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Short communication

Surface hardening induced by high flux plasma in tungsten revealed by nano-indentation

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ABSTRACT

Surface hardness of tungsten after high flux deuterium plasma exposure has been characterized by nanoindentation. The effect of plasma exposure was rationalized on the basis of available theoretical models. Resistance to plastic penetration is enhanced within the 100 nm sub-surface region, attributed to the pinning of geometrically necessary dislocations on nanometric deuterium cavities – signature of plasma-induced defects and deuterium retention. Sub-surface extension of thereby registered plasma-induced damage is in excellent agreement with the results of alternative measurements. The study demonstrates suitability of nano-indentation to probe the impact of deposition of plasma-induced defects in tungsten on near surface plasticity under ITER-relevant plasma exposure conditions.

1. Introduction

Mechanical and microstructural properties of sub-surface region play a crucial role in the performance of materials for high temperature applications, operating as armour in extreme environment, such as in the fusion reactor [1]. Extracting heat from 150 million °C plasma coming in cycles represents a major challenge for modern worldwide technology to select and qualify an appropriate material. Tungsten (W) is one of several plasma facing materials considered for the operation in ITER and DEMO [2,3] and therefore investigation of its performance under high flux plasma is one of the "hottest" issues for the applied solid state – plasma physics community, in its direct and indirect meaning.

The emergence of nano-scale experimental techniques and of physically robust computational models has promoted a growing interest for the investigation of W sub-surface region at nano- and atomic-scales (see e.g. Refs. [4,5]). From the view point of W-plasma interaction, the outer surface will experience cyclic heat loads, leading to the nucleation of cracks and to their propagation towards bulk [6]. Permeation of plasma particle (hydrogen isotopes - HI) forming nano-cavities (presumably hydrogen bubbles) [7] is expected to bring an extra hardening by constraining the dislocation-mediated plasticity and therefore preventing the release of the thermo-mechanical stresses. The mechanical failure is unacceptable for the proper functioning of the armour material, as it leads to the extra heat propagation on the one hand, and creates a risk of plasma contamination, on the other hand.

Further efforts are therefore required to understand the sub-surface W hardness and its evolution under plasma and heat exposures. Unfortunately, conventional mechanical testing (uniaxial tensile or Vickers hardness punch tests) are not applicable because the typical penetration depth of HI under high flux plasma exposure is limited to ~10 μm [8,9] i.e. major amount of HI is stored within the first grain facing the surface. Nano-indentation (NI), with a resolution of tens of nanometers, is therefore an ideal tool to deal with the problem. Armstrong et al. [10–12] have already demonstrated that NI can be successfully applied to W and W-rhenium alloy to quantify ion-induced damage and the impact on the resistance to plastic deformation. A good correlation between the ion damage depth profile and hardness variation was found. Here, we continue the investigation of the surface hardness of W and focus on the high flux plasma-induced damage. We investigate

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the sub-surface hardness of W samples exposed to ITER-relevant deuterium (D) plasma at different temperatures.

The investigated samples were cut from a double forged bar of tungsten of 99.99 purity. The original bulk material has been hot rolled into a cylinder at 1600 C and then forged (i.e. compressed by several precents) in radial direction and then along the cylinder axis to maximize the density. The main impurities, as reported by the manufacturer, are listed in Table 1 provided as supplementary material. The production route, initial microstructure and nominal mechanical properties are reported in Refs. [13,14]. The forging was followed by stress relief at 1000 °C and annealing at 1600 °C for 1 h in inert environment to protect from oxidation. The annealing uniformed the shape of grains and reduced the dislocation density down to about $(2-8) \times 10^{12}$ m^{-2}, as measured by transmission electron microscopy (TEM). Scanning electron microscopy (SEM) and electron back scattering diffraction (EBSD) revealed that a typical size of random grains is in the range 50–150 μm, and sub-grains of size 2.5–5 μm are present. The samples for plasma exposure and subsequent NI testing were cut to $10 \times 10 \times 1$ mm dimension, re-annealed and mechanically polished down to 0.25 μm to reach mirror surface finish.

Exposures were performed at the linear plasma generator Pilot-PSI [15,16], which delivers high-density deuterium (D) plasmas mimicking the 'sub-displacement threshold' plasma wall interaction conditions expected in the ITER. The energy of D ions was ~50 eV, while imposing more than 900 eV is required to initiate atomic displacement in W. Although the plasma beam was non-uniform (full width at half maximum, FWHM, ~10 mm), the relatively small sample surface ensured a limited temperature gradient (473 ± 10 K) across the surface during exposure, as measured and confirmed by an infra-red camera (FLIR A645 sc). The maximum particle flux in the centre of the sample was 10^{24} $D/m^2/s$. The flux was calculated from the plasma electron density and electron temperature, as measured by Thomson scattering [17]. The samples were exposed to a fluence of $F = 5 \times 10^{25}$ D/m^2.

The exposed and reference samples were tested using an Agilent G200 nanoindenter in order to determine the Young's modulus and hardness. The indentation measurements were performed in the continuous stiffness mode (CSM) [18] with the standard XP head equipped with a Berkovich diamond tip. The oscillation amplitude and frequency were respectively 2 nm and 45 Hz. The indentation strain rate was set to 0.05 s^{-1} and the specimens were indented down to a penetration depth $h_{NI} = 1.5$ μm. At least 25 indents spaced by 60 μm have been performed on each specimen, and the hardness estimated using the classical Oliver & Pharr method [18]. The tip area function was calibrated by performing a series of indents in a reference fused silica sample.

Nano-indentation was performed on three samples in different areas preselected by targeting in the optical microscope to avoid apparent surface defects. Several randomly chosen load-displacement curves for each probed sample are provided in the supplementary material (see Fig. S1). Limited variation in the load-displacement curves for the individual indents prove the overall homogeneity of the material and acceptable quality of surface even after high temperature exposure. Deviations in the loading curves for a given sample should be attributed to a particular grain orientation and size, in addition to the roughness induced uncertainty on the indentation depth. Let us here discuss the averaged results without taking into account particular properties of individual indented grains, although it might be important when considering the plasma-exposed samples as will follow from the discussion.

The effect of plasma-induced damage on the surface hardness was found to be very pronounced within $h_{NI} = 100$ nm. The absolute value of the Young's modulus E and of the hardness H measured by NI is subject to an error coming from the uncertainty of the contact surface [18,20,21], which however decreases with increasing penetration depth. To avoid the uncertainty related to nano-metric roughness and pile-ups formation obscuring the estimation of the actual contact area, only the ratio of the hardness-to-reduced modulus, H/E_r^2, will be presented as proposed in Refs. [21,22], and grounded by discussion. The roughening amplified by the plasma exposure is unavoidable. Any polishing to get rid of this roughness would modify the near surface state of hardening over thicknesses comparable to the region of interest for this study. The absolute value of the measured hardness is reported in supplementary material (see Fig. S2 and its capture for brief description).

The variation of H/E_r^2 as a function of penetration depth is shown in Fig. 1, with a decreasing trend typical of the plastic indentation response at shallow depth, accommodated mainly by geometrically necessary dislocations (GNDs) [23]. H/E_r^2 is a measure of the material resistance to plastic penetration, being independent on the contact area [21]. Physically, at very small indentation depth, H/E_r^2 characterizes the resistance of material to the injection of GNDs and their propagation towards bulk. The obtained results demonstrate that resistance to plastic flow strongly deviates in the sub-surface region of the plasma-exposed samples within first 80–100 nm, and beyond all the curves converge to the same value over the entire indentation depth. Non-negligible deviation in H/E_r^2 obtained for the 460 K- and 820 K-exposed samples is tentatively attributed to the difference in the size and density of plasma-induced defects formed (mainly cavities, believed to be deuterium bubbles), as their nucleation and growth will be affected by exposure temperature (as confirmed by the thermal desorption spectroscopy measurements presented elsewhere).

At this stage, we assume that the Young's modulus is not significantly affected by the exposure and by the presence of the cavities. Hence, the observed effect in Fig. 1(a) can be mainly attributed to the change in the resistance to plastic flow as quantified by H.

The apparent increase of H/E_r^2 with plasma exposure is to be attributed to the strengthening originating from the interaction of GNDs with two features, namely: (i) a higher density of statistically stored dislocation networks (SSD), and (ii) cavities (presumably, deuterium bubbles) and dislocation loops present in the subsurface up to a depth of several micrometers, as revealed by TEM examination partially reported in Refs. [7], TEM micrographs showing cavities and loops in the sub-surface of the exposed samples are added in the supplementary material.

A conventional way to measure the sub-surface deposition of D is the nuclear reaction analysis (NRA) method, whose results are presented in Fig. 1(b) for the same W sample measured after the exposure in equivalent high flux plasma conditions, and reported by us earlier [19]. The relative change in hardness is also added in the figure, being plotted versus the indentation depth which is scaled by a factor of five to reflect the size of the zone that undergoes plastic deformation [24]. Clearly, the deuterium deposition, measured by NRA, nicely correlates with the plasma-induced hardness increase (which is larger than the penetration depth by a factor of ~5).

According to Nix and Gao [23], a dependence of the form of $\frac{H}{H0} = \sqrt{1 + h^*/h}$ prevails up to a certain indentation depth h^*, which sets the magnitude of the depths over which GNDs activity dominates SSDs. H_0 is the "bulk" hardness, taken here at a depth of 1.5 μm, and h^* can be found by linear interpolation of $(H/H_0)^2$. Consideration of the reduced hardness (H/H_0) is a phenomenological way to estimate the space-scale of the microstructure providing the resistance to the penetration of GNDs (as h^* characterizes the depths