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Lattice swelling and modulus change in a helium-implanted tungsten alloy: X-ray micro-diffraction, surface acoustic wave measurements, and multiscale modelling

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Abstract—Using X-ray micro-diffraction and surface acoustic wave spectroscopy, we measure lattice swelling and elastic modulus changes in a W-1% Re alloy after implantation with 3110 appm of helium. An observed lattice expansion of a fraction of a per cent gives rise to an order of magnitude larger reduction in the surface acoustic wave velocity. A multiscale model, combining elasticity and density functional theory, is applied to the interpretation of observations. The measured lattice swelling is consistent with the relaxation volume of self-interstitial and helium-filled vacancy defects that dominate the helium-implanted material microstructure. Larger scale atomistic simulations using an empirical potential confirm the findings of the elasticity and density functional theory model for swelling. The reduction of surface acoustic wave velocity predicted by density functional theory calculations agrees remarkably well with experimental observations.

Keywords: Helium implantation; Micro-diffraction; Elastic properties; Density functional theory; Empirical potential

1. Introduction

Tungsten (W) and tungsten-based alloys are the main candidate materials for plasma facing divertor surfaces in future fusion power plants [1] due to their high melting point, good resistance to sputtering, high thermal conductivity, and low tritium retention rate [2–5]. In addition to the radiation damage produced by collision cascades [6], bombardment of tungsten with 14.1 MeV fusion neutrons also transmutes it into other chemical elements through nuclear reactions. Calculations show that significant amounts of rhenium (Re) (∼0.2 at.% year^{-1}), tantalum (∼0.1 at.% year^{-1}) and osmium (∼0.1 at.% year^{-1}) will be produced during the operation of a DEMO fusion reactor [7]. The amount of helium (He) produced by transmutation in tungsten is relatively small (between 0.1 and 10 appm year^{-1}) [7]. During operation tungsten surfaces in the divertor will also be exposed to large fluxes of hydrogen isotopes and helium ions with a broad spectrum of energies, resulting in a high heat flux of up to 15 MW/m^2 [8].

High-flux low-energy helium ion implantation causes significant modifications of the material surface (e.g. formation of sponge-like structures, "fuzz" and bubbles) even at ion energies below the sputtering threshold (∼100 eV) [9–11]. Similar effects have been observed at intermediate helium ion energies (30 keV) and temperatures from 1000 K to 1400 K [12,13]. At high temperatures it is expected that the implanted helium atoms diffuse deeper into the bulk, affecting material behaviour due to their strong interaction with irradiation defects [14]. For example, it has been found experimentally, by nano-indentation, that the combined effect of helium implantation and cascade damage from self-ion irradiation on hardness is far greater than that of cascade damage alone [15].

An important question concerns the dominant mechanism by which helium is retained in the tungsten matrix. Positron Annihilation Spectroscopy (PAS) studies [16,17] and ab-initio calculations [18] indicate that helium-induced microstructure in metals is driven by the propensity of helium atoms to form bound complexes with vacancies. These may be pre-existing vacancies or vacancies formed

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by helium agglomeration and self-trapping leading to the spontaneous production of Frenkel pairs [19,20]. Helium also has a significant effect on radiation-induced microstructure through the suppression of vacancy and self-interstitial atom (SIA) recombination since helium atoms rapidly fill vacant lattice sites [21]. At elevated temperatures the resulting availability of excess SIAs stimulates nucleation and growth of interstitial dislocation loops [22].

Due to these complex interactions, carrying out quantitative analysis of the microstructure formed as a result of helium ion implantation proves challenging [23]. The majority of recoil events produced by helium ions during implantation have energy lower than ~100 eV (see Fig. 1(c)). Such low energy recoil events produce only individual Frenkel pairs, and hence the microstructure of helium ion irradiated tungsten is expected to be dominated by helium-filled vacancies and vacancy clusters, and SIA defects. The objective of the analysis given below is to correlate experimentally measured strains and elastic properties with the notion of such a damage microstructure through the use of data on defect properties derived from *ab initio* calculations, elasticity theory and larger scale atomistic calculations with an empirical potential.

2. Experimental measurements

2.1. Sample preparation

To mimic transmutation-induced production of rhenium in tungsten, a W-1 at.% Re alloy was manufactured by arc melting from high purity elemental powders [24]. 1 mm thick slices were polished using diamond paste and 50 nm colloidal silica suspension to produce a flat, damage-free surface. Optical micrographs show equiaxed grains with sizes ranging from 100 to 1000 μm (Fig. 1(a)). Electron back scattered diffraction (EBSD) indicated no significant texture.

Helium ions were implanted at 300 °C to a depth of ~2.8 μm using a 2 MeV ion accelerator at the National Ion Beam Centre, University of Surrey, UK. To achieve a near uniform helium ion concentration in excess of 3000 appm throughout the implanted layer, implantations were carried out at 12 different ion-energies and fluences [24]. The resulting implantation profile, predicted by the Stopping and Range of Ions in Matter (SRIM) code [25], assuming a Frenkel pair formation threshold energy of 68 eV [26], is shown in Fig. 1(b). At depths between 1 and 2 μm the calculated implanted helium dose is 3110 ± 270 appm. The associated displacement damage is 0.24 ± 0.02 displacements per atom (dpa). Analysis of recoils caused by the implanted helium ions (Fig. 1(c)) shows that they are predominantly low energy events. Hence we expect mainly the formation of individual Frenkel pairs during implantation, rather than clusters of defects [27]. This is also confirmed by transmission electron microscopy (TEM) of pure tungsten samples implanted with helium under the same conditions, which showed no visible defects after implantation, indicating that all of the implantation-induced defects formed at 300 °C are below the TEM resolution limit [15].

2.2. Micro-diffraction measurements

Lattice swelling due to helium implantation was measured by micro-beam Laue diffraction at beamline 34-ID-E at the Advanced Photon Source, Argonne National Lab, USA. Fig. 2(a) shows a schematic of the experimental setup. The incident, polychromatic (7–30 keV) X-ray beam was focussed by Kirkpatrick-Baez (KB) mirrors to a probe spot with 600 nm vertical and 400 nm horizontal full width at half maximum and near Lorentzian shape. The sample was positioned at the focus in 45° reflection geometry. Diffraction patterns were recorded on a Perkin Elmer flat panel detector mounted in 90° reflection geometry above the sample. The Differential Aperture X-ray Microscopy (DAXM) technique was used to determine the depth in the sample from which different scattered contributions originated [28–30]. For DAXM measurements a 50 μm diameter platinum wire was scanned through the diffracted beams. By triangulating using the wire edge, depth-resolved Laue patterns were reconstructed at 500 nm intervals along the incident beam direction. A detailed description of the experimental setup and analysis routines is provided elsewhere [29,31,32].

Laue diffraction patterns containing more than 20 peaks were collected at 3 positions in the implanted sample. The

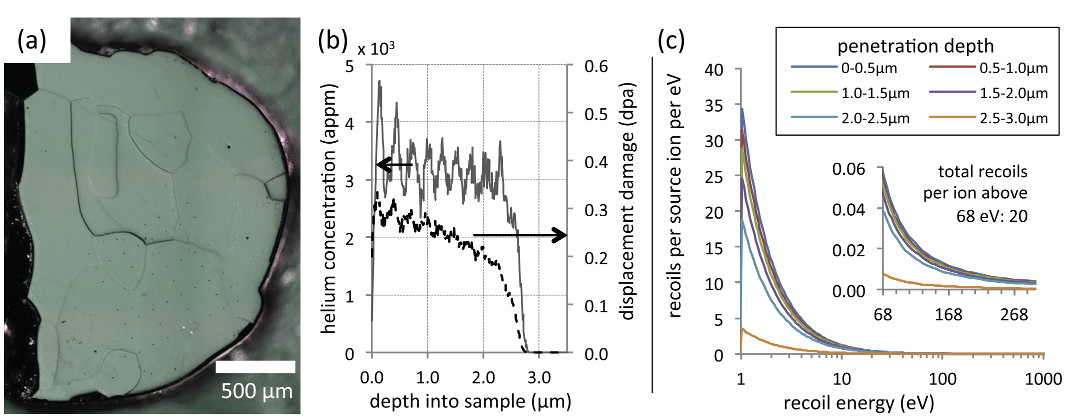

Fig. 1. (a) Representative optical micrograph of the W-1% Re material. (b) SRIM calculated profile of injected helium ion concentration (grey curve) and implantation-induced displacement damage (black curve) as a function of depth in the sample. (c) Primary recoil atom energy spectrum per source ion calculated for different depths in the sample. On average each source ion produces 20 recoils with energy greater than the Frenkel pair formation threshold energy of 68 eV.