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Small-scale characterisation of irradiated nuclear materials: Part I – Microstructure

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ABSTRACT

The behaviour of nanometre-scale precipitates in oxide dispersion strengthened (ODS) ferritic alloys and tungsten–rhenium alloys for nuclear applications has been examined by atom probe tomography (APT). Low Re content tungsten alloys showed no evidence of Re clustering following self-ion irradiation whereas the 25 at.% Re resulted in cluster formation. The size and composition of clusters varied depending on the material form during irradiation (pre-sharpened needle or bulk). These results highlight the care that must be taken in interpreting data from ion irradiated pre-sharpened needles due to the presence of free surfaces. Self-ion irradiation of the ODS ferritic alloy resulted in a change in the composition of the clusters, indicating a transition from a near-stoichiometric $Y_2Ti_2O_7$ composition towards a Ti_2YO_5.

1. Introduction

The next generation of nuclear reactors – both fission and fusion – will see the materials from which they are constructed being subjected to greater extremes than those currently in service; these extremes primarily being high temperatures and greater damage levels [1,2]. As such it is necessary to develop new materials that can withstand these extremes more than the materials that are currently in use today. Two highly prominent materials for advanced reactor designs are oxide dispersion strengthened ferritic alloys (including the nanostructured ferritic alloy class) and tungsten. Both of these materials can suffer from radiation-induced microstructural changes that are detrimental to the mechanical properties of the alloys.

1.1. Tungsten

Tungsten is the prime candidate material for the plasma facing components (PFCs) of future fusion devices such as ITER and DEMO [3–5]. During operations the deuterium–tritium reaction results in the production of 14.1 MeV energetic neutrons that may interact with the PFC material. This interaction can result in two types of material modification: lattice damage caused by the permanent displacement of atoms from their lattice sites, and transmutation reactions whereby the matrix atoms are chemically altered into another element. The lattice damage generated will take the form of fundamental defects e.g. interstitials and vacancies, and more complex defects such as dislocations, stacking faults and voids. Chemical transmutation occurs as a function of neutron energy and fluence [6]. It has been predicted that under fusion-like conditions the most common transmutation products from a pure tungsten component after 5 years of full-power operation will include W, Re, Os, Ta, Hf, He and H [4,6] with the most common transmutation products being Re and Os (5 and 3 at.% respectively after 5 years operation).

Pure tungsten is inherently brittle at low temperatures (~400–500 °C [7,8]) but through alloying with Re the brittle-to-ductile transition temperature can be reduced. Therefore the production of Re due to transmutation should be beneficial. However the displacing irradiation can result in radiation-enhanced clustering of the transmutation products [9]. The resulting clusters can be σ or χ phases (ReW and Re_3W respectively) that can facilitate embrittlement [9]. It is therefore necessary to understand the parameter space within which W–Re (and W–Re–Os) cluster formation occurs so that steps can be taken to mitigate the diminution of mechanical properties.

1.2. Oxide dispersion strengthened ferritic alloys

The class of steels known as oxide dispersion strengthened (ODS) ferritic alloys (also known as nanostructured ferritic alloys) consist of a dispersion of ultra-fine oxide particles throughout the matrix. These oxide particles serve to improve the mechanical properties of the system, particularly at high temperatures, of the

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system through inhibiting dislocation motion and grain boundary sliding. In nuclear applications the oxide particles have been suggested to act as point defect sinks [10,11] to improve radiation tolerance, and as preferential sites for the formation of nano-scale He bubbles therefore reducing swelling compared to non-ODS steels [12–15]. The ability of the oxide particles to improve these properties depends on the structure and composition of the particles [10,11,16,17] and their stability under irradiation. Typical compositions of ODS steels include between 9 and 14 at.% Cr for oxidation resistance (most commonly 14 at.%); W for solid solution hardening; Y_2O_3 that is put into solid solution during the initial, mechanical alloying, process but then during consolidation at high temperatures forms precipitates; and Ti to inhibit significant growth of the oxide particles; the balance being made up of Fe and impurities [18]. For this reason these steels are often referred to as 14YWT, reflecting the constituent elements.

Previous studies of the radiation behaviour of the particles under irradiation [19,20] showed that at temperatures of 100 °C and below, oxide particles can be dissolved back into the matrix whereas at higher temperatures, refinement and growth can be observed [21,22]. This clearly indicates that the radiation has some effect on the oxide particles, and that a more detailed understanding of this radiation response is required.

2. Experimental methods

2.1. Tungsten

The tungsten materials used in this study came in the form of wire or bulk specimens. The alloy wires were 0.25 mm in diameter with compositions of W–3 wt.%Re, W–5 wt.%Re and W–25 wt.%Re (W–3Re, W–5Re, and W–25Re respectively); purchased from Goodfellows and with a total impurity concentration below ~210 ppm. The wires were annealed at 1400 °C for 12 h to ensure a dislocation free structure prior to electropolishing in a 5 wt.% NaOH solution to form a needle for atom probe analysis.

Bulk samples of W–25Re were prepared by vacuum arc melting of tungsten (Sigma Aldrich) and rhenium (AEE) powders. The impurity levels of the material was determined by X-ray fluorescence to be below ~210 ppm [23]. The surface of the bulk material was mechanically polished to a mirror-like finish using colloidal silica as the final polishing medium.

Both bulk and needle-shaped specimens were irradiated with 2 MeV W ions to a fluence of 1×10^{14} ions cm^{-2} (equivalent to 1.5 displacements per atom, dpa) at temperatures of 300 and 500 °C using the Tandem accelerator at the University of Surrey Ion Beam Centre. During irradiation the needle-shaped specimens were orientated parallel to the direction of the ion beam i.e. the irradiation was head-on to the apex of the needle. The ion fluences were converted to damage levels in dpa using information obtained using the Stopping and Ranges of Ions in Matter (SRIM) simulation program [24] using the Kinchin–Pease model and a displacement energy, E_d, of 68 eV [25].

Following ion irradiation the samples were prepared from the bulk material for atom probe analysis using a focussed ion beam (FIB) technique [26]. Samples from both bulk and needle-shaped specimens were analysed using a Cameca LEAP 3000X HR operating in laser-mode with a specimen temperature of 50 K and a pulse energy of 0.6–0.8 nJ.

2.2. Oxide dispersion strengthened steel–14YT

The oxide dispersion strengthened steel (referred to from hereon-in as 14YT) was prepared by the weighing and mixing of high purity of elemental powders (Fe, Ti, Cr and Y_2O_3) to give a composition of Fe–14Cr–0.2Ti–0.3Y_2O_3 (all wt.%). The full composition is given in Table 2. The solid solution hardener W was not included in this composition in order to study the fundamental radiation response of the nanoclusters with no contribution from the W. The mixed powders were mechanically alloyed by ball milling (Simolayer CM-08) in an argon atmosphere for 4 h at 1000 rpm with a ball-to-powder ratio of 10:1. The milled powder was consolidated by extrusion at 1150 °C and then quenched prior to being tempered at 750 °C for 2 h and air-cooled.

Following sample preparation and polishing the specimens were irradiated at the Indira Gandhi Centre for Atomic Research (IGCAR), Kalpakkam, India using 5 MeV Fe^{3+} ions up to a total dose of 50 dpa at a temperature of 700 °C. The dpa was calculated based on SRIM calculations using the Kinchin–Pease model with a displacement energy E_d of 40 eV. Post ion-irradiation samples to be analysed by atom probe tomography were prepared from the bulk using a FIB technique [26]. The samples were then analysed using a Cameca 4000X HR in laser-mode with a specimen temperature of 50 K, pulse fraction of 20% and pulse energy of 03–0.4 nJ.

3. Results and discussion

3.1. Tungsten

Following ion irradiation of the W–3Re and W–5Re the data was analysed in a manner to search for evidence of clustering of the Re. A plot of the Re concentration distribution for the W–5Re un-implanted and 300 and 500 °C irradiated samples is shown in Fig. 1. The figure shows that for the conditions under which the specimen was irradiated (300 and 500 °C, 1.5 dpa), the distribution of the Re atoms is indistinguishable from that of the un-irradiated specimen i.e. the Re remains in solution and no segregation occurs (although improvements in detector efficiency should enable clusters containing 2–9 atoms to be identified [27]). This indicates that no clustering occurs in these samples and as such more focus will now be given to the W–25Re specimen.

Atom probe reconstructions of the W–25Re alloy following irradiation show strong evidence for cluster formation. These clusters are formed solely from the process of irradiation based on the lack of cluster formation following annealing of the un-irradiated alloys at 1200 °C for 12 h. Atom maps of the Re atoms from the W–25Re material irradiated to 1.5 dpa under 4 different conditions (300 °C, needle; 300 °C, bulk; 500 °C, needle; 500 °C, bulk) are shown in Fig. 2. Spatially separated clusters are evident throughout and it is particularly noticeable that the number density of the clusters is significantly higher in the lower, 300 °C, irradiation than in that of the 500 °C irradiation. In order to obtain more quantitative data on the clusters, the maximum separation method [28] was used to identify and separate the clusters from the matrix atoms. The

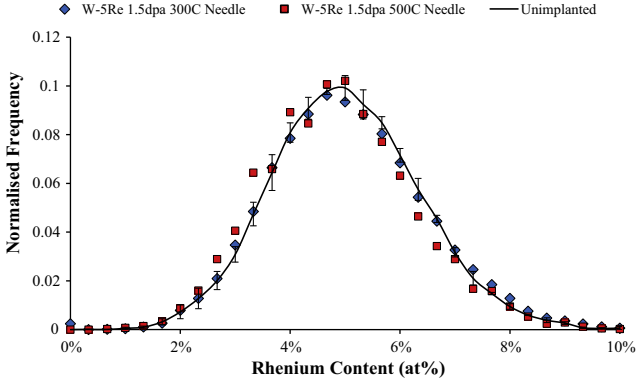

Fig. 1. Bulk rhenium concentration distribution in irradiated and non-irradiated W–5 wt.%Re.