In-situ study:Faulted Loop and Void Behavior in Single Beam Bulk Irradiated Fe-21Cr-32Ni Model Alloy

Muhammet AYANOGLU[1] and Arthur T. MOTTA

Department of Mechanical and Nuclear Engineering, Pennsylvania State University

Introduction

The microstructure of the materials in the reactor core continuously evolves during reactor operation due to the atomic displacements resulting from interactions with high-energy neutrons. For some proposed fast reactors, the amount of displacement damage is expected to reach up to ~200 dpa (displacements per atom) at operating temperatures above 400ºC[1]. Therefore, understanding of how materials behave under irradiation is essential. The major challenge of neutron irradiation experiments is the requirement of very long exposure times, as well as the handling of highly radioactive samples needing the usage of hot cells which increases costs.

As an alternative, ion irradiation has remarkable advantages. For example, high damage rates can be achieved such that, doses equivalent to years of irradiations can be achieved in days with no or very low residual activity. Greater control of irradiation conditions allows a wide variety of different irradiation conditions can easily be adjusted to study their individual effects.

In this study, faulted loop and void behavior in single beam bulk irradiated Fe21Cr-32Ni, a simple model alloy analogue to the commercial alloy 800H, was investigated using transmission electron microscopy (TEM). In order to extend our understanding on faulted loop and void behavior, additional in-situ experiments were performed at Intermediate Voltage Electron Microscopy (IVEM) with the TEM samples using 1 MeV Kr ions. The results of these irradiations are discussed here.

Experimental Methods

Bulk 21Cr32Ni samples were irradiated at the Michigan Ion Beam Laboratory (MIBL) to doses of 1 dpa, 10 dpa and 20 dpa with single beam 5 MeV Fe^{++} ions at 440ºC and a dose rate of 1×10^{-3} dpa/s. The target dose was achieved at around ~0.6 µm depth from the irradiated surface. TEM specimens were then prepared from the bulk materials by using focused ion beam (FIB) with 2 keV final thinning. The irradiated microstructure was studied using bright field/dark field (BF/DF) micrographs.

To allow for measurement of defect density, the thicknesses of the samples were systematically measured using Convergent Electron Beam Diffraction (CBED) and Energy Filtered TEM (EFTEM). The damage profile and ion range of 5 MeV Fe ions in 21Cr32Ni alloy were determined by using Stopping Range of Ion in Matter (SRIM) with the Kinchin-Pease-Quick calculation mode and displacements energy of 40 eV.

The faulted loops were quantified in terms of defect diameter and density using rel-rod dark field imaging. The overfocus/underfocus technique was used to image voids and ImageJ software was used to measure their size.

In-situ irradiation experiments were performed using 1 MeV Kr^{++} with the damage rate of $1x10^{-3}$ dpa/s at temperatures ranging from -223ºC to 440ºC.

Results

Fig. 1 shows a typical damage profile and ion range of 5 MeV Fe ions into 21Cr32Ni model alloy calculated with SRIM. According to this

[1] mua214@psu.edu

damage profile, the peak dose is found at ~1.3 µm while the maximum range of ions is ~2 µm.

Rel-rod dark field images captured near ~0.6 µm depth from the surface are shown in Fig. 2. Since only one family is shown, these represent only 1/4 of all faulted loops observed in the 1, 10 and 20 dpa bulk irradiated samples. Faulted loops were observed to be uniformly distributed in the entire irradiated region, and no faulted loops were observed beyond the ion range. Quantitative results show that between 1 and 20 dpa the average faulted loop diameter decreases by a factor of ~2 while the loop density increases by factor of ~10.

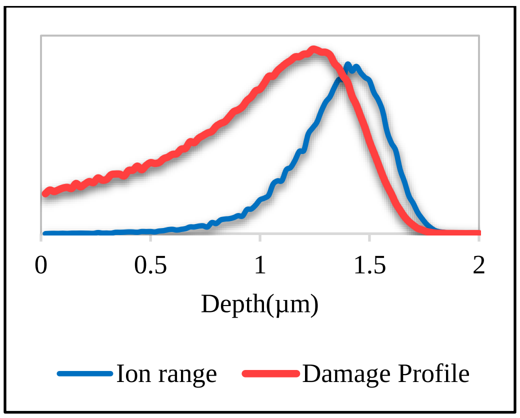

Fig.1 Damage profile and Ion range as calculated by SRIM for 5 MeV Fe atoms into Fe21Cr32Ni model alloy.

To further investigate the faulted loop behavior during irradiation, a FIB sample was lifted out from material bulk irradiated to 1 dpa and then further re-irradiated at IVEM. In this experiment, a rel-rod condition was maintained and one of the large faulted loop (~116 nm in diameter) was tracked throughout the re-irradiation.

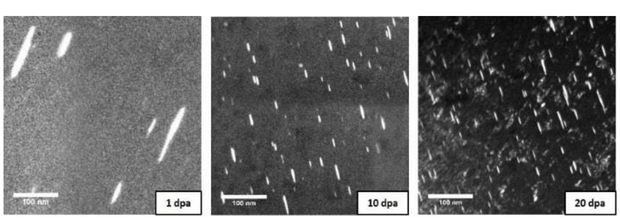

Fig.2 Dark field micrographs of Fe21Cr32Ni model alloy using the rel rod reflections of faulted loops after irradiation to 1, 10 and 20 dpa at 440ºC.

Fig. 3 shows a video capture time sequence of loop unfaulting observed after an additional dose of +1.25 dpa (2.25 dpa in total) during the in-situ irradiation. In this figure, the initial time of zero indicates the starting point when the loop was first observed to be unfaulting through the interaction with another dislocation. The total unfaulting time for this particular loop was ~60 sec and the given dose is ~+1.3 dpa. In other words, once the loop started unfaulting, it took only ~+0.05 dpa for become unfaulted, and thus invisble to the rel rod imaging. During irradiation, formation of new small faulted loops was observed (not shown in the figure). This real time observations provide insights on how faulted loops behave during the irradiation.

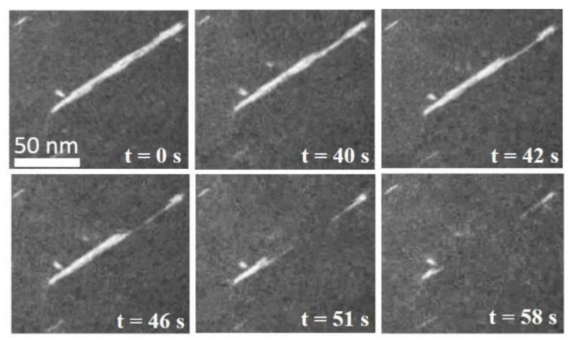

Fig.3 Rel-rod dark field images showing time sequence of loop unfaulting during the in-situ irradiation.

Further in-situ experiments were performed with the FIB samples lifted out from the 1 dpa bulk irradiated material at a wide variety of temperatures between -223ºC to 440ºC to study the void behavior in TEM foil during the in situ re-irradiation. Interestingly, voids were observed to shrink during re irradiation. This occurred at all temperatures even at cryogenic temperatures where both interstitials and vacancies are expected to be immobile. Fig. 4 shows the shrinkage and disappearance of individual voids during IVEM re-irradiation at 440ºC.

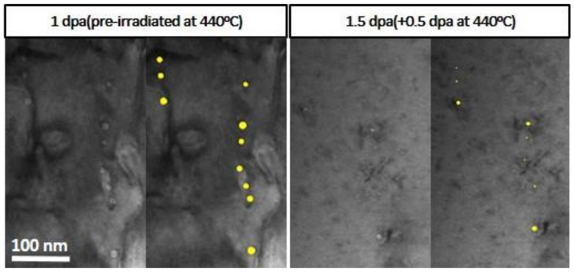

Fig.4 Void shrinkage observed in 21Cr32Ni model alloy during the IVEM irradiation of 1 dpa pre-irradiated sample. The yellow dots indicate the void positions.

The individual void diameters were measured at different doses and the average shrinkage rate for all voids was calculated (Fig.5). Results show that the average shrinkage rate $(dR/dt)_{ave}$ shows little variation with temperature. The absolute value of the average shrinkage rate increases with irradiation time as shown in Fig.5. This behavior is due to the fact that the void diameter decreases with time resulting in a higher surface-to-volume ratio and a faster shrinkage rate.

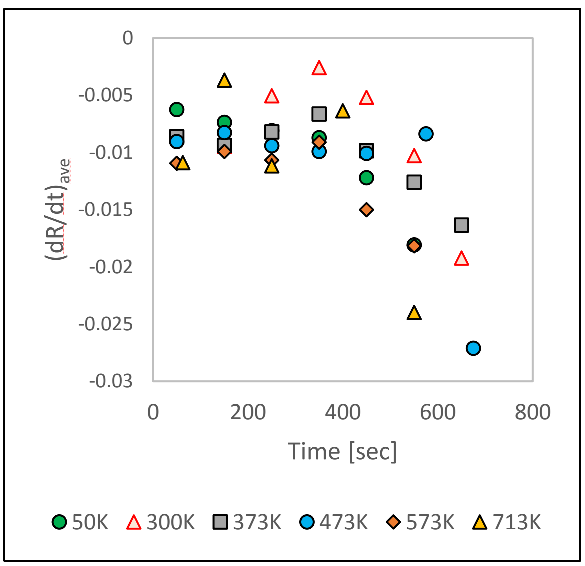

Fig.5 Average void shrinkage rate during IVEM irradiation vs. time for various irradiation temperatures.

Annealing experiments in which samples are held at temperature for ~30 minutes at room temperature 400ºC in the absence of irradiation showed that the voids are stable during annealing which indicates that thermal emission of vacancies is not an effective mechanism for cavity shrinkage. A significant net interstitial flux would be needed to explain the void shrinkage results and it is clear that this only occurs during thin foil irradiation. The mechanism for void shrinkage is under investigation but its presence allows us to put some constraints on defect migration and formation energies.

Conclusion

The microstructural evolution of 21Cr32Ni model alloy irradiated to 1, 10 and 20 dpa single beam 5 MeV Fe ions was investigated.

The average faulted loop diameter decreases between 1 and 10 dpa likely due to loop unfaulting, remaining more or less constant up to a dose of 20 dpa, whereas the loop density increases. In-situ experiments performed with the FIB samples lifted out from 1 dpa bulk irradiated material showed that the faulted large loops become unfaulted during the irradiation. In addition, new faulted loops start forming which explains the faulted loop behavior at elevated doses.

Void behavior was investigated in-situ at different temperatures. It was observed that the voids shrink even at cryogenic temperatures where both interstitials and vacancies are expected to be immobile. More interestingly, comparison of average void shrinkage rate shows almost no significant temperature dependence of the observed void shrinkage rate.

Reference

[1]J. GAN, J. I. COLE, T. R. ALLEN, Shutthanandan and S., Thevuthasan, S., Journal of Nucl. Mat., 351, (2006), p. 223-227.

[2] L. K. MANSUR, *Nuclear Technology* 40.1 (1978): 5-34.