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RADIATION DAMAGE STUDIES IN CTR MATERIALS
USING DUAL-BEAM IRRADIATION

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INTRODUCTION

Basic radiation damage mechanisms in materials can be conveniently studied through the use of ion bombardment techniques. In particular, radiation damage similar to that expected in the first wall of a fusion reactor can be simulated to a reasonable degree in a dual ion accelerator system. In a dual ion system, heavy ions, e.g., Ni^{++} simulate the neutron damage and He^+ ions simulate the helium generated by n, α reactions in a reactor. Because of the much higher dose rate from heavy ions compared to neutrons, high damage levels can be achieved in a very short time. In addition, experimental parameters are easily controlled, there is no induced radioactivity and costs are much less compared to reactor experiments. The main and limiting drawback to the ion bombardment technique is the small depth of radiation damage, typically extending only 1-2 μm from the surface. As a result, only transmission electron microscopy (TEM) has been used to study the radiation damage.

Radiation damage studies using accelerator techniques are underway at Pacific Northwest Laboratory (PNL) and this report summarizes some of the recent studies. A brief description of the dual ion accelerator facility is first presented. This is followed by a brief summary of some recent studies on refractory metals. These studies include the effect of helium on radiation damage in molybdenum and the role of oxide precipitates on radiation damage in Nb-Zr alloys.

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EXPERIMENTAL APPARATUS AND TECHNIQUES

A complete description of the apparatus can be found in Reference 1. The following discussion summarizes the apparatus and describes recent changes in the system.

Accelerator System

The beam lines from two accelerators intersect at approximately 32° to form the dual beam ion irradiation system, Figure 1. A 2 MeV tandem accelerator is used for the heavy ions. Negative ions are produced in a sputter ion source, accelerated to the terminal potential, stripped of electrons and accelerated to ground potential at the target. The 1.8 MeV light ions, such as He^+ , are produced in a Van der Graaff accelerator. These ions are further degraded to a lower energy inside the target chamber in a manner to be described later. Additional pumps have been installed in the beam lines near the target chamber, Figure 1, in order to keep the vacuum in the target chamber as high as possible.

Target Chamber

The target chamber is an all metal sealed, ion pumped high vacuum system. Operation in a vacuum as low as 1×10^{-8} torr is possible. Within the target chamber are the specimen holders and furnaces, beam alignment aperture, Faraday cups, helium beam degrader foils and crystal detectors. Each of these features are described in the following sections.

Specimen Holder. Specimens are clamped onto heaters which are positioned on the outer diameter of a rotating apparatus referred to as the carousel, Figure 2. The specimens are 3 mm diameter disks suitable for insertion in the electron microscope. However, only the center 2 mm are bombarded. Up to 8 specimens can be irradiated before reloading and each specimen can be individually heated to 1450 K. Temperature measurement and control is provided by an infrared pyrometer. One specimen position has a thermocouple which provides an emissivity standard for the particular material under irradiation.

Beam Alignment and Measurement. Alignment of the two beams on the specimen is carried out through the use of an aperture. This 2 mm beam alignment aperture is on the same relative position on the carousel as the specimens. The carousel is adjusted until both beams are passing through the aperture as measured by Faraday cups placed behind the carousel, Figure 2. This position is noted by the reticle on the infrared pyrometer. During an experiment, each specimen to be irradiated is rotated into this marked position. For maximum dose rate, the heavy ion beam is focused until one-half the total intensity in passing through the 2 mm aperture. As the beam shape is approximately Gaussian, the intensity at the beam edge is still 80% of maximum.

Beam current is detected in several ways. Prior to placing the two beams on the specimen, each beam current is measured on a separate Faraday cup placed behind the specimen. During irradiation, the beam current is measured intermittently by means of a swinging paddle inserted into the beams ahead of the specimen (Figure 2). Measurement of beam current on the specimen has not been too successful because of stray currents. Large changes in current such as loss of the beam would be detected, however.

Degradation of Helium Beam. The helium energy must be lowered so as to match the helium deposition with the damage produced by the heavy ions. A 5 μm thick aluminum foil placed just inside the target chamber degrades the energy from 1.8 MeV to a range of 200-400 KeV. A surface barrier detector has been placed so as to pick up a portion of the scattered beam that passed through the foil, Figure 2. The energy distribution of this scattered beam is then recorded on a multi-channel analyzer. From the energy distribution and range measurements, the distribution of the implanted helium can be calculated.

The amount of helium was experimentally determined through the analysis of the size and density of gas bubbles observed in helium bombarded molybdenum. Assuming the gas bubbles to be filled with an equilibrium amount of gas, the amount of helium was calculated from microstructures taken from different depths in the bombarded region. The amount of helium was then plotted as a function of depth, Figure 3. The shape of the helium distribution agrees well with calculated curves but the amount of helium determined experimentally is

less than calculated by a factor of 3-4. Backscattering experiments are planned to find the cause of this discrepancy.

Microstructural Analysis

After the irradiation, the damage induced microstructures are analyzed in the electron microscope. Electrothinning techniques have been developed so that regions within one micron of the bombarded surface can be selectively isolated and observed in the electron microscope. All features of the induced radiation damage, i.e., dislocation loops, dislocation network, voids and precipitates are analyzed using standard transmission electron microscopy techniques.

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RECENT RESULTS

Dual Beam Irradiation of Molybdenum

The effect of simultaneous helium injection on internal void formation during heavy ion irradiation in molybdenum has been studied. Such studies provide information useful in the understanding of radiation damage to fusion first wall materials in which large amounts of helium will be generated. Comparison of the ion irradiation damage with neutron fission reactor damage has also been made.

The molybdenum was bombarded with 7.2 MeV Ta^{+++} under a range of irradiation conditions. The temperature range extended from 1170 K to 1470 K. The dose level ranged from 1 dpa (atom displacements per atom) to 80 dpa at a dose rate of 5×10^{-3} dpa/sec. For comparison, a typical dose rate in a fusion first wall is in the range of 5×10^{-7} to 1×10^{-6} dpa/sec. The ratio of ppm of helium to dpa was in the range of 5 to 10. This compares with a value of ~ 7 for a first wall made of molybdenum.

Simultaneous helium injection resulted in a smaller void size and higher void density after high dose levels at 1270 K compared to no helium injection, Figure 4a and 4b. Apparently, the helium is promoting void nucleation to higher dose levels than would normally occur in the absence of helium. There is, however, little effect of the helium on the void volume fraction (swelling) or swelling rate since the higher void density is compensated by the smaller void size, Figure 5a, 5b.

The line drawn in Figure 5 actually represents the dose dependence of swelling for fission neutron irradiated molybdenum in the temperature range 870-1070 K but reasonably represents the swelling rate in the ion bombarded molybdenum. A temperature shift, i.e. 1270 to 1070, however, has been found necessary when comparing ion and neutron irradiation data due to the large difference in dose rate. The swelling does tend to level off at high doses in the case of ion irradiation. If neutron data extended to such high levels, a similar phenomenon might be observed.

The temperature dependence of swelling was little affected by the presence of helium, Figure 6. The large scatter in the data could hide some subtle differences. However, large differences in void size and size distribution were noted at the highest temperature of 1470 K. In specimens with no helium, the voids were reasonably homogeneous in size and size distribution, Figure 7a. In specimens containing helium, the void distribution was very heterogeneous in that clusters of small voids were interspersed among a low density of large voids, Figure 7b. These observations suggest that more significant helium effects could be observed at still higher temperatures.

An inherently high void nucleation rate in molybdenum has been put forth as the principle reason for a lack of a strong helium effect. The high void density is the main sink for both the irradiation produced vacancies and interstitials, so little void growth can occur. Similar conclusions have been reached in other studies on iron and nickel base alloys.⁽³⁾ Namely, in those alloys that showed high void nucleation in the absence of helium, the swelling was little affected by simultaneous helium injection.

Ion Bombardment of Nb-Zr Alloy

The influence of small precipitates on the damage microstructure in a refractory alloy was the principle objective of this study. Nb-Zr alloys can be internally oxidized so as to form a fine dispersion of ZrO_2 in the matrix. A comparison of the damage microstructure both with and without prior internal oxidation was therefore made.

A Nb-1 wt% Zr alloy was internally oxidized at 1000 K for 32 hr. The resulting ZrO_2 precipitates were extremely small, ≤ 3 nm, and of very high number density. Other Nb-1Zr specimens were annealed for similar times in high vacuum to serve as unoxidized standards.

The alloys were bombarded with 5 MeV Ni^{++} ions over the temperature range of 970-1170 K. Previous work on Nb and Nb-1Zr has shown a swelling peak within this range.^(4,5) Doses up to 45 dpa were attained at a dose rate $\geq 5 \times 10^{-3}$ dpa per second. No helium beam was used in this study.

Bombardment of specimens with no prior internal oxidation produced an inhomogeneous complex structure of dislocations, precipitates and voids, Figure 8a. The irradiation induced precipitates were identified as ZrO_2 . The precipitates were attached to voids in many cases but it was not possible to determine which feature nucleated first. The void volume fraction increased with temperature, then leveled off in contrast to the swelling peak observed in the previous study.⁽⁵⁾

Bombardment of the internally oxidized Nb-1Zr produced quite different structures. A dense dislocation network developed at the lower irradiation temperature, Figure 8b. Voids were only observed at the highest irradiation temperature, 1170 K. As a consequence, the swelling was much less than in the unoxidized specimens. Some particle coarsening was also noted at 1170 K.

A model based on the radiation enhanced diffusion of zirconium in the alloy can explain the precipitate formation in the unoxidized alloys. To account for the amount of precipitation, however, some oxygen must have been picked up during the irradiation. The presence of these radiation induced precipitates demonstrates that phase stability in even simple alloys can be a complicating factor in understanding radiation damage mechanisms.

The presence of a fine dispersion of precipitates prior to irradiation does appear to retard void formation in the Nb-Zr alloy. One possible explanation is that the precipitate interface is serving as a neutral sink for both interstitials and vacancies.⁽⁶⁾

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CONCLUSION

A dual beam, ion irradiation facility for studying radiation damage problems applicable to fusion first walls has been described. Experiments on molybdenum have shown that co-implanted helium can alter the internal void microstructure during heavy ion irradiation but does not appreciably change the total swelling. Heavy ion irradiation of a Nb-1 Zr alloy have shown that irradiation can induce precipitate formation and that existing precipitates can influence the damage microstructure.

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- Fig. 1. Layout of dual beam ion irradiation facility at Pacific Northwest Laboratory
- Fig. 2. Target chamber for dual beam facility
- Fig. 3. Helium concentration as a function of depth for He^+ bombarded molybdenum
- Fig. 4. Void size and number density vs. dose for Ta^{+++} ion bombarded molybdenum. (a) no helium (b) simultaneous helium injection
- Fig. 5. Void-volume fraction (swelling) vs. dose for Ta^{+++} in bombarded molybdenum. (a) no helium (b) simultaneous helium injection.
- Fig. 6. Void volume fraction (swelling) vs. irradiation temperature for Ta^{+++} ion bombarded molybdenum, with and without simultaneous helium injection.
- Fig. 7. Void microstructure of molybdenum bombarded with Ta^{+++} ions at 1470°K to 40 dpa. (a) no helium (b) simultaneous helium injection
- Fig. 8. Microstructure of Nb-1Zr alloy bombarded with 5 meV Ni^{++} ions at 1170 K to 15 dpa. (a) no pre-internal oxidation (b) internally oxidized 1000 K for 32 hours.

Fig. 1

PNL ION BOMBARDMENT CHAMBER

Fig. 3

4A

4B

7A

7B

8A

8B