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Summary

The effect of interstitial gas atoms on cavity formation in self-ion irradiated nickel specimens is studied utilizing a 700-kV accelerator to inject H^+ or He^+ ions into nickel foils. A tandem accelerator then irradiates the foils with 14-MeV nickel to produce displacement damage. Details of the irradiation facilities are presented. Sample preparation and analysis techniques are described. Preliminary results and plans for future experiments are described.

Introduction

The development of engineering materials for utilization in irradiation environments, such as those present in fusion and fast breeder reactors, poses great problems for the reactor design engineer. Such irradiation environments produce high energy neutrons which can cause damage to crystalline metals by the production of displacement cascades, introduction of gas atoms into the metal matrix by transmutation reactions such as (n,p) and (n,α) and/or the initiation of phase changes in alloys. This damage can alter the mechanical properties of the material and will probably dictate the useful lifetime of the reactor.

Since few intense sources of 14-MeV neutrons for materials research currently exist,[1] studies using electrostatic accelerators have long been used to model the effects of neutron irradiation environments.[2-5] Accelerators may also be used to model the effects of transmutational gas evolution by utilizing dual-ion irradiation[3-10] or by preinjection of light ions.[6-10] These studies can be compared to the available 14-MeV and fission neutron irradiation data[9] to help predict microstructural evolution in fusion reactor materials.

The use of accelerators for irradiation studies has some shortcomings, however. Accelerators can create displacement damage only within a very small volume of material, usually a few micrometers below the incident surface. The high damage rates required to model long term irradiation conditions in a reasonable time period alter the temperature dependence of microstructure development as predicted by Bullough and Perrin.[11] The addition of excess interstitial atoms during a self-ion irradiation can affect the nucleation rate of cavities[12] thus altering the microstructure observed in the peak damage regions at the end of range of the incident ion. Accelerator irradiation studies must take these factors into account.

In this paper, we describe the irradiation facilities used to preinject hydrogen or helium gas atoms into nickel specimens. These specimens are then irradiated with 14-MeV nickel ions to produce displacement damage. We prepare the irradiated samples for Transmission Electron Microscopy (TEM) examination utilizing a cross-section technique. Results of preliminary irradiations for specimens with and without interstitial hydrogen are presented.

Experimental Facilities and Techniques

Light Ion Irradiation Facility

The hydrogen and helium gas atom preinjections are completed using the University of Wisconsin Nuclear Engineering Light Ion Accelerator Facility. This facility consists of a 700-kV electrostatic accelerator, beam focussing and analyzing equipment, vacuum system and a specimen holder-heater. A schematic of the accelerator facility, shown in Figure 1, indicates the associated pumping and beam handling components of the system.

An emittance measuring device[13] which continuously samples the phase space area occupied by the beam produced the results summarized in Table I. The emittance

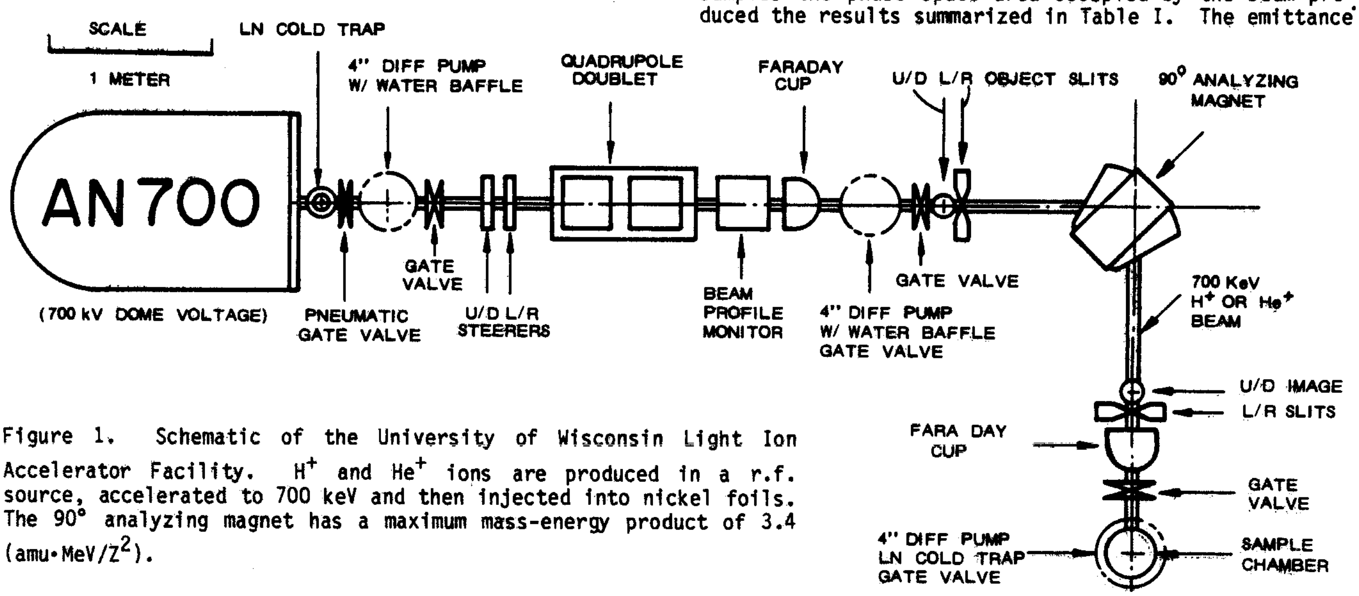

Figure 1. Schematic of the University of Wisconsin Light Ion Accelerator Facility. H^+ and He^+ ions are produced in a r.f. source, accelerated to 700 keV and then injected into nickel foils. The 90° analyzing magnet has a maximum mass-energy product of 3.4 (amu·MeV/Z^2).

Table I. Summary of AN 700 Accelerator Emittance Characteristics

Dome Voltage (kV)	Ion	Beam Current (μA)	Emittance (90% Contour) (π mm mrad MeV$^{1/2}$)
700	H^+	5	2.62
500	H^+	4	2.81
200	H^+	4	6.32
700	He^+	5	3.92
500	He^+	8	4.19

for the contour containing 90% of the beam decreases as the accelerating potential increases for both H^+ and He^+ beams. This increase is expected since higher accelerating voltages reduce space-charge effects. We used the measured emittances in transport calculations through beam handling components of the accelerator system.

The three major beam handling components include up-down and left-right steerers, a magnetic quadrupole doublet and a 90° analyzing magnet. The steerers are an in-house design while the quadrupole doublet and analyzing magnet were manufactured by Magnion, Inc. and ANAC, Inc. respectively.

The sample holder accommodates three 3-mm discs and three 0.5-cm x 1.0-cm foils. The holder electrically isolates the sample and masks the irradiated area to allow for accurate measurement of the incident beam current. A 10-keV electron current from a biased tungsten filament heats the sample holder which in turn radiatively heats the isolated sample to temperatures in the range of 100-800°C. Chromel-alumel thermocuples measure the temperature behind each sample position.

Heavy Ion Irradiation Facility

We irradiate the specimen with 14-MeV nickel ions from a model EN tandem accelerator. Figure 2 is a schematic of the tandem accelerator facility which is equipped with a sputter-type negative-ion source.[17-19] The specimen holder assembly [19] consists of a carousel with eight individual sample holders. The carousel arrangement allows individual heating of samples during irradiation. Chromel-alumel thermocouples in thermal contact with each specimen allow constant temperature monitoring during irradiation. The range of operating temperatures for this sample holder-heater assembly is roughly 200-700°C.

Specimen Preparation and Analysis

An important aspect of this study is the sample preparation technique that allows examination[20,21] of the samples in cross section. This technique provides a great deal of information from each sample since both dose and dose rate are depth dependent.

Before irradiation we anneal the pure nickel foils for one hour at 1000°C in a high purity argon atmosphere. The specimens must have extremely clean surfaces so that metal may be plated to them following irradiation. To this end we first grind the samples on abrasive paper and then mechanically polish them using a 3-μm alumina slurry. A final electropolish in a solution of 60% H_2SO_4 and 40% H_2O at -20°C produces a mirror-like surface.

To limit surface oxidation following irradiation we store specimens in alcohol until we begin the electroplating process. Up to 1.5 mm of nickel is

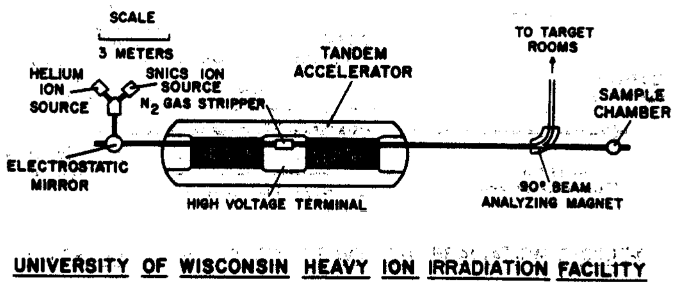

Figure 2. Schematic of the University of Wisconsin Tandem Accelerator Facility. Negative nickel ions from the sputter source are injected into the accelerator. In this study, 14-MeV Ni^{3+} ions are used to irradiate nickel foils in the sample chamber.

plated onto each side of the foil as described by Whitley.[14] We then mount the plated samples in epoxy and cut them with a diamond saw into slices normal to the original foil surface. A twin jet electropolishing device thins the standard 3-mm TEM discs that we punch from these slices. The depth distribution of the voids is determined by division of the micrographs into regions of uniform thickness parallel to the irradiated surface. Void size is determined using a Zeiss particle counter. We measure the TEM foil thickness using stereo microscopy techniques which allow the calculation of the void number density and the void volume fraction.

Results and Future Experiments

Whitley studied the effect of interstitial hydrogen atoms on the depth dependent microstructure in heavy-ion irradiated nickel.[14] The micrographs in Figure 3 show the void density as a function of depth in the sample. The ion beam entered the foil from the left. Figure 3a shows copious void formation at the peak damage region in a specimen that contained hydrogen introduced during the electropolishing phase of sample preparation. A thoroughly outgassed sample which was heated to 900°C in an ultra-high vacuum environment prior to heavy-ion irradiation (Figure 3b) shows a much lower void density than the non-outgassed sample. Figure 4 shows a difference of two orders of magnitude in the void density between a thoroughly outgassed sample and a non-outgassed sample. For these specimens the interstitial hydrogen enhances nucleation by stabilizing the void embroyos.

The introduction of a stabilizing gas atom via a sample preparation technique such as electropolishing offers only qualitative information. The quantity of gas which diffuses into the metal matrix is not readily calculable since the partial pressure of the gas at the metal-electrolyte surface is not known. Quantitative results are possible when one injects the sample with a known amount of interstitial gas (hydrogen or helium) prior to heavy-ion irradiation.

The Light Ion Accelerator Facility has recently been equipped with a 90° analyzing magnet. Mass analysis allows us to separate the H^+ or He^+ beams from any contaminant beams and to distinguish between the hydrogen ion H^+ beam and the molecular hydrogen H_2^+ beam. We have observed beam currents of 2.5-3.5 μA on a 3 mm

Figure 3. TEM micrographs comparing the void microstructure in heavy-ion irradiated nickel specimens with (a) and without (b) interstitial hydrogen (from Whitley[14]).

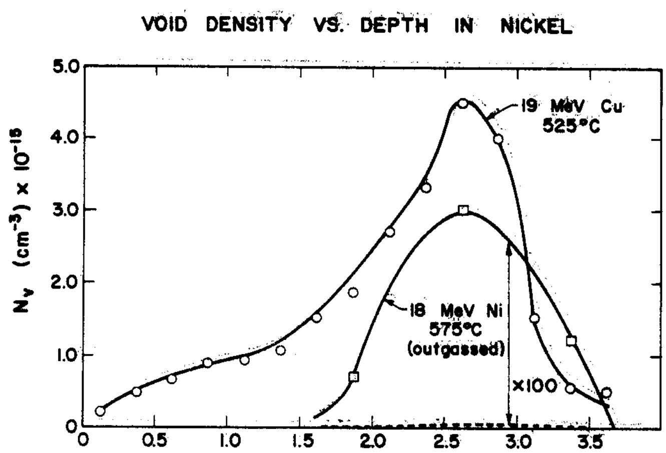

Figure 4. Void number density vs. depth for heavy-ion irradiated nickel with and without interstitial hydrogen (from Whitley[14]).

target for both H^+ and He^+ beams. These readings are equivalent to fluxes of 2.2-3.1 x 10^{14} ions/cm^2-sec.

Other future applications of the Light Ion Accelerator Facility include the study of near-surface damage in H^+ and He^+ irradiated nickel, nickel-aluminum, nickel-silicon and 316 stainless steel alloys. These same alloys will also be preinjected with hydrogen and helium gas atoms and subsequently irradiated with heavy ions to study the effects of interstitial gas atoms on the phase stability of these alloys under irradiation. Cross-section sample preparation techniques similar to those described above are now under development for the alloys proposed for study.

Acknowledgment

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