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Deuterium retention in dense and disordered nanostructured tungsten coatings

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ABSTRACT

A systematic and attentive comparison of the deuterium (D) retention in tungsten (W) coatings with different nano-crystalline structures after the plasma exposure in comparison with polycrystalline tungsten (PCW) is presented. While a wide database is available for PCW, only a few data about the D retention in coatings with different structures exist. The D retention in W coatings produced by three different deposition techniques on different substrates was studied with respect to the influence of (a) coating crystallite size, (b) coating thickness, (c) specimen temperature during D plasma exposure, (d) presence of argon (Ar) used as working gas during the coating deposition and (e) substrate material. It is shown that the variation of the processing parameters, such as temperature, deposition rate, Ar implantation, etc. even within one deposition method results in different grain size distributions and structure of coating and has a significant effect on the D retention. It is revealed that the substrate material and the presence of Ar in a coating play a minor role in the D retention in the coating. It is shown that both the D concentration and the D retention in coatings drastically increase with decreasing the grain size. Consequently, in the case of using of W coating as a protective layer of a structural material, a compromise in the development of nanostructured tungsten films is necessary to keep the hydrogen isotope concentration at an acceptable level.

1. Introduction

A prediction of fuel retention in plasma-facing materials (PFMs) for ITER and future fusion reactors depends on the understanding of the fundamental hydrogen isotope retention mechanisms in these materials. Tungsten (W) is a promising material because of the high melting point, low sputtering rate and low hydrogen solubility. A lot of effort has been devoted to investigating deuterium (D) retention in undamaged [1–7] and radiation-damaged [8–13] polycrystalline tungsten (PCW). Those studies confirmed that the D retention in undamaged PCW should be smaller compared to the D retention expected for other candidates, e.g. carbon and beryllium, which is a positive aspect with respect to the control of in-vessel tritium inventory [14–17]. However, the D retention at radiation-induced defects in PCW can be a concern [11–13]. Moreover, the D retention in W coatings used as plasma-facing materials for today's tokamaks such as ASDEX Upgrade with the full-W wall and JET with the ITER-like wall (project JET-ILW) is known to differ from that of bulk tungsten [18–22]. This is attributed to the different micro-structure and defect density. During the last decade, the research in the field of W coatings for fusion applications was mainly focused on the thermo-mechanical properties, particularly, on their resistance to high heat fluxes ($T_{max} \leq 2.000\,^{\circ}C$) [23,24]. Dense nanostructured W coatings produced by Combined Magnetron Sputtering and Ion Implantation (CMSII) technology was chosen as a most promising coating for plasma-facing material application after successful high heat flux testing up to 10 MW/m^2 in GLADIS facility [25]. But the information concerning the D retention for this type of coating is quite limited [18,21,22]. In [21],

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the D retention in CMSII-W coatings on isotropic fine grain graphite (FGG) substrate was investigated for various incident ion energies ranging from 20 to 200 eV per deuterium atom with fluences ranging from $1 \times 10^{23} m^{-2}$ to $2 \times 10^{25} m^{-2}$. It was shown that the D accumulation in CMSII-W coatings is higher compared to bulk PCW in all range of investigated temperatures from 320 to 650 K. The D retention in nano-structured CMSII-W coating with a thickness of 7–10 μm was higher compared to other types of coatings, such as physical vapour deposited W coating with a thickness of 4–5 μm and plasma-sprayed W coating of 200 μm for all investigated D irradiation conditions. It was suggested that the D retention correlates with the microstructure of W coatings. The high initial density of intrinsic defects in CMSII-W coating results in high D retention. The data of the D retention in CMSII-W coatings after exposure in ASDEX Upgrade reported recently in [18] are in a very good agreement with laboratory data reported in [21].

In future fusion power plants, PFMs will operate under a severe environment involving high heat flux, intensive irradiation and particle implantation (x-ray, gamma, neutrons, ions, neutrals and electrons). 14 MeV neutrons generate radiation defects in the bulk of PFMs. Retention at radiation-induced defects in PCW can be a concern [11–13]. The effect of radiation damage on the D retention in nanostructured W coatings was investigated only in two papers [22,26]. It was reported in [22,26] that the D concentration at radiation-induced defects produced by self-ion irradiation to a damage level of 3 displacements per atom (dpa) is the same for dense CMSII-W coatings and disordered coatings produced by Pulsed Laser Deposition (PLD) method and it is the same as in polycrystalline PCW. In other words, this implies that the density of radiation-induced defects is the same for all types of W coatings, regardless of the structure of each W material. On the other hand, if we define the radiation tolerance as the ratio of the density of initial intrinsic defects in un-irradiated material to the density of radiation-induced defects then the radiation tolerance increases with decreasing the grain size. In the paper [22], it was shown that CMSII-W coating exhibits enhanced radiation tolerance compared to PCW, namely, neutron irradiation will produce smaller change in coating properties compared to PCW. In the paper [26], it was shown that radiation-induced defects created by self-ion irradiation to a damage level of 3 dpa at room temperature do not affect the D concentration in PLD-W films with crystallite size below 7 nm. This implies that the formation of radiation-induced defects is suppressed in nanostructured W films with crystallite size below 7 nm because pre-existing nanoscaled boundaries behave as effective sinks for point defects, thus, enhancing defect annihilation and promote Frenkel pair recombination. This is in agreement with results reported in [27–29] which demonstrated that materials based on nanoscale interfaces have improved radiation resistance.

It was also mentioned in [26] that despite the fact that nano-structured W films with small crystallite size possess a potential for improved radiation resistance compared to polycrystalline tungsten, the nano-structured W coatings have a high retention of hydrogen isotopes in comparison with PCW. Moreover, the build-up of D at the interface between coating and substrate can be a concern for both un-irradiated and neutron-irradiated materials [26] because it can cause hydrogen-induced cracking. However, PCW as a pure material has not been used so far for the entire first wall in fusion devices because of high cost implications to the project, its brittleness and bulkiness resulting in large structural weight. Therefore, tungsten coatings on a lighter substrate was found to be a good solution to investigate specific aspects concerning the compatibility of fusion plasma with a W wall. At the same time, the deposition of thick W coating on the structural material may be only one possible option to protect the structural material from interaction with the plasma.

The objective of the present work is to study in details the D retention in nano-structured W coatings exposed to D plasma in well-defined laboratory conditions in order to understand the fundamental retention mechanisms in those materials for fusion application. In contrast to tokamak devices, it is possible to control experimental parameters in plasma or ion beam irradiation experiments, for example, ion flux, fluence, ion energy and temperature of W specimen. By varying the deposition parameters, the controlled W film structure can be obtained even within the same deposition method. We will investigate dependences of the D retention in W coatings on (i) the substrate material, (ii) the nano-crystalline structure, (iii) the thickness of the coating and (iv) the presence of impurities in the coating. The D retention at the interface between the coating and the substrate is also studied.

2. Experimental

2.1. Specimens

W coatings used in the present study were prepared by:

1) combined magnetron-sputtering and ion implantation technique (CMSII-W) on FGG, CFC and Eurofer substrates (NILPRP Bucharest),
2) standard vacuum magnetron-sputtering (SMS-W) on FGG and Eurofer substrates (IPP, Garching),
3) pulsed laser deposition (PLD) method on FGG and Eurofer substrates (Politecnico di Milano).

FGG is isotropic fine grain graphite, CFC is carbon-fibre composite and Eurofer is reduced-activation ferritic/martensitic steel. Those C-based materials have been widely used as PFMs in various tokamaks in the last decades, and Eurofer steel is a candidate material for the structure material of future fusion reactors. The thickness of substrates was 1 mm.

The deposition of CMSII-W coatings was done at National Institute for Laser, Plasma and Radiation Physics, Bucharest [23,24] in argon (Ar) atmosphere using a standard two axes rotation device with the rotation speed of 2 rot/min. Typical parameters for the high voltage pulse discharge are: $U = 40$ kV; $\tau \approx 20$ μs and $f = 25$ Hz, $U_{bias} = -700$ V for sputter cleaning between pulses and −100 V for deposition, the argon flow rate is 25 sccm and the deposition pressure is 0.8 Pa. The temperature of the substrate during the deposition was about 300 °C. Such deposition method provides a nano-structure and high densification of the coating. The deposition of CMSII-W film on CFC substrates has been done for two fibre planes: pitch bundles oriented perpendicular and PAN bundles oriented parallel to the coated surface. The Ar content in the coating was detected by Rutherford backscattering spectrometry (RBS) to be ~1% [26].

Some coatings were produced by runs with the oscillating jigging device. The difference in the coating production between the two systems is the following:

- With the oscillating jigging device, substrates remain all the time in front of the magnetrons. The deposition process is continuous. The deposition rate for W was about 1.4 nm/s.
- With the two axes rotation device (standard run), substrates rotate in front of the magnetrons. In this way they stay in front of the magnetron only half of the processing time. The other half of time the substrates do not see the magnetron targets. The deposition rate for W was about 0.7 nm/s. The deposition process is not continuous. In this case, the coating has bigger crystallite size.