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DEPTH DEPENDENT VOID SWELLING RATES IN SELF-ION IRRADIATED NICKEL*

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Specimens of high purity nickel were irradiated with high energy heavy ions and the resultant microstructure examined along the ion path using a cross sectioning sample preparation technique. Void nucleation was found to be very sensitive to hydrogen introduced into the samples by electropolishing prior to irradiation. Samples containing hydrogen had void densities of about two orders of magnitude greater than outgassed samples. A series of hydrogen doped samples were irradiated with 14 MeV nickel ions to fluences from 2 x 10^{15} ions/cm^2 to 1.4 x 10^{17} ions/cm^2 (peak damage from 2 to 150 dpa). The increase in the collision cross section as the incident ion slows down causes an increase in displacement rate with depth. The variation in the observed void density, void size and void swelling rates with depth indicate the importance of displacement rate on void simulation studies.

1. INTRODUCTION

The first walls of Controlled Thermonulcear Reactors (CTR) will be subject to displacement damage and high generation rates of helium from (n,α) type reactions and hydrogen from (n,p) reactions. Hydrogen isotopes will also be introduced into the first wall by interactions with both the plasma and residual hydrogen in the system. Since helium is well-known to aid void nucleation, [1] the present study was performed to investigate the sensitivity of void formation to hydrogen concentration. In the first part of this study, specimens of high purity nickel were injected with hydrogen by electropolishing prior to irradiation. The specimens were subsequently irradiated at 525°C with 14 MeV nickel or copper ions. The resultant microstructure was then compared to that of a thoroughly outgassed specimen.

The post-irradiation analysis of the specimens was performed using a technique which allows the depth dependence of the microstructure to be examined directly. [2,3,4] Under self-ion irradiation, both the damage rate and the total damage level vary by approximately a factor of ten along the ion range. To study this displacement rate effect, a series of nickel specimens were irradiated at 525°C with nickel ions at fluences ranging from 2 x 10^{15} to 1.4 x 10^{17} ions/cm^2 (peak dose from 2 to 150 dpa) and the void swelling rates were measured as a function of depth. By examining the structure in cross section, the effects of variation in displacement rate, along with the variation in other features such as PKA spectrum could be studied or observed.

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2. EXPERIMENTAL TECHNIQUE

High purity (>99.995%) nickel samples were irradiated with high energy heavy ions accelerated in a Tandem Van de Graaff accelerator. The depth dependent displacement rates were calculated using the code of Brice [5] with the results for 14 MeV nickel incident on nickel shown in Figure 1. The details of the accel-

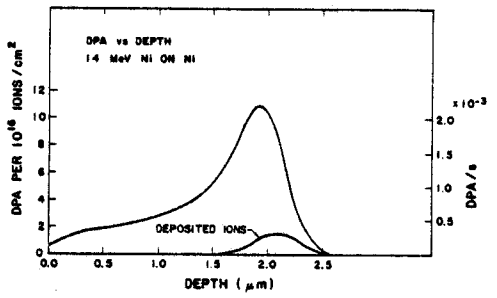

Figure 1 - Damage profile as calculated from Brice Code. [5]

erator facilities and of the high temperature ultrahigh vacuum target chamber are described elsewhere. [6] The nickel specimens in the form of 1 x 1.2 x 0.2 cm strips were annealed in either an inert gas atmosphere or an ultrahigh vacuum (<10^{-6} Pa) furnace for 1 hr. at 850°C. To introduce hydrogen into the specimens and to clean the surfaces prior to irradiation, the samples were electropolished in a solution of 60% H_2SO_4 and 40% water at room temperature. In some samples, the electropolishing was followed by an anneal at 900°C for one hour in an ultrahigh vacuum high temperature furnace to provide gas free samples for comparison to those doped with hydrogen. The post-irradiation

examination technique, which is described in detail in references [2] and [3], involved electroplating ~1.5 mm of nickel onto each side of the irradiated foil and then slicing the specimen in cross-section using a low speed diamond saw.

3. RESULTS AND DISCUSSION

3.1 Outgassing studies

In this study, the introduction of hydrogen by electropolishing was found to dramatically enhance the void nucleation rate.

This paper reports on irradiations using copper and nickel ions. Similar work using aluminum and carbon ions has been reported elsewhere. [2,4] In all cases, the void microstructures consisted of high (~10^{15}/cm^3) void densities with few dislocation loops observed. In the outgassed foils, however, the structure consisted mainly of small unfaulted loops with a few very large voids present near the ion end-of-range. The loop density increased with depth reaching a peak density of approximately 10^{15}/cm^3 with loop diameters of about 40 nm. Typical depth dependent microstructures from an electropolished and an outgassed specimen are shown in Figure 2. The foil surface is visible on the left-hand-side of the micrograph with the incident ions having traveled from left to right and come to rest near the right-hand-side of the micrographs.

The void densities measured for an electropolished and an outgassed foil are shown in Figure 3 (note the change of scale of the outgassed curve). While the outgassed foils had void densities over two orders of magnitude smaller than electropolished foils, the void diameter had increased from 20 nm in the electropolished foils to about 100 nm in the outgassed foils. This increase in void size led to similar swelling values in both cases, indicating that the hydrogen introduced by electropolishing was only aiding nucleation and not drastically altering the void swelling rate.

In this study, electropolishing was definitely identified as a sample preparation procedure that was introducing an agent into the samples capable of aiding void nucleation.

Electropolishing has been previously observed to enhance void formation [7] presumably by the introduction of hydrogen. Hydrogen induced deformation of thin foils has been observed after electropolishing at room temperature. [8] Other studies on thoroughly outgassed copper and nickel [9] have shown soluble gases to aid void nucleation. Electropolishing apparently charges samples with hydrogen [7] when the polishing is carried out in anything except very low temperature baths (i.e., <-50°C). At higher temperatures, the hydrogen diffuses into the sample faster than material is being removed and hence saturates the sample. Hydrogen would most probably enhance void nucleation by adsorbing onto the void surface and lowering the surface energy or by internal pressure of H_2.

To aid void nucleation, the hydrogen introduced into the samples during the electropolish must not diffuse out of the specimen at irradiation temperatures. However, calculations of hydrogen behavior in metals [10] have revealed hydrogen-vacancy binding energies of only about 0.2 eV. It was also found that molecular hydrogen would not form in the metal lattice. One way hydrogen could be kept in the samples during the heating to irradiation temperatures would be by trapping at other impurities. Carbon and oxygen are both present in small amounts and could form hydrogen compounds that would effectively trap the hydrogen. During irradiation, either the hydrogen would be freed from these traps and migrate to the void nuclei or the hydrogen compound would migrate to the void nuclei. At high temperatures (>900°C), the hydrogen would be freed from the traps and diffuse from the samples. If this mechanism is valid, then the amount of hydrogen available to aid nucleation will depend only on the impurity concentration and on the introduction of sufficient hydrogen to saturate the available trapping sites. The exact sample handling procedure and the details of how the hydrogen was introduced (such as the polishing time) will not be important. In this study, electropolishing was found to give reproducible void densities and was used in lieu of the more usual helium injection.

3.2 Results of 14 MeV nickel ion irradiation

To aid in interpreting the depth dependent void growth rates, a series of seven samples were irradiated at 525°C with 14 MeV nickel ions to fluences ranging from 2 x 10^{15} to 1.4 x 10^{17} ions/cm^2. The samples were prepared for irradiation by annealing for one hour at 850°C in an inert atmosphere and then electropolished. The development of the void structure as a function of ion fluence is shown in the micrographs of Figure 4. The original foil surface is visible near the left-hand-side of each micrograph with the incident ions having traveled from left to right.

Voids were observed throughout the damage region at all fluence levels (down to 0.4 dpa). There was a denuded region at the front surface of 100 to 150 nm, with the voids adjacent to this denuded region being unusually large at the lower fluences but of normal dimensions at the higher fluences. Voids were observed up to depths of about 3.2 μm. At the highest fluence level (150 dpa at the peak), the voids show signs of partial ordering. This ordering is the early stage of a void lattice formation which has been previously observed in nickel. [11] The void shapes were truncated octahedra except at the highest ion fluence, where the shapes were nearly cubic.

Figure 2 - Microstructure of Ni bombarded with 5×10^{15} ions cm^{-2} at 525°C with (a) and without (b) hydrogen in the matrix

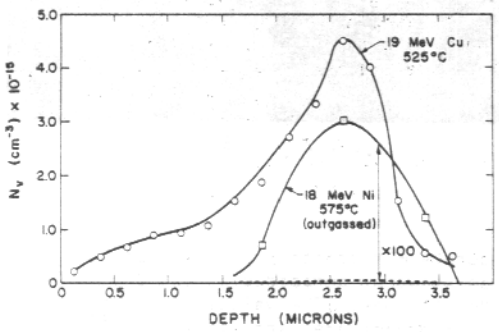

Figure 3 - Number density of voids from samples in Figure 2.

Void data was collected from these samples using 0.25 μm depth intervals and a set of curves such as the one shown in Figure 5 was obtained for each of the seven fluence levels. To assist in interpreting this data they are plotted in Figures 6-7 by taking data from three depth intervals in each sample and plotting the data against the dpa value of each point as determined from Figure 1 using the appropriate ion fluence. Hence, the set of data points from a given depth will represent data where the main irradiation variable is the total ion fluence (that is, data that are comparable with that taken by conventional sample preparation procedures). A comparison of data sets from different depths of the same sample will involve changes in dose rate, PKA spectrum, etc. From the void density curves of Figure 6, the void density is seen to saturate very early (<0.5 dpa). At a given dpa value, the void density increases with increasing depth, a result expected from the higher dose rate in these regions. The void size curve of Figure 7 also shows the expected behavior of increasing void size with increasing dose, and decreasing void size with increasing dose rate (i.e., depth). Void sizes are either constant or drop slightly above 20 to 50 dpa.

The swelling values from these specimens at the three depths are shown in Figure 8. In

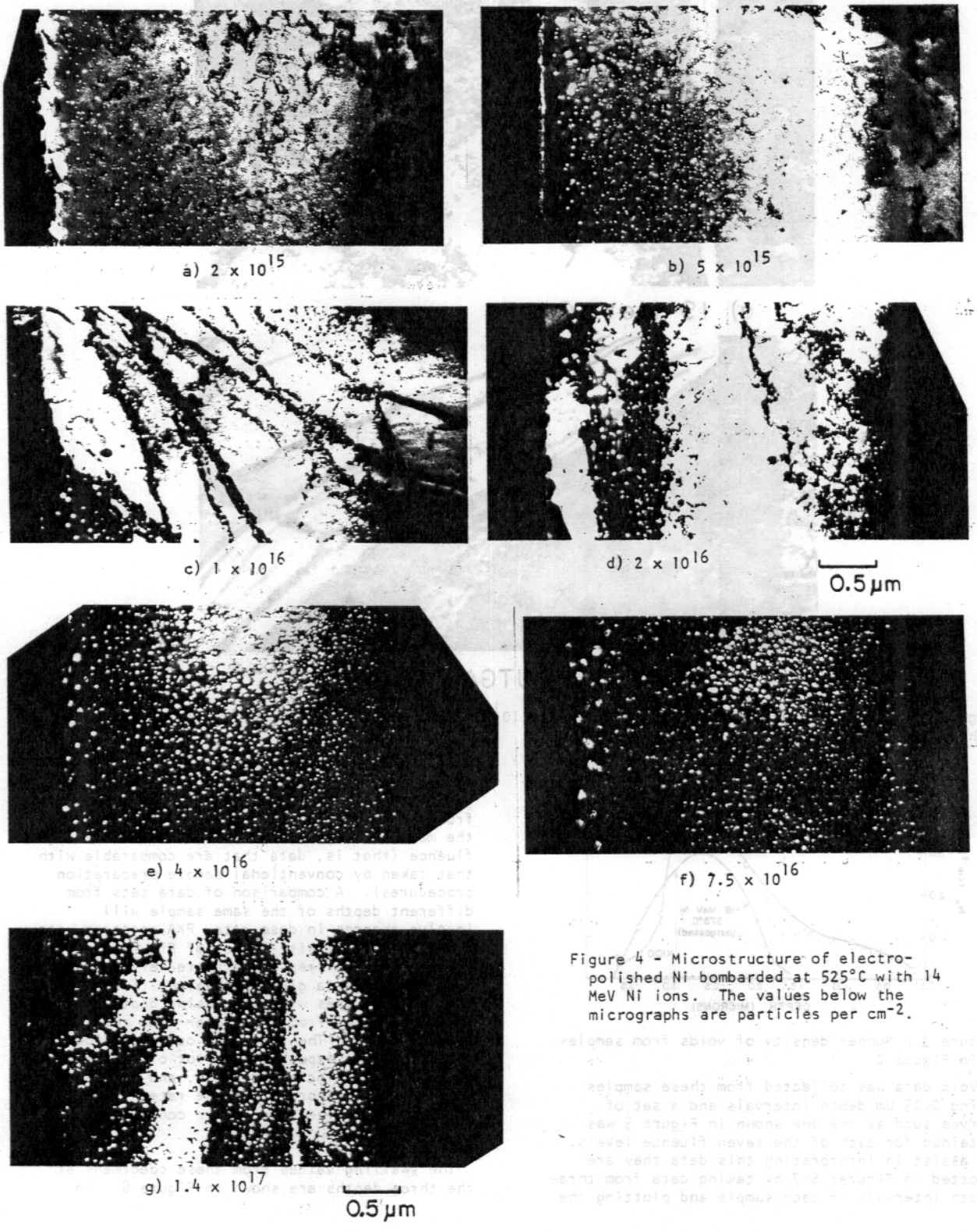

Figure 4 - Microstructure of electropolished Ni bombarded at 525°C with 14 MeV Ni ions. The values below the micrographs are particles per cm^{-2}.

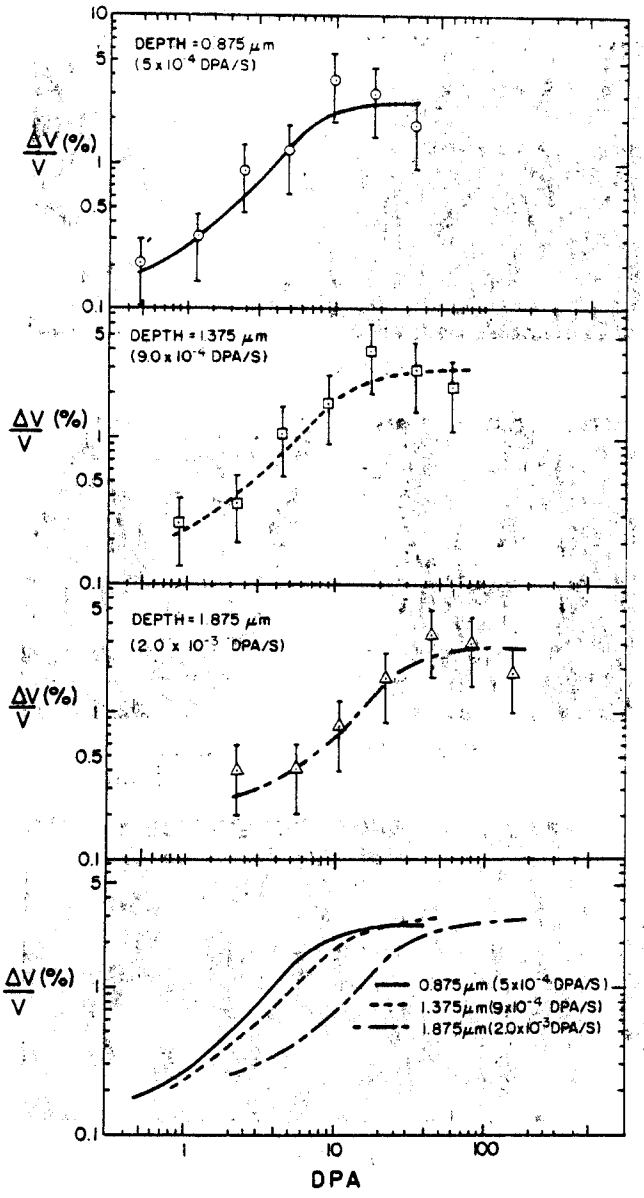

Figure 5 - Swelling data at different depths in Ni bombarded at 525°C with 14 MeV ions. (See Figure 4 for microstructure.)

general, the swelling curves for each depth increase approximately linearly with dose up to about 20 to 50 dpa, at which time the swelling levels off at a value of 3%. At a given dpa value, swelling decreases with increasing depth (i.e., increasing dose rate). In the growth regime at each depth, the swelling rates are roughly equal. The swelling value saturates at a larger dpa value for the other depths, but the final swelling values at saturation are about equal.

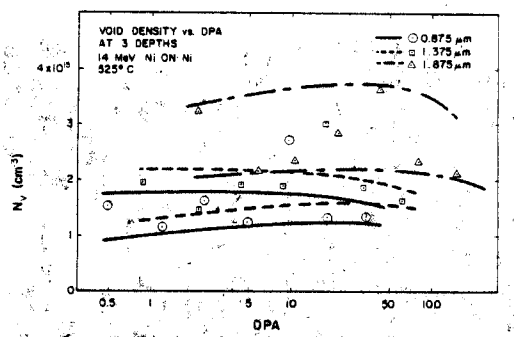

Figure 6 - Void density in Ni bombarded at 525°C with 14 MeV Ni ions. Note early saturation level.

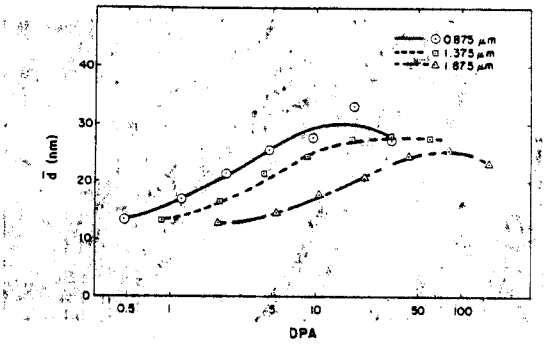

Figure 7 - Effect of damage on void size in Ni bombarded at 525°C with 14 MeV Ni ions.

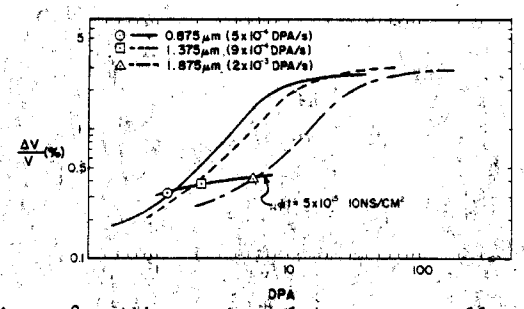

Figure 8 - Illustration of damage rate effect in Ni bombarded at 525°C with 14 MeV Ni ions. The data points come from the same sample but slightly different depths (damage rates).

Even small variations in dose rate with depth, less than a factor of two for example, are very important in modifying depth dependent behavior. The data of Figure 5 is replotted in Figure 8 to illustrate this point. The three data points are the swelling values at three depths within a single sample, namely the sample irradiated to a total ion fluence of 5×10^{15} ions/cm^2. The variation in swelling between these points with increasing dose is not indicative of the swelling rate expected by taking data from only one depth (i.e., one displacement rate) and increasing the total ion fluence. These results indicate that data taken from different depths in a sample do not give a valid representation of the swelling versus dose behavior.

4. CONCLUSIONS

1. In thoroughly outgassed nickel, the void nucleation rate was dramatically reduced up to 10 dpa with observed void densities of $\sim 10^{13}/cm^3$.

2. When sufficient hydrogen was introduced by electropolishing, void nucleation was enhanced with densities of $\sim 10^{15}/cm^3$ observed at 0.2 dpa.

3. At 525°C, the void density saturated at low dose (< 2 dpa). The saturation of the void size between 20 and 50 dpa caused the swelling to saturate at ~3%. At the highest doses, a void superlattice was beginning to form.

4. The damage level necessary for swelling to saturate increased with depth (most probably displacement rate).

5. The swelling value at saturation did not depend on depth (displacement rate).

6. One must be extremely careful when comparing data from many specimens, to always use that information obtained under identical damage conditions (i.e., displacement rate, PKA spectra, etc.). These conditions can change drastically over short distances in ion bombarded solids.

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