Macroscopic rate equation modeling of trapping/detrapping of hydrogen isotopes in tungsten materials

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ABSTRACT

Relevant parameters for trapping of Hydrogen Isotopes (HIs) in polycrystalline tungsten are determined with the MHIMS code (Migration of Hydrogen Isotopes in MaterialS) which is used to reproduce Thermal Desorption Spectrometry experiments. Three types of traps are found: two intrinsic traps (detrapping energy of 0.87 eV and 1.00 eV) and one extrinsic trap created by ion irradiation (detrapping energy of 1.50 eV). Then MHIMS is used to simulate HIs retention at different fluences and different implantation temperatures. Simulation results agree well with experimental data. It is shown that at 300 K the retention is limited by diffusion in the bulk. For implantation temperatures above 500 K, the retention is limited by trap creation processes. Above 600 K, the retention drops by two orders of magnitude as compared to the retention at 300 K. With the determined detrapping energies, HIs outgassing at room temperature is predicted. After ions implantation at 300 K, 45% of the initial retention is lost to vacuum in 300 000 s while during this time the remaining trapped HIs diffuse twice as deep into the bulk.

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1. Introduction

Tungsten (W) is a serious candidate material for plasma facing components (PFC) in fusion devices such as ITER and DEMO thanks to its thermal properties. Due to plasma/wall interaction, W-based PFC will be subject to high particle fluxes (10^{20}–10^{24} m^2/s) of hydrogen isotopes (HIs). Fuel particles implanted in the subsurface region of PFC could diffuse into the tungsten matrix and be trapped deeper in the bulk. It causes safety issues because of the regulation-limited amount of tritium in the vessel walls as well as operation concerns due to possible uncontrolled HIs recycling fluxes that can affect global plasma stability. So, migration and trapping of deuterium have been extensively studied; see for instance reviews by Causey [1], Skinner et al. [2] and Tanabe [3].

The development of a global tokamak wall model interacting with hydrogen isotopes by taking into account all the physical processes (particles implantation, migration, trapping, outgassing …) is necessary in order to extrapolate the fuel retention in W-based plasma facing materials for ITER. In this approach, macroscopic rate equations (MRE) models are an efficient way to investigate migration and trapping of HIs in metallic materials from nanometers to centimeters scales. The rate equation model for hydrogen diffusion including hydrogen trapping in materials was originally discussed by McNabb et al. [4] and used for a number of metals such as steel [5,6] and tungsten [7–11].

In the present work, a code based on a rate equations model has been developed to deal with the trapping of HIs in W. It is named MHIMS (Migration of Hydrogen Isotopes in MaterialS) [12] and it can be seen as a light version of the HIIPC models developed by Sang et al. [13] for tokamaks inventory simulations. In addition to the reasonable computational resources necessary for running MHIMS, we will detail here how MHIMS is a good tool to extract, from laboratory experiments, fundamental parameters of the HIs–tungsten interaction and how it can be used for experimentally relevant predictions. MHIMS is able to model particles implantation with traps creation during plasma wall interaction,

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particles depth profiles evolution in the material and Thermal Desorption Spectrometry (TDS) measurements. In the first part of this paper, the equations of the model are described (Section 2). Then, simulation results are benchmarked against well controlled laboratory experiments from the literature performed with poly-crystalline tungsten samples that we consider as a reference case (Section 3). Using this benchmarked set of parameters, we compare MHIMS simulations of the evolution of the retention as a function of fluence and implantation temperature with a larger set of laboratory experiments. The objective of this part of our work is to test the robustness of our model and its predictive ability (section IV). Finally, predictions on deuterium outgassing at room temperature are made and their influences on the interpretation of laboratory experiments are discussed (Section 5).

2. Description of MHIMS model

An earlier introduction to our model can be found in Refs. [12], but we provide here a more comprehensive and updated account. Fig. 1 presents the general energy diagram of a hydrogen atom inside a metal with two types of trapping sites present in the bulk of the material. $E_R + E_S$ is the energy barrier of the hydrogen atom penetration into the metal matrix. E_R is the energy barrier for the H atom to overcome in order to reach the surface (a preliminary step for surface recombination). E_D is the barrier of diffusion of H in the metal matrix through solute sites. $E_{B,1}$ and $E_{B,2}$ are binding energies of HIs located in two different trap types present in the metal. Thus, energy barriers to come out of these traps, called detrapping energies, are respectively $E_{T,1} = E_{B,1} + E_D$ and $E_{T,2} = E_{B,2} + E_D$.

In our MRE model, HIs are split into two populations: mobile (or solute) and trapped species. C_m stands for the concentration of mobile particles and $C_{t,i}$ for the concentration of trapped particles in the ith trap type. In the following, the concentrations will be normalized to the metal density i.e. they are expressed in atomic fraction (at.fr.).

The temporal variation of each population is described by Equations (1) and (2).

$$\frac{\partial C_{t,i}}{\partial t} = -S_{trap,i \to mobile} + S_{mobile \to trap,i} \tag{1}$$

$$\frac{\partial C_m}{\partial t} = D(T) \cdot \frac{\partial^2 C_m}{\partial x^2} - \sum \frac{\partial C_{t,i}}{\partial t} + S_{ext} \tag{2}$$

where $S_{mobile \to trap,i}$ is the source of mobile particles being trapped, $S_{trap,i \to mobile}$ corresponds to trapped particle being detrapped and S_{ext} is the exterior source of particles entering the volume. $D(T) = D_0 \cdot e^{-\frac{E_D}{k \cdot T}}$ is the diffusion coefficient of HIs in the metal matrix (in m^2 s^{-1}) with the energy barrier E_D represented in Fig. 1 (k is the Boltzmann constant). Dealing with HIs, the diffusion coefficient has to be mass dependent, thus as in Refs. [10,11] we used $D_{HI} = \frac{D_H}{\sqrt{HI\ atomic\ mass}}$ for the diffusion of any Hydrogen Isotope. As a

remark, if an equilibrium is considered between trapped and solute particle, the model correspond to Oriani's one [5] which envisaged the HIs trapping and migration as an apparent diffusion from trap to trap: in such model, the retention variation with fluence would vary as fluence$^{0.5}$ (i.e. limited by diffusion).

For each trap type, there are a finite number of available traps. We also assume that each trap captures only one HI atom. The trap density is noted n_i and it can evolve with space (inhomogeneous spatial distribution) and with time, since we include trap creation in this model (see further in the text) in contrast to some previous MRE models [7,10,11]. The number of solute sites is fixed. We call n_{solute} the number of solute sites per tungsten atoms and it is considered that $n_{solute} \gg \sum n_i$. Therefore we consider that for each trap, its first neighbor is a solute site. It is also considered that the solute concentration is low: $C_m \ll n_{solute}$ which means that for each trapped particle, there is a free neighbor solute site which permits detrapping. Following these assumptions, $S_{mobile \to trap,i}$ and $S_{trap,i \to mobile}$ can be expressed as follows [8]:

$$S_{trap,i \to mobile} = \frac{1}{\tau_a} \cdot C_{t,i} \tag{4}$$

$$S_{mobile \to trap,i} = \frac{1}{\tau_b} \cdot \frac{C_m}{n_{solute}} \cdot (n_i - C_{t,i}) \tag{5}$$

Here, τ_a and τ_b are time constants for respectively the detrapping and trapping processes. As in Refs. [7,10,11], the detrapping time constant τ_a can be expressed as a frequency term. Using the energy barrier $E_{T,i} = E_D + E_{B,i}$ of Fig. 1, $\frac{1}{\tau_a} = \nu_0 \cdot e^{-\frac{E_{T,i}}{k \cdot T}}$ and Equation (4) becomes:

$$S_{trap,i \to mobile} = \nu_0 \cdot C_{t,i} \cdot e^{-\frac{E_{T,i}}{k \cdot T}} \tag{6}$$

where ν_0 is the pre-exponential factor (or attempt frequency) in s^{-1}. It is important to specify the value of ν_0 when this set of equations is used to fit TDS experiments. Indeed, for a same detrapping energy, the simulated peak can be shifted by about 35 K if ν_0 changes only by an order of magnitude. In the following, ν_0 is taken equal to 10^{13} s^{-1} accordingly with [7,10]. In Refs. [8,9], ν_0 is expressed as a function of the lattice constant and of the diffusion coefficient and $\nu_0 \sim 3 \times 10^{13}$ s^{-1} for hydrogen and $\nu_0 \sim 2 \times 10^{13}$ s^{-1} for deuterium which is of the same order of magnitude. The trapping source can also be understood as a detrapping effect, where the HI atom detraps from a solute site before falling in a trap site of type i. The trapping attempt frequency $1/\tau_b$ is usually expressed as a function of the diffusion coefficient by $\tau_b = \lambda^2 / D(T)$, where λ is the distance between 2 solute sites or between a solute and a trap site. So the trapping source term (5) becomes:

$$S_{mobile \to trap,i} = \frac{D(T) \cdot n_i}{\lambda^2 \cdot n_{solute}} \cdot C_m \cdot \left(1 - \frac{C_{t,i}}{n_i}\right) \tag{7}$$
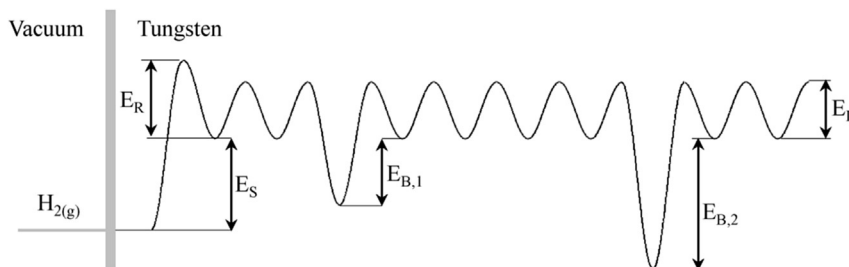

Fig. 1. Potential energy diagram for a hydrogen atom in tungsten.