Investigation of the performance of different types of zirconium microstructures under extreme irradiation conditions

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Abstract

The safe and continued operation of the US nuclear power plants requires improvement of the radiation resistant properties of materials used in nuclear reactors. Zirconium is a material of particular interest due to its use in fuel cladding. Studies performed on other materials have shown that grain boundaries can play a significant role on the radiation resistant properties of a material. Thus, the focus of our research is to investigate the performance of different zirconium microstructures under irradiation conditions similar to those in commercial nuclear reactors. Analysis of the surface morphology of zirconium both pre- and post-irradiation was conducted with Scanning Electron Microscopy (SEM). Cold-rolled (small-grain microstructure) and annealed (large-grained microstructure) zirconium samples were mechanically polished in order to be irradiated. Room temperature irradiation of zirconium samples was conducted at energies of 100 eV and 1 keV with He^{+} ions at a flux of 1 $x10^{20}$ m^{-2} using a gridded ion source. High temperature (350°C and 700^{0}C) He^{+} irradiations were performed at an energy of 100 eV using a gridless end-hall ion source at the same flux. Transmission Electron Microscopy (TEM) was conducted to determine the grain size of the zirconium samples. Preliminary results show greater surface damage on the rolled zirconium samples than on the annealed samples for all irradiation cases. The difference in damage was most evident in high temperature irradiations. Further work is necessary to evaluate why the small-grain zirconium exhibited greater damage. Future testing will be performed using higher fluxes, temperatures and energies.

1. Introduction

Zirconium is a material of particular importance to nuclear reactors due to its use in fuel cladding. Some of the properties that make zirconium an ideal material for fuel cladding is its low neutron absorption and high resistance to corrosion.

Current methods for improving the physical properties of zirconium are mainly based on alloying. Zircaloy-4 was a zirconium alloy developed for use in nuclear reactors and its composition is detailed in Table 1.

Table 1: Zircaloy-4 element composition [1]

Element	Weight (%)
Sn	1.2-1.7
Fe	0.12-0.18
Cr	0.05-1.5
Ni	0.007

Despite the favorable physical properties of zircaloy-4, the extreme radiation environment found in nuclear reactors continues to result in degradation of physical properties and eventually leads to material failure. Thus, it is important to improve the radiation resistance of zirconium and its alloys.

Studies performed on tungsten and carbon steels [2,3] have shown that increasing the grain boundary density of these materials enhances the tolerance to radiation induced defects. Both studies dealt with ultra-fine grain (100nm – 1000 nm) and nanocrystalline (less than 100 nm) tungsten and carbon steel.

The study performed by El- Atwani et. al. irradiated tungsten with He^+ ions and demonstrated that grain boundaries not only act as defect sinks, but they can also act as helium traps. Thus, materials with high grain boundary density have a decreased concentration of He intergranularly. However, the nucleation of helium bubbles at the grain boundaries can result in degradation of mechanical properties (embrittlement, reduced creep resistance) and enhanced grain boundary grooving [2]. The study performed by El-Atwani et. al was meant to model tungsten irradiation in plasma facing conditions of fusion reactors. Helium in fission reactors has a negligible impact on radiation damage of the fission reactor components. However, the experimental conditions of the zirconium study discussed in this report are similar to those conducted by El-Atwani et. al.

The study performed by Murty et. al. involved the irradiation of carbon steel with neutrons. These conditions are a better model for the radiation environment of a fission reactor and can be the subject of future work for the current study on zirconium.

Despite the differences in materials and irradiation conditions, both studies on tungsten and carbon steels reported the lowest damage to have occurred for materials with grains below 50 nm.

2. Experimental

2.1. Materials

The materials employed for this investigation were cold-rolled and annealed zirconium samples. Cold-rolled zirconium samples correspond to a small-grain microstructure, while annealed zirconium samples correspond to a large-grain microstructure.

2.2. Mechanical polishing

Both types of zirconium samples were mechanically polished prior to irradiation. Mechanical polishing was performed using Pase Technologies 1000-grit SiC abrasive paper and 1200-grit Alumina abrasive paper. Further polishing was done using South Bay Technologies diamond abrasive films ranging from 30 microns to 0.5 microns.

2.3. Irradiation and surface characterization

Room Temperature (RT) irradiations of both types of zirconium samples were performed at the Interaction of Materials with Particles and Components Testing (IMPACT) experimental facility at Purdue University. The energies used were 100 eV and 1 keV using He^+ ions at a flux of $1x10^{20}$ $s^{-1}m^{-2}$ with a gridded ion source. The total fluence was $5x10^{22}$ m^{-2}.

High temperature irradiations were performed in the High Heat Flux (HHF) laboratory at the Center for Materials under Extreme Environment (CMUXE). Irradiations were performed at temperatures of 350°C and 700°C using He^+ ions at an energy of 100 eV using a gridless end-hall ion source. The flux and fluence were the same as in the RT irradiations.

The pre- and post-irradiation surface characterization of all zirconium samples was conducted using a Hitachi S4800 FESEM in Birck Nanotechnology Center at Purdue University. Cross-section imaging was conducted using a FEI xT Nova NanoLab Dual Beam Focused Ion Beam/Scanning Electron Microscope (FIB/SEM).

2.4. Grain size determination

Unirradiated cold-rolled and annealed zirconium samples were characterized using TEM in order to determine the grain size. Preparation of the samples for TEM required electropolishing in a 1 liter solution of 85% methanol-15% perchloric acid at a temperature between -30°C and -40°C. The electropolishing voltage was 20 V and the current was 80 to 100 mA. TEM characterization was performed using a FEI Titan 80/300 field emission TEM with a 300 kV operating voltage.

3. Results and discussion

As seen in Figure 1, TEM micrographs showed that the annealed zirconium samples have large grains (5-10 μm) with sharp grain boundaries. The rolled sample grains were found to be of ultrafine quality, though the size of the grains was difficult to determine due to non-sharp grain boundaries.

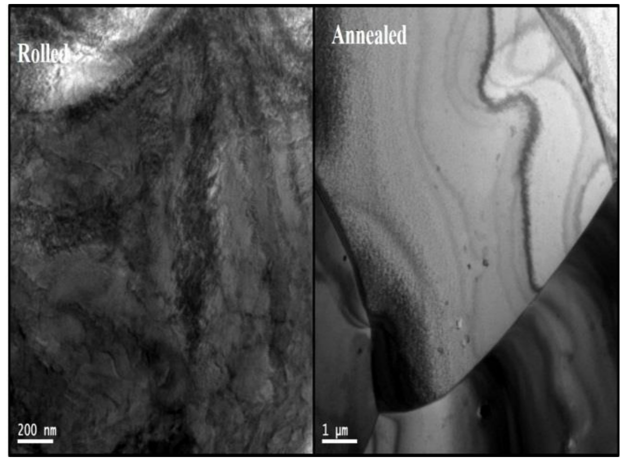

Figure 1: TEM micrographs of the rolled and the annealed samples

SEM micrographs of the unirradiated zirconium samples were taken for comparison purposes with the irradiated samples. Figure 2 shows little morphological effects due to mechanical polishing of the samples.

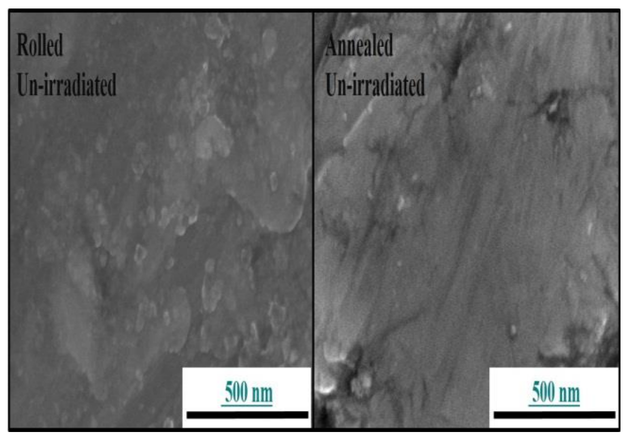

Figure 2: SEM micrographs of the un-irradiated rolled and annealed samples

Figure 3 shows the surface morphology after RT irradiation with an energy of 100 eV. Surface damage on both samples was characterized by small blisters and large holes, though the damage was more evident in the rolled sample. Defects are primarily formed due to bubble formation and possible loop punching or trap mutation. However, defect migration is slow at RT so only small bubbles formed.

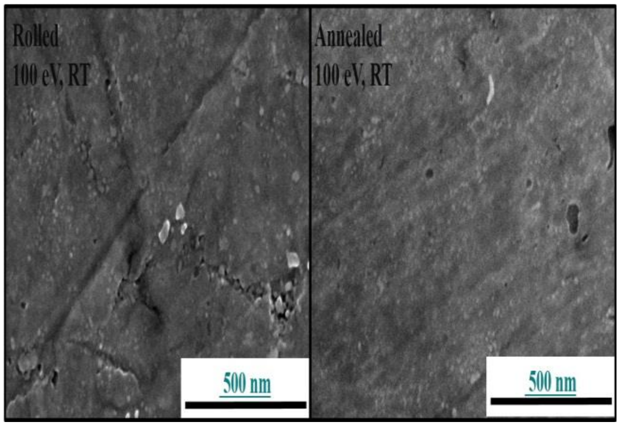

Figure 3: SEM micrographs of both samples irradiated with 100 eV He$^+$ at RT

Increasing the temperature to 350°C (temperature relevant to Boiling Water Reactors) resulted in an increase in the density of surface voids and blisters. Figure 4 shows the surface damage to both samples. The rolled sample surface was characterized by high blister density and faceted voids. The latter were likely created by helium bubbles bursting on the surface. Despite a high density of surface voids on the annealed sample, the damage was less than what was observed on the rolled sample.

Further increasing the temperature to 700°C (temperature relevant to Pressurized Water Reactors) resulted in even greater damage to the zirconium surface. Figure 5 continues to show greater damage to the rolled sample in comparison with the annealed sample. At this temperature and energy, the rolled sample surface was covered with blisters and intergranular micro-cracks. Micro-crack formation is attributed to enhanced grain boundary grooving due to helium bubble formation at the grain boundaries coupled with irradiation enhanced diffusion.

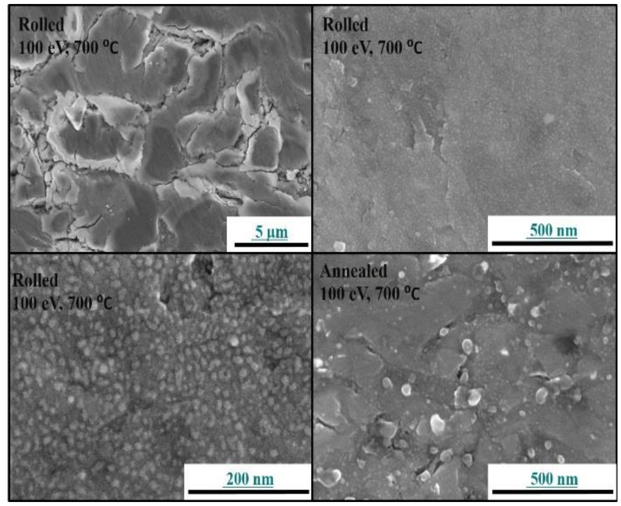

Figure 5: SEM micrographs of both samples irradiated with 100 eV He$^+$ at 700°C

Increasing the irradiation energy to 1 keV at RT led to greater damage on both samples. For this irradiation condition, the surface damage was comparable for both samples as seen in Figure 6.

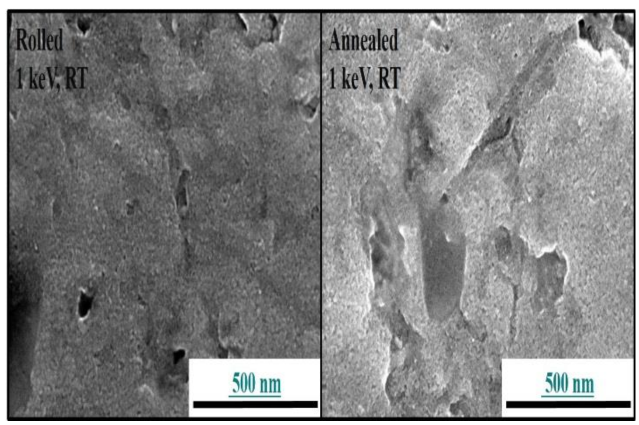

Figure 6: SEM micrographs of both sample irradiated with 1 keV He$^+$ at RT.

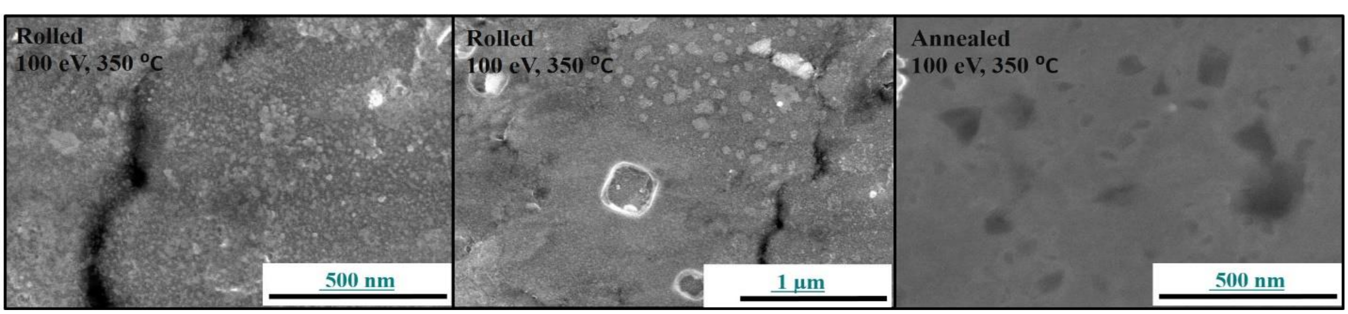

Figure 4: SEM micrographs of both samples irradiated with 100 eV He$^+$ at 350°C

Cross-section images of both samples were taken in order to better compare the damage from irradiation. As seen in Figure 3.7, the thickness of the damaged layer for 1 keV and RT conditions was comparable in both samples. The low contrast below the damaged layer in the rolled sample demonstrates its ultrafine microstructure. At 700°C and 100 eV, the thickness of the damaged layer was much greater in the rolled sample. The thickness of the damaged layer in the rolled sample was several microns in size which exceeded the He penetration depth (10 nm) from TRansport of Ions in Matter (TRIM) simulations.

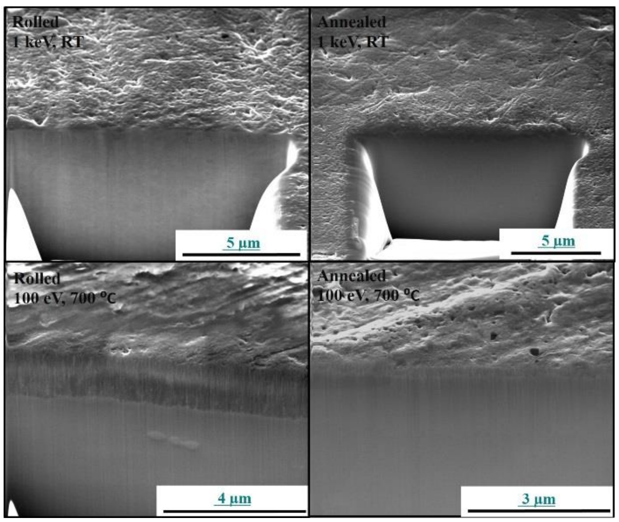

Figure 7: Cross-sectional FIB/SEM images of both samples irradiated with 1 keV at RT and 100 eV at 700°C

4. Conclusions

The purpose of this study was to investigate and compare the performance of two types of zirconium microstructures under extreme irradiation conditions in order to determine which would be more suitable for nuclear reactor applications.

The majority of the irradiation conditions that were tested resulted in greater damage to the rolled sample. This deviated from the expectation that increasing the grain boundary density would result in enhanced radiation resistance. Further work is necessary to explain why this deviation occurred.

The general trend observed was that increasing the temperature led to greater damage to the surface of the samples. Thus, similar high temperature experiments will be conducted with an energy of 1 keV. Higher flux irradiations will be conducted to simulate reactor conditions more accurately. Thermal Desorption Spectroscopy will be conducted on both types of irradiated samples to study He trapping phenomena.

The same experiments will then be repeated for reactor grade zircaloy samples.

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