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High temperature annealing of ion irradiated tungsten

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Abstract—Transmission electron microscopy of high temperature annealing of pure tungsten irradiated by self-ions was conducted to elucidate microstructural and defect evolution in temperature ranges relevant to fusion reactor applications (500–1200 °C). Bulk isochronal and isothermal annealing of ion irradiated pure tungsten (2 MeV W^+ ions, 500 °C, 10^{14} W^+/cm^2) with temperatures of 800, 950, 1100 and 1400 °C, from 0.5 to 8 h, was followed by *ex situ* characterisation of defect size, number density, Burgers vector and nature. Loops with diameters larger than 2–3 nm were considered for detailed analysis, among which all loops had $\boldsymbol{b} = \frac{1}{2}\langle 1\,1\,1\rangle$ and were predominantly of interstitial nature. *In situ* annealing experiments from 300 up to 1200 °C were also carried out, including dynamic temperature ramp-ups. These confirmed an acceleration of loop loss above 900 °C. At different temperatures within this range, dislocations exhibited behaviour such as initial isolated loop hopping followed by large-scale rearrangements into loop chains, coalescence and finally line–loop interactions and widespread absorption by free-surfaces at increasing temperatures. An activation energy for the annealing of dislocation length was derived, finding $E_a = 1.34 \pm 0.2$ eV for the 700–1100 °C range.

Keywords: Tungsten; Ion irradiation; Annealing; TEM; Fusion

1. Introduction

Tungsten is seen as the primary candidate material for plasma-facing components (PFCs) in future fusion reactors. Pure tungsten has been chosen as ITER's PFC material [1], and substantial research is being undertaken internationally to improve its performance (through alloying, tailoring microstructure [2], and compositing), and to understand its fundamental mechanical behaviour [3]. A major issue with tungsten is its acute brittleness and high brittle-to-ductile transition temperature (BDTT) of ~400–500 °C [4–6], rendered more severe by radiation damage, which immediately creates dislocation loops and other defect structures (SIAs, voids), and in the longer term can produce brittle phases as a result of elemental transmutation, both of which are detrimental to tungsten's mechanical performance [7].

This paper attempts to address questions arising when tungsten is irradiated in high temperature environments found in fusion reactors. What is the behaviour of radiation damage at high temperatures in tungsten? What evolution pathway do loops found in typical irradiated samples undertake, and can dislocations be annealed away completely, and at what temperature? To this aim, in order to understand the recovery behaviour of displacement damage, transmission electron microscopy (TEM) analysis, used successfully on other materials [8,9], was performed on ultra-high purity (UHP) tungsten samples following 2 MeV W^+ ion irradiation (as a surrogate for neutron irradiation) and subsequent vacuum annealing. The annealing temperatures ranged from 800 °C to 1400 °C, for times ranging from 1 to 8 h.

Finally, *in situ* annealing experiments were carried out, both isochronal tests and dynamic ramp-up tests, detailed in subsequent sections. These were useful to observe dislocation behaviour during annealing and to understand the underlying mechanisms associated with damage evolution, as well as the transition between recovery stages and the role which free surfaces play.

1.1. Previous recovery studies

Several studies have probed the effect of temperature *during* irradiation in tungsten, using ion irradiation [10] and

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neutrons [11]. Numerous studies have also been undertaken with regard to the recovery of irradiation-induced defects post-irradiation. However, these have mostly centred around resistivity measurements [12–19], with some early field ion microscopy work [20,21] and a single early TEM study on single crystal tungsten [22]. A similar TEM work has also been done in molybdenum [23]. Resistivity measurements, although useful in identifying the temperature and activation energies of recovery stages (calculated using the Meechan–Brinkman theory [24]), do not provide a direct method of the observation of defects, their nature and how they change with different annealing conditions. This study looks to provide a more complete picture by: (1) quantifying the damage immediately after irradiation, (2) understanding the time and temperature dependence of recovery, (3) directly observing the mechanisms associated with the radiation defect recovery. Such a study has found to be lacking in the literature, with systematic experiments being limited to loops and stacking fault tetrahedra formed during quenching processes of fcc metals [8].

Studies based on fast neutron-irradiated specimens ($\geqslant$ 1 MeV), have identified five stages of annealing common to BCC metals based on the classifications of Thompson [25]. There seem to be numerous inconsistencies in the literature on the number of recovery stages, their nomenclature, temperature and physical interpretation, particularly at higher temperatures [26].

Stage I occurs below −170 °C and is attributed to the movement of free interstitials. Stage II, between −170 °C and 350 °C, is a steady recovery attributed to the release of interstitials from traps with a wide range of interaction energies from 0.25 to 1.7 eV [27]. This explains the many sub-stages observed for example in resistivity studies [17]. These two stages occur below fusion reactor operating temperatures, and will not be considered further here. For fusion-relevant temperatures (from 500 to 1000 °C [28]), the recovery of defects created by fast neutrons in tungsten has been observed to occur in three major stages (III–V), at $\sim$0.15 T_m/350 °C, $\sim$0.22 T_m/640 °C, $\sim$0.31 T_m/970 °C [16].

Stage III, with an onset of $\sim$350 °C and activation energy of $\sim$1.7 eV, was initially considered to be due to self-interstitial migration, primarily to immobile traps [18]. This explanation was later revised to being due to monovacancy mobility activation [29–31,27]; this interpretation is also supported by DFT calculations [32]. The onset temperature has also been shown to shift to lower temperatures with increasing neutron fluence [13].

Higher temperature recovery stages in tungsten have been largely un-researched since the 1970s, with interpretation being limited to resistivity measurements and early FIM experiments [17,33,20].

Stage IV recovery, at 0.22T_m, is still subject to debate [14], being generally attributed to vacancy-impurity complexes [33], or di-vacancies [17]. Recent DFT simulations have shown that di-vacancies are not favoured in tungsten [34]. Other *ab initio* computational studies yield activation energies between 3.00 and 3.43 eV for vacancy-carbon complexes, that may explain this stage as being controlled by the dissolution of vacancy-carbon complexes [35], noting that the experimental activation energy calculated from resistivity measurements is 3.3 eV [15]. Similar conclusions for this stage may be drawn by comparing to other materials, such as α-Fe, where the recovery stage above Stage III observed by simulations and positron lifetime measurements was ascribed to VC_n complexes [36–38].

Stage V recovery, starting at 0.31T_m,[1] was initially explained as being due to vacancy migration, based on early field-ion microscopy studies [20,21], but later attributed to the disappearance of "defect clusters" or formation of voids [19]. It is still unclear what is the cause of this recovery stage.

The majority of studies have been carried out on polycrystalline tungsten. In single-crystal tungsten,[2] defects were found to be stable up to 1900 °C, [17] presumably due to the absence of sinks at grain boundaries. Bykov et al. [17] identified three stages of annealing: 500–800 ° (Stage IV), 950–1200 ° (Stage V), and 1200–1900 ° (Stage VI).

1.2. Irradiation-induced defects

Ion irradiation introduces several types of defects in the material such as self-interstitial atoms (SIAs), vacancy clusters, dislocation loops, voids. Damage formation specific to W^+-ion irradiated UHP tungsten (and alloys) has been characterised [39,40] and shows the presence of both $\frac{1}{2}\langle 111\rangle$ and $\langle 100\rangle$-type Burgers vector loops, as suggested by the Eyre–Bullough mechanism, with a decreasing fraction of the latter with increasing irradiation temperature (from $\sim$79% $\frac{1}{2}\langle 111\rangle$ loop fraction at 300 °C to $\sim$90% at 750 °C, at 1.5 dpa).

2. Experimental procedure

2.1. Sample preparation

Polycrystalline ultra-high purity tungsten (typically W >99.996 wt%) was sourced from Plansee Gruppe, Austria, in the form of 150 μm foils. These contained traces of C (10 ppm), P (<10 ppm), Si and O (5 ppm) impurities. 3 mm diameter TEM discs were punched from the foils, and mechanically polished to a thickness of 100 μm using diamond lapping films (grit size 15, 9, 6, 3 and 1 μm). The discs were then annealed in a vacuum furnace for 24 h at 1400 °C to promote grain growth and to ensure a dislocation-free microstructure. The vacuum furnace used a diffusion pump, and had a pressure below 10^{-6} mbar, to prevent surface contamination. Furthermore, subsequent to annealing, the discs were electropolished in a bath of 0.5 wt% NaOH aqueous solution at close to 0 °C to remove any potential oxides or contamination during annealing and ensure a mirror-like surface finish.

2.2. Ion irradiation

The polished bulk samples were irradiated at the National Ion Beam Facility at the University of Surrey, United Kingdom, using W^+ ions at 2 MeV, at a temperature of 500 °C to a fluence of 10^{14} ions/cm^2, producing an estimated dose of 1.5 dpa. The dose rate was 2.73×10^{10} ions/cm^2/s.

The dose was calculated using SRIM 2008, using a displacement threshold energy of 55.3 eV [41]. More details on the SRIM computations can be found in the Appendix.

[1] This stage is referred to as Stage IV in [15].
[2] No crystal orientation is given in this study [17].