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BEAM INTERACTIONS WITH MATERIALS AND ATOMS

Standardization of accelerator irradiation procedures for simulation of neutron induced damage in reactor structural materials

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Self-ion irradiation is widely used as a method to simulate neutron damage in reactor structural materials. Accelerator-based simulation of void swelling, however, introduces a number of neutron-atypical features which require careful data extraction and, in some cases, introduction of innovative irradiation techniques to alleviate these issues. We briefly summarize three such atypical features: defect imbalance effects, pulsed beam effects, and carbon contamination. The latter issue has just been recently recognized as being relevant to simulation of void swelling and is discussed here in greater detail. It is shown that carbon ions are entrained in the ion beam by Coulomb force drag and accelerated toward the target surface. Beam-contaminant interactions are modeled using molecular dynamics simulation. By applying a multiple beam deflection technique, carbon and other contaminants can be effectively filtered out, as demonstrated in an irradiation of HT-9 alloy by 3.5 MeV Fe ions.

1. Introduction

Incomplete interstitial-vacancy recombination during neutron damage and subsequent defect clustering processes lead to development of extended defects in reactor structural materials. Subsequent microstructural evolution leads to dimensional changes via void swelling and creep, mechanical property changes such as hardening and embrittlement, and local chemical composition changes involving grain boundary segregation and precipitation [1]. These changes involve property degradation and can significantly limit reactor performance and operation limits. For advanced reactor concepts with even harsher environments of temperatures and neutron damage levels, it is critical to develop accelerator-based ion irradiation testing to reach the damage regions well beyond that obtained in the existing testing reactors. The currently available ATR (Advanced Testing Reactor) at Idaho National Laboratory is able to reach only about 8 dpa (displacements per atom) per year, requiring decades to reach 400 dpa. In comparison, self-ion accelerators typically can attain >100 dpa per day.

The dpa concept has been widely accepted as a damage equivalency parameter to convert neutron damage to self-ion damage [1]. It represents a first order approximation and does not consider details such as damage cascade size effects or dpa rate effects. Most importantly, self- ion testing to study void swelling has significant limitations that arise from various "neutron-atypical" features. To mitigate such atypical issues, "best practice" procedures are being developed.

One neutron-atypical feature is "defect imbalance" along the ion path in the target arising from two primary phenomena. First, the use of mono-directional and mono-energetic ion beams induces momentum transfer from projectiles to target atoms skewed somewhat in the forward direction, producing interstitial profiles with depth that are slightly deeper than those of vacancies [2,3]. Such spatial distribution differences lead to slight vacancy enrichment in the shallow depth regions and interstitial enrichment closer to the projected range of implanted ions. Second, interstitial enrichment at deeper depth is further augmented by the implanted extra atoms themselves, greatly suppressing void nucleation and growth [4]. Previous studies have shown that void swelling occurs only in the shallow depth region and peaks roughly at about half of the projected ion range, instead at the dpa peak region [2]. Cross-sectional characterization of irradiated specimens is therefore a necessity and sampling for swelling analysis must avoid the dpa peak region.

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A second neutron-atypical feature is the "pulsed beam" effect, which occurs when a rastered or sweeping beam is used. Several recent studies have shown dramatic differences in void morphology at different rastering frequencies [5,6]. In general, the higher the rastering frequency, the less the swelling. Early studies suggested that between two beam pulses void nuclei tend to dissolve [7,8]. The earliest ASTM standard on ion-neutron simulation suggested that a static defocused and non-rastered beam must be used to simulate non-pulsed neutron damage [9].

The third neutron-atypical feature is "carbon contamination" and its tendency to suppress void swelling. It has been well known that electron or ion beam irradiation can induce a surface contamination layer which is rich in oxygen and carbon. Such contamination persists even when the target chamber pressure is better than 8×10^{-8} torr, but occurs only in the beam-irradiated region. Until recently the mechanism was unclear. It is proposed here that carbon, oxygen and nitrogen are entrained in the beam by Coulomb drag interactions with the energetic beam ions and delivered to the target surface with energies of low eVs. Carbon is especially known to suppress void swelling in both fcc and bcc iron-base alloys in the temperature range of interest [10,11]. While oxygen is usually captured at the surface, carbon can diffuse into the bulk, interacting with vacancies and can decorate larger open volume defects such as voids and bubbles. Extra carbon atoms can also induce carbide formation if the original carbon levels are high enough. Both carbon atoms and carbide precipitates are known to play significant roles in defect clustering processes, and can thereby impact the swelling behavior.

In this paper, we provide experimental evidence of beam-induced carbon contamination and describe an innovative beam management strategy to minimize carbon contamination. Details of contamination effects on void swelling will be reported elsewhere [12]. The present paper mainly focuses on understanding the mechanism of carbon contamination, illustrated using molecular dynamics simulations.

2. Experimental procedure

Ferritic-martensitic alloy HT-9 was irradiated with 3.5 MeV Fe ions at 475 °C to a fluence producing 600 peak dpa. Based on previous studies on HT9, the irradiation temperature was selected to maximize the swelling. The beam spot size was 6 mm × 6 mm, and was static without rastering. The damage was calculated by using the Kinchin-Pease mode in SRIM code and the displacement threshold energy was 40 eV. During irradiation, liquid nitrogen cooling was used to cool a long cold trap along the last beam flight segment to improve the vacuum such that the target chamber pressure was always better than 8×10^{-8} torr.

Two nearly-identical irradiation campaigns were conducted. The first campaign did not employ any beam filtering. The second employed multiple beam small-angle deflectors to reduce carbon contamination. The key was to create a zigzag beam trajectory to periodically kick contaminants out of the ion beam, due to mass and charge differences between contaminant atoms and the ion beam. Three magnetic beam deflectors, manufactured by NEC Inc., were positioned along the beam line ahead of the target. Furthermore, the last deflector was positioned as close as possible to the target chamber to minimize entrainment in the last beam flying segment.

The depth profiles of carbon and nitrogen were obtained by using secondary ion mass spectrometry (SIMS) with a cesium analysis beam of 4 keV, but only carbon will be addressed in this paper since the carbon introduction was more consequential. Structural characterization was performed by using transmission electron microscopy (TEM). The focused ion beam (FIB) lift out technique was used for TEM specimen preparation.

3. Modeling procedure

The mechanism of carbon entrainment by Fe ions was studied by molecular dynamics simulation, using the LAMMPS (Large-scale Atomic Molecular Massively Parallel Simulator) computer code [13]. The Fe-C interaction potentials were described by the Fe-C Tersoff potential which was smoothly linked to ZBL potential at short distances [14]. The cell size was 20 nm × 20 nm × 20 nm and contains 500 randomly-positioned C atoms. All of these C atoms were negative and single-charged. The system was thermally relaxed to reach equilibrium at 300 K for 500 ps using NVT ensemble. Subsequently, 20 Fe atoms of 1 MeV were introduced randomly in the cell's central region of 1 nm wide under NVE ensemble. The Fe atoms were allowed to re-enter the cell for repeated bombardments. The time step was 0.002 fs.

4. Results and discussion

Fig. 1 compares cross-sectional dark field TEM images of HT-9 after irradiation up to 600 peak dpa both with and without the multiple beam deflection technique. Superimposed on the figure are the SRIM-predicted dpa profiles. The top shows schematic beam trajectories for each campaign. Fig. 1a is the traditional approach without beam deflection. A high density of needle-like carbides are clearly visible in the region from the surface to a depth of ~1 μm. The bottom of Fig. 1a shows a high resolution bright field TEM image of one needle-like carbide which is determined to be M_3C. Prior to irradiation, $M_{23}C_6$ carbides are observed along grain boundaries. After irradiation, plate-like M_7C_3 and needle-like M_3C carbides were observed within the irradiated region. Fig. 1b shows the innovation using the multiple beam deflection technique. The damage region is free of needle-like carbides. Obviously beam deflection is able to effectively reduce the amount of irradiation induced carbides. Note that big pre-existing grain boundary

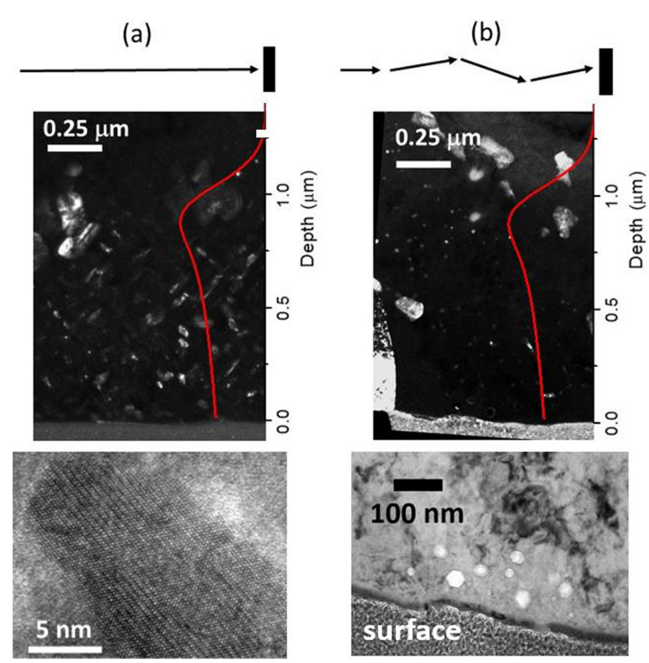

Fig. 1. (a) Dark field TEM image (top) showing carbides and bright field TEM image (bottom) of a typical carbide in HT-9 after irradiation up to 600 peak dpa at 475 °C without the multiple beam deflection technique and (b) dark field TEM image (top) showing reduced carbides and bright filed TEM image (bottom) of surface voids in HT-9 with the multiple beam deflection technique applied to filter out contaminants. Superimposed is the dpa profile and the ions enter the specimen from the bottom of the figure.

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