Journal of Nuclear Materials 471 (2016) 266–271

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

MeV per nucleon ion irradiation of nuclear materials with high energy synchrotron X-ray characterization

M.J. Pellin [a, *], Abdellatif M. Yacout [b], Kun Mo [b], Jonathan Almer [c], S. Bhattacharya [a, d], Walid Mohamed [b], D. Seidman [d], Bei Ye [b], D. Yun [b], Ruqing Xu [c], Shaofei Zhu [e]

[a] Materials Science Division, USA
[b] Nuclear Engineering Division, USA
[c] Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA
[d] Department of Materials Science and Engineering, Northwestern University, 2220 Campus Dr., Evanston, IL 60208, USA
[e] Physics Division, Argonne National Laboratory, Argonne, IL 60439, USA

ARTICLE INFO

Article history:
Received 18 June 2015
Received in revised form
4 January 2016
Accepted 5 January 2016
Available online 14 January 2016

Keywords:
Radiation damage
Characterization
x-ray
energetic ion
nuclear fuels
UMo

ABSTRACT

The combination of MeV/Nucleon ion irradiation (e.g. 133 MeV Xe) and high energy synchrotron x-ray characterization (e.g. at the Argonne Advanced Photon Source, APS) provides a powerful characterization method to understand radiation effects and to rapidly screen materials for the nuclear reactor environment. Ions in this energy range penetrate ~10 μm into materials. Over this range, the physical interactions vary (electronic stopping, nuclear stopping and added interstitials). Spatially specific x-ray (and TEM and nanoindentation) analysis allow individual quantification of these various effects. Hard x-rays provide the penetration depth needed to analyze even nuclear fuels. Here, this combination of synchrotron x-ray and MeV/Nucleon ion irradiation is demonstrated on U-Mo fuels. A preliminary look at HT-9 steels is also presented. We suggest that a hard x-ray facility with in situ MeV/nucleon irradiation capability would substantially accelerate the rate of discovery for extreme materials.

1. Introduction

Improved nuclear materials are necessary to advance the use of nuclear energy as a clean, safe and affordable energy source [1–3]. While the semiconductor, automotive [4], aerospace [5–7], and other industries have benefited from transformative improvements in materials over the past two decades, new material development for the nuclear industry has lagged. Reactor lifetime extension, enhanced accident tolerance, higher burn-up fuels, and advanced reactor designs all require fuel materials, structural materials and claddings that are able to withstand high radiation doses at elevated temperatures, in potentially corrosive environments, and under significant thermal and mechanical stresses.

Development of new materials and service life extension of existing materials have been hindered by the long in-reactor testing times needed to demonstrate resistance to radiation damage, consistent with rigorous qualification requirements. The traditional approach of performing neutron irradiation in test reactors, followed by post-irradiation testing and examination, is extremely time consuming and expensive. For example, test-reactor irradiation at a typical radiation damage dose rate (3 dpa per year) would take over 30 years to reach 100 dpa. Following such an irradiation evaluation is often hampered by the induced radioactivity of the sample. Such protracted testing greatly hinders the ability to iterate on composition, fabrication variables, and operating conditions to achieve desired levels of performance.

Radiation damage induced by energetic ions has been used as an alternative to reactor irradiation for the study of radiation damage in materials [8]. Comparisons between for instance proton and neutron irradiation effects, suggest that at the very least ion irradiation can be used to understand many of the phenomena found in neutron irradiation environments [9]. More recently, it is suggested that the use of MeV per nucleon ions as an irradiation source, coupled with synchrotron X-ray analysis can provide unique insights into the evolution of radiation damage in a solid [10–12].

Among the unique opportunities provided by this combination

* Corresponding author.
E-mail address: Pellin@anl.gov (M.J. Pellin).

http://dx.doi.org/10.1016/j.jnucmat.2016.01.004
0022-3115/

are: 1) a unique opportunity to simulate the effects of fission fragments in nuclear fuels; 2) The increased penetration depth of energetic ions allows the "bulk behavior" to be examined for cladding and structural materials, essentially eliminating surface-sink effects not representative of neutron damage. Moreover, distinct damage mechanisms are separated over the ~10 μm ion penetration depth, with the collisional (neutron like) damage isolated both from the surface sink and from other ion processes such as added interstitials; 3) The ion irradiation coupled with the penetrating ability of focusable hard x-rays allows the interrogation of individual grains within solid material sample. It reveals in a single sample many of the features of radiation damage, including the effects of added interstitials, swift ion effects, and the role of interstitial/vacancy trapping. 4) The single-effect physics investigations enabled by such combined MeV per nucleon irradiation and synchrotron X-ray advances are invaluable for validating computational models and elucidating the differences between ion and neutron irradiation and 5) at higher energies the sputtering yield of ions incident on the surface drops allowing much higher total damage to be placed in a sample.

As a test of the technique two types of samples are chosen for irradiation. U-Mo fuel samples are used to illustrate the use of "fission fragment equivalents" to produce radiation damage since the primary damage in nuclear fuels arises from fission fragments. Xe ions with 84 MeV energy were chosen to be representative of typical fission fragment. Second, a nuclear structural material typically considered for cladding in fast reactors, HT9 was chosen for irradiation. In structural materials, the damage in a nuclear reactor arises from neutron irradiation. The HT9 was also irradiated with 84 MeV Fe ions. In this case, 84 MeV Fe ions have a more complex interaction with the material, which varies as a function of depth, when compared to neutron irradiation. By using synchrotron X-rays at the Advanced Photon Source (APS) to resolve damage as a function of depth, we seek to reveal the collisional part of the ion damage that is most similar to that caused by neutrons. We note that the paper is meant to highlight the breadth of capability this technique offers rather than to provide a detailed comparison of radiation damage in these materials. This will be set forth in subsequent publications.

2. Experimental procedure

In summary, during this study irradiation of two types of UMo fuels, U-7Mo and U-10Mo (7 and 10 wt. % Mo, respectively) were irradiated with Xe ions. The U-10Mo was irradiated were irradiated with 105 MeV Xe^{26+} ions. In this case the angle of incidence was 45°. The final fluence of the irradiation was 2×10^{16} Xe/cm^2 which is equivalent to a peak damage of about 150 dpa according to the SRIM-2008 calculation with a full cascade model [13]. U-7Mo w were irradiated with 84 MeV Xe^{26+} ions along the target normal (achieving a similar penetration depth to the U-10Mo samples) to a calculated 150 dpa peak damage.

Also irradiated was a nuclear structural material, Fe-12Cr-1Mo-0.5 W-0.5Ni-0.25 V-0.2 C (wt.%) is referred to here as HT9. High-energy Fe^+ ion-implementation with 84 MeV Fe^+ ions was conducted on this material at ATLAS. The radiation damage region is about 7.4 μm to the outer surface, and the peak radiation damage is ~40 DPA, based on SRIM calculations.

3. Materials

Two types of materials selected for this study, and as indicated in Fig. 1, a Gaussian ion beam with varying intensity as a function of sample position was used on each the two types of samples.

The first samples correspond to U–Mo fuels with high uranium densities have been developed for use in high performance research reactors in order to convert those reactors from the use of highly enriched uranium (HEU) to the use of low enriched uranium (LEU). Because of this interest, these fuels have been extensively studied in the nuclear reactor environment. As such they make good candidates for MeV/nucleon ion irradiation comparisons.

Two morphologies of this fuel type have been examined. Depleted U–10 wt. % Mo (denoted as U-10Mo) monolithic miniature size plates were used. Mechanical grinding and polishing were performed to remove the Zr clad on one side of the plate. The final finish of the polishing was by 1200 grit SiC paper. This sample was then irradiated through the polished surface at a 45-degree angle.

Also irradiated is a U–7 wt. % Mo (denoted as U–7Mo) particulate fuel sample. A miniature fuel plate [10 cm (length); 2.5 cm (width); 0.14 cm (thickness)] was fabricated at ANL for the ion irradiation. The plate had an 8 g U/cm^3 dispersion of atomized natural U-7Mo alloy powder in a pure Al matrix. The atomized natural U–7Mo powder was provided by Korea Atomic Energy Research Institute (KAERI). For the ion irradiation experiment, small disks of 1.7 mm diameter were punched through the thickness of the fuel plates. One side of the cladding was removed using abrasive paper (with a final polish done with 5 μm paper) to get access to the zone containing U-7Mo powder. During irradiation, the polished surface was exposed to the ion beam after loading into the sample holder displayed in Fig. 1b.

An advanced nuclear structural material, Fe-12Cr-1Mo-0.5 W-0.5Ni-0.25 V-0.2 C (wt.%) is referred to here as HT9. HT9 steel has been an attractive candidate material for various nuclear applications since 1970s [14]. High-energy Fe^+ ion-implementation with 84 MeV Fe^+ ions was conducted on this material at ATLAS. The ion beam had a 10 mm diameter Gaussian beam shape. Under irradiation the sample temperature rose to ~250 °C. The radiation damage region is about 7.4 μm to the outer surface, and the peak radiation damage is ~40 DPA, based on SRIM calculations.

3.1. APS sample geometry

Two different synchrotron diffraction techniques were used here: (1) high-energy X-ray diffraction (at 1-ID beamline at APS); and (2) microbeam Laue diffraction (at 34-ID beamline at APS). A typical high-energy x-ray diffraction experiment involved preparing an irradiated specimen as described above using a focused Ga ion beam (FIB), and welding it on top of a tungsten needle. (Fig. 2) The energy of synchrotron X-ray is adjusted to ~62 keV in the experiment. The X-ray beam was vertically focused to ~2 μm and horizontally defined by slits to 20 μm. For the microbeam Laue diffraction (0.5×0.5 μm^2 in full-width half-maximum), due to a lower energy and smaller beam size, the X-rays only probed a single grain within a bulk U-10Mo sample (10 mm × 3 mm × 0.4 mm). The sample geometry used in this experiment has been previously described [15].

3.2. Irradiation

All irradiations were performed at the Argonne Tandem Linac Accelerator System (ATLAS). With thermocouples connected to the sample stage and an Infra-red camera, the thermal rise of the sample was determined during ion irradiation. The total current was constantly measured to each sample with a digital volt meter with the sample biased by a few volts to hold secondary electrons on the sample surface. Since the charge state of the chosen ions is determined in a mass separator, this current can easily be transformed into the total dose received at the sample. While not changed here during irradiation, the beam angle of incidence can be adjusted by rotating the sample placed in front of the beam. This