIA encountering a vacancy) or a sink, as well aggregation, either by adding a point defect to a cluster or by forming a complex between a defect and a trap for it. These events occur only on the basis of geometrical considerations (overlap of reaction volumes) and do not participate in defining the progressing of time. The possibility of introducing different classes of immobile traps and sinks, characterized by specific geometrical shapes (spheres, infinite cylinders, surfaces, etc.) and suitable to mimic voids or other trapping nanofeatures, as well as dislocations and grain boundaries, is also implemented. The code is therefore equipped to mimic fairly realistic microstructures and irradiation conditions.

The choice of the parameter set is a difficult task and quite an open question. For each object, one has to define all the possible events that that object can perform, with their appropriate probabilities. The object properties are: the types of objects (e.g. a single vacancy v, a He atom, a di-vacancy, 2v, a single self interstitial atom (SIA), etc.), the forward reactions that these objects can perform with the appropriate probabilities and capture radius (e.g. the annihilation of a vacancy with a self interstitial: v + SIA $\rightarrow \varnothing$, or the formation of a di-vacancy: v + $v \rightarrow$ 2v), the backward reactions with their probabilities (e.g. the dissociation of a vacancy from a di-vacancy).

To obtain the OKMC data, ab initio calculations in the framework of the functional density and the Vienna Ab initio Simulation Package VASP [3] were used. The calculations were performed in the framework of Blöchl's projector augmented-wave (PAW) method [4] within the Generalized Gradient Approximation (GGA) of Perdew and Wang [5]. The pseudo-potentials were taken from the VASP library. The supercell approach with periodic boundary conditions (PBC) was used to simulate point defects as well as pure phases. Brillouin zone sampling was performed using the Monkhorst and Pack scheme [6]. The plane wave cut-off energy was 350 eV in order to get converged results. 54 atom supercells with 125 kpoints as well as 128 atoms with 27 kpoints were used to check the convergence of the calculations with supercell size. For clarity and because the results were converged, only the results obtained with the largest supercell size, i.e. the 128 atom supercell calculations will be presented in this article. All the structures have been relaxed by conjugate gradient, keeping the volume constant. The uncertainty on the ab initio results is 0.01 eV.

The binding energy $E_b(A_1, A_2)$ between two entities A and B is obtained as

$$E_b(A_1, A_2) = [E(A_1) + E(A_2)] - [E(A_1 + A_2) + E_{\text{Ref.}}] \tag{3}$$

where E_{Ref} is the energy of the supercell without A_1 and A_2, $E(A_j)$ is the energy of the supercell containing A_j only and $E(A_1 + A_2)$ is the energy of the supercell containing both A_1 and A_2 in interaction with each other. All the supercells contain the same number of metal sites, i.e. have the same size. Except when otherwise stated, the reference state of the binding energies presented in this work is always the energy of a supercell without any defects, i.e. a perfect crystal. With such a scheme a positive binding energy means attraction between the entities. For more than two entities, Eq. (3) can be extended as follows:

$$Eb(A_1, A_2, \ldots, A_n) = \sum_{i=1,\ldots,n} E(A_i) - [E(A_1 + A_2 + \cdots + A_n) + (n-1)E_{\text{ref}}] \tag{4}$$

with the same convention as in Eq. (3)

3. Results

3.1. Helium and vacancy clustering

The most stable interstitial configuration for He is the tetrahedral (T) site, the energy difference between the two possible interstitial sites ΔE_{T-O}, where O stands for octahedral, being equal to 0.22 eV. Furthermore, the substitutional configuration is 1.46 eV lower than the tetrahedral one.

The binding energy between interstitial He atoms [10] (Table 1), as well the binding energy of a single He atom or a single vacancy with small He-vacancy complexes have been determined. The results are shown in Table 2.