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EFFECT OF HYDROGEN ON VOID PRODUCTION IN NICKEL*

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Samples of high purity nickel were preinjected with hydrogen at 25 C and subsequently irradiated with 14-MeV nickel ions at 525 C. The resulting microstructure was examined by transmission electron microscopy analysis utilizing a transverse sectioning technique which allowed examination throughout the entire 3 micrometer range of the bombarding ion. It was found that approximately 1-10 appm of hydrogen had a major effect on the production of voids. The hydrogen increased the swelling by a factor of 5 at the 10 appm level over the entire 25 dpa range studied as compared to a similar hydrogen free sample. It is evident from this study that any nickel component in a fusion reactor which is exposed to hydrogen (e.g., through discharge cleaning) prior to operation of the reactor, will be very susceptible to enhanced swelling and possibly reduced mechanical performance.

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1. Introduction

The role of hydrogen as a nucleating agent for void formation in a fusion reactor environment has been largely ignored primarily due to concern with the effects of helium in a similar role. It has been assumed that due to the high diffusivity of hydrogen in most candidate first wall materials[1], the effect of hydrogen on the cavity nucleation process would be minimal. With the development of complex computer codes such as DIFFUSE [2] and other calculations which predict the tritium inventory, it has become possible to estimate the amount of hydrogen retained in the fusion reactor first wall.

Look and Baskes [3] have employed a Gas Driven Permeation (GDP) model to calculate the steady state hydrogen isotope concentration in an operating fusion reactor. A range of 1 to 10 appm retained hydrogen was predicted for a stainless steel first-wall. Recent calculations by Kerst and Swansiger[4] and experimental results by Causey, et al.[5] studying the effects of Plasma Driven Permeation (PDP) suggest the trapped hydrogen inventory may be double that produced by GDP. This implies a hydrogen concentration of up to 20 appm may be present in fusion reactor first-wall materials.

This study investigates the effect of interstitial hydrogen atoms on void nucleation by implanting nickel foils with hydrogen and subsequently irradiating with energetic nickel ions to produce displacement damage. The irradiated foils are prepared for transmission electron microscopy examination utilizing a transverse sectioning technique which permits evaluation over the entire 3 micrometer range of the irradiating ion. This technique also allows evaluation over a range of damage levels (7 to 27 dpa) at varying damage rates ($1\text{-}5\times10^{-3}$ dpa/s).

2. Experimental Technique

High purity (99.995%) polycrystalline nickel foils were annealed and metallographically polished, as described in detail elsewhere [6]. The foils were implanted with H_3^+ ions produced by an electrostatic accelerator. The implantation energies ranged from 200 to 700 keV. Initial implantations were completed at 700 kV followed by injection at decreasing 100 kV increments to 200 kV. This technique introduced the hydrogen at a relatively uniform concentration from a depth of about 1 micrometer to the foil surface. Hydrogen ion fluences of 3×10^{18} H_3^+/m^2, 1.5×10^{19} H_3^+/m^2, and 3×10^{19} H_3^+/m^2 represent injected concentrations of 100, 500, and 1000 appm assuming 100 percent trapping. The actual trapping efficiency is assumed to be on the order of 1 percent as described in a subsequent section of this report. Hence, it is expected these injected fluences represent retained concentrations of 1-10 appm. All hydrogen implantations were completed at 25 C.

Implanted samples were then irradiated with 14-MeV Ni ions to a fluence of 2.5×10^{20} ions/m^2 at 525 C. Theoretical calculations using the Brice code[7] have predicted a range of 2.5 micrometers for these irradiation conditions. The peak damage region at a depth of 2.5 micrometers is exposed to a dose of 27 dpa, while the damage level at a depth of 1 micrometer is about 7 dpa. Nominal vacuum during the hydrogen implantations and nickel ion irradiations was 1.0×10^{-5} Pa. Irradiated samples were prepared for TEM evaluation using the transverse sectioning technique described in detail by Whitley [8]. The samples were examined using a JEOL 100B transmission electron microscope. The depth distribution of voids was determined by division of the micrograph into regions of thickness 0.25 micrometer which were parallel to the irradiated surface. The void size was determined using a Zeiss particle analyzer and the foil thickness was determined by stereo microscopy techniques.

3. Results

The microstructure observed for nickel foils irradiated with nickel ions to a fluence of 2.5×10^{20} ions/m^2 at 525 C with varying preinjected hydrogen fluences is shown in Figure 1. An increase in the void number density with increasing injected hydrogen concentration is readily observed all throughout the entire damage region. However, to simplify this presentation, the data presented in Figures 2-4 were obtained at two depths (1 micrometer and 2.5 micrometer). This represents damage levels of 7 dpa and 27 dpa and damage rates of 1×10^{-3} dpa/s and 5×10^{-3} dpa/s, respectively.

The most dramatic effect of injected hydrogen concentration variation is noted in the marked decrease in the mean void diameter as shown in Figure 2. With no hydrogen injection the mean void diameter is approximately 70 nm. The injection of even the smallest amount of hydrogen (100 appm injected, estimated 1 appm retained) reduces the mean void diameter to about 28 nm. This mean void diameter remains relatively constant with increasing hydrogen injection up to 1000 appm injected. It should also be noted that there is little variation in the mean void diameter with increasing damage levels. Since the mean void diameter throughout the entire damage region (not just the 0 to 1 micrometer injected region) shows little variation, it might be suggested that there exists some trapping of hydrogen throughout the sample and not just in the damage region produced by the preinjection.

The variation in void number density as a function of injected hydrogen concentration is shown in Figure 3. An increase in void number density is noted for increasing injected hydrogen concentration from 0 to 500 appm (estimated 0-5 appm retained). A saturation in void number density appears when the injected hydrogen concentration reaches a level of 500 appm (5 appm retained). This suggests the beginning of swelling saturation.

The effect of the variation of hydrogen content on swelling in these samples is shown in Figure 4. This figure shows an increase in swelling associated with increasing hydrogen concentrations. The top curve shows the variation in swelling noted at the peak damage region (27 dpa), while the bottom curve shows the variation in swelling at a depth of 1 micrometer (7 dpa). Also note that as the hydrogen concentration increases, the rate of swelling is not linear but appears to be saturating. This saturation was also suggested by the void number density data.

4. Discussion

The high diffusivity of hydrogen in nickel poses questions about the amount of hydrogen which is retained following a preinjection at 25 C as performed in this study. Recent experimental results by Bessenbacher, et al.,[9-10] indicate that the fraction of hydrogen retained in nickel following implantation at an energy of 10-keV was related to the number of defects present in the material. These experiments indicate the fraction of implanted hydrogen retained ranged between 1 and 10 percent at 25 C. Similar experiments and theoretical considerations have been completed by Wilson, et al., [11-12] in 316 stainless steel. Wilson predamaged steel samples with 300-keV He ions and irradiated with 1-10 keV deuterium ions. These experiments produced results which suggest a 1 percent trapping of hydrogen implanted at 25 C. The present study employed hydrogen implantation energies over the range 200-700 keV for a H_3^+ ion beam (67-233 keV/H). Based on the previously published experimental data, this study will assume a trapping efficiency of 1 percent. This assumption yields a retained hydrogen concentration of 1, 5 and 10 appm for injected hydrogen concentrations of 100, 500 and 1000 appm. This represents a conservative estimate considering the higher implantation energies employed in this work. These higher implantation energies result in deeper hydrogen implantation and higher damage levels associated with the implantation.

A comparison of swelling versus displacement damage for various injected hydrogen concentrations is presented in Figure 5. The increase in swelling with increasing hydrogen concentration is again evident. The sample injected with 100 appm hydrogen (1 appm retained) shows twice the swelling of a sample with no hydrogen injection. The sample injected with 500 appm hydrogen (5 appm retained) exhibits swelling about 3.5 times greater than the no hydrogen case. The 1000 appm hydrogen injected sample (10 appm retained) shows the greatest increase with a swelling level 5 times greater than a similar sample with no injected hydrogen.

The saturation in swelling with increasing damage for each hydrogen concentration is also shown in Figure 5. However, another factor must be considered when evaluating this saturation. The damage rate increases with increasing damage level since both are depth dependent. This damage rate variation may also contribute to the observed saturation in swelling.

A similar saturation in swelling has been noted in other heavy-ion irradiations[8,13-15] and has been attributed to damage rate effects.

The effect of hydrogen on cavity nucleation in an irradiation environment has very serious ramifications to operating fusion reactors. The introduction of interstitial hydrogen into first-wall and structural materials by GDP (dissociation and diffusion) or by PDP (energetic implantation) will provide sites for the nucleation of cavities at the onset of displacement damage production. This early nucleation may greatly enhance the overall swelling rate and thus reduce the lifetime of first-wall and structural components.

5. Conclusions

The irradiated nickel foils displayed an increase in swelling with increasing injected hydrogen concentration. The sample with a retained hydrogen concentration of 10 appm exhibited swelling 5 times greater than a similar sample with no hydrogen injection. All hydrogen injected samples had almost uniform mean void diameter throughout the entire 2.5 micrometer damage region even though the hydrogen was only injected to a depth of 1 micrometer. This suggests migration of hydrogen to traps throughout the sample rather than trapping only in the preinjection damage zone.

A tendency toward saturation of swelling with increasing hydrogen concentration occurred as the injected hydrogen concentration approached 1000 appm (10 appm retained). A saturation of swelling above a damage level of 10 dpa was also observed in all samples. This saturation may be attributed to higher damage rates at the these damage levels.

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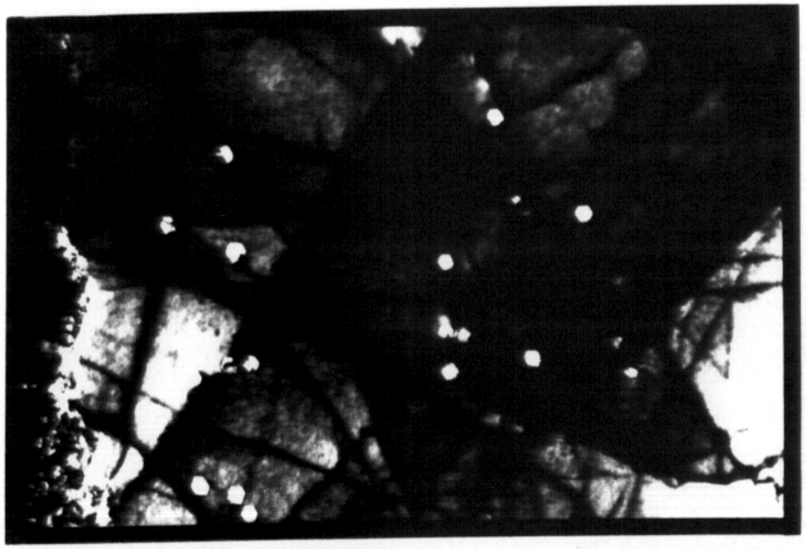
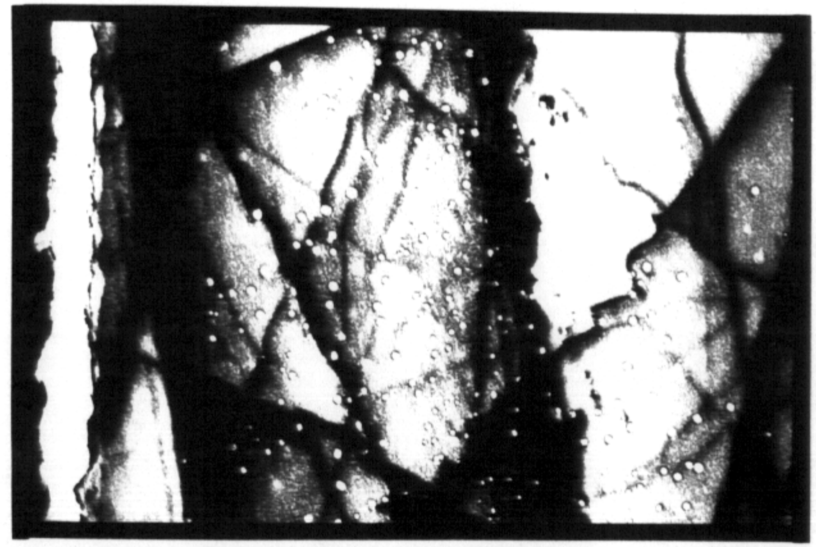
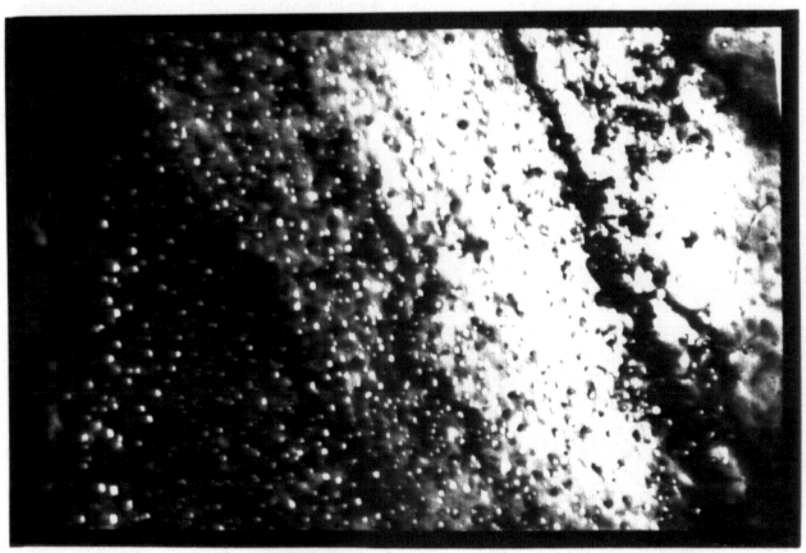
VARIATION IN VOID MICROSTRUCTURE WITH INCREASING HYDROGEN PREINJECTION AT 525 C



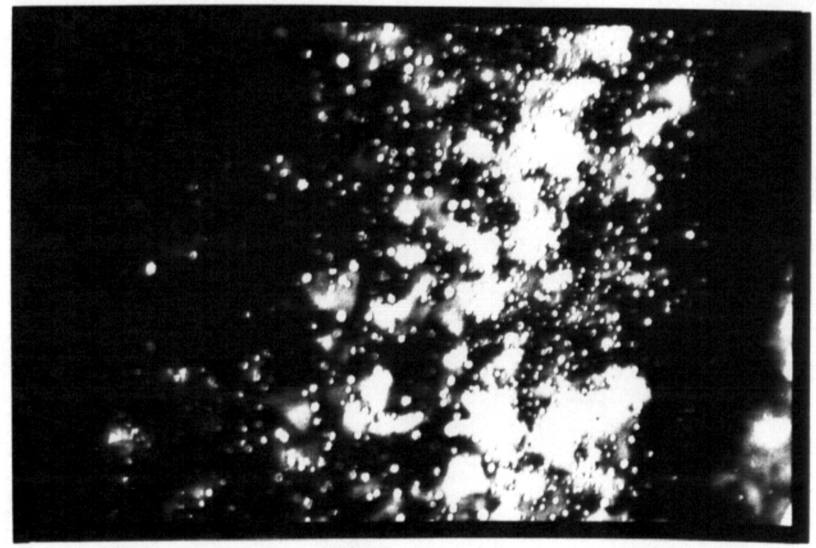
a) No gas preinjection



b) 100 appm H ($3x10^{18}$ H_3/m^2)



c) 500 appm H ($1.5x10^{19}$ H_3/m^2)



d) 1000 appm H ($3x10^{19}$ H_3/m^2)

0.5μm

Figure 1. Void microstructure at various injected hydrogen concentrations for nickel irradiated with nickel ions to a fluence of $2.5x10^{20}$ ions/m^2 at 525 C.

EFFECT OF HYDROGEN CONCENTRATION ON VOID DIAMETER IN PURE NICKEL

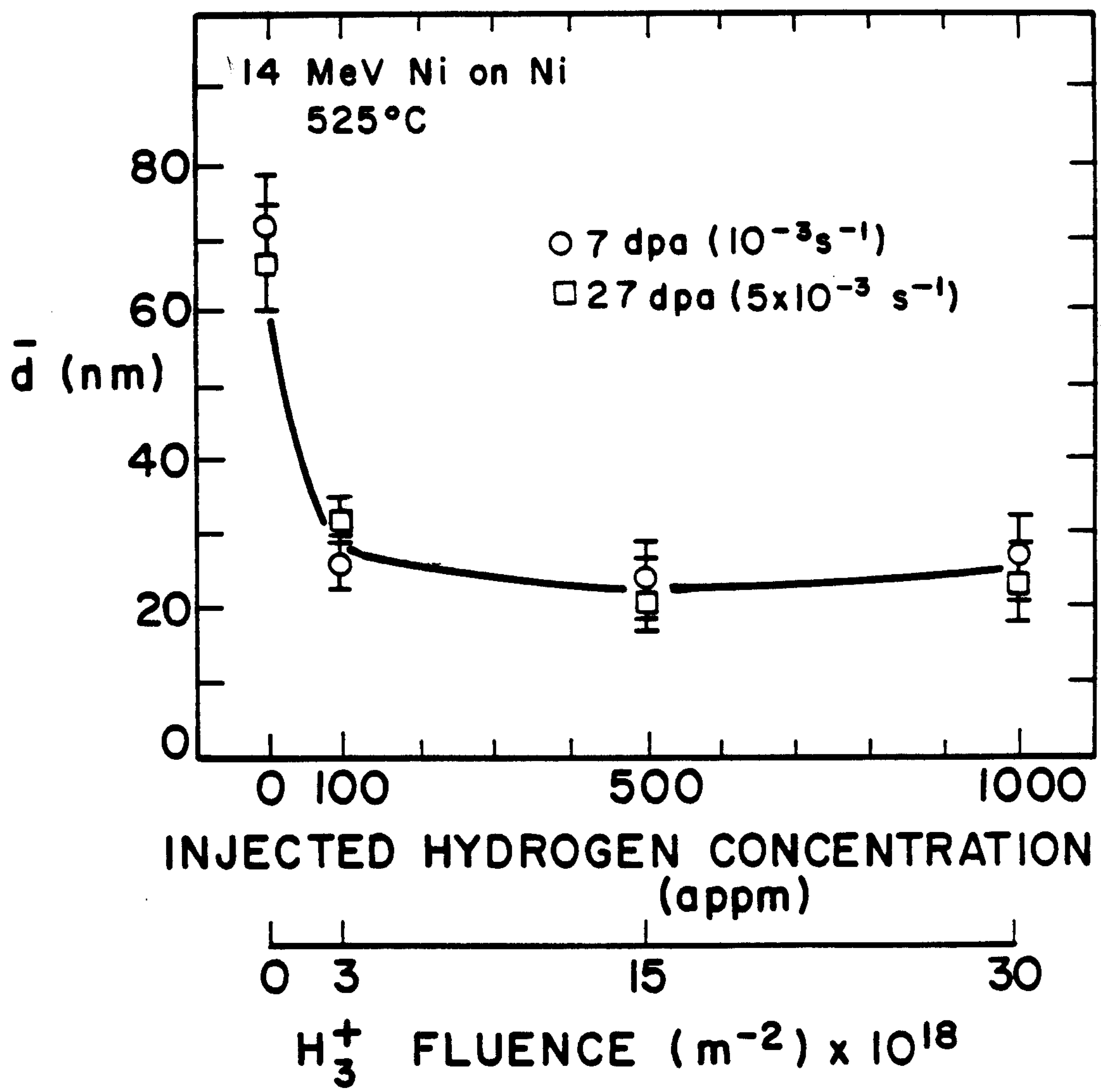


Figure 2. Mean void diameter vs. injected hydrogen concentration for nickel irradiated to a fluence of $2.5x10^{20}$ ions/m^2 at 525 C.

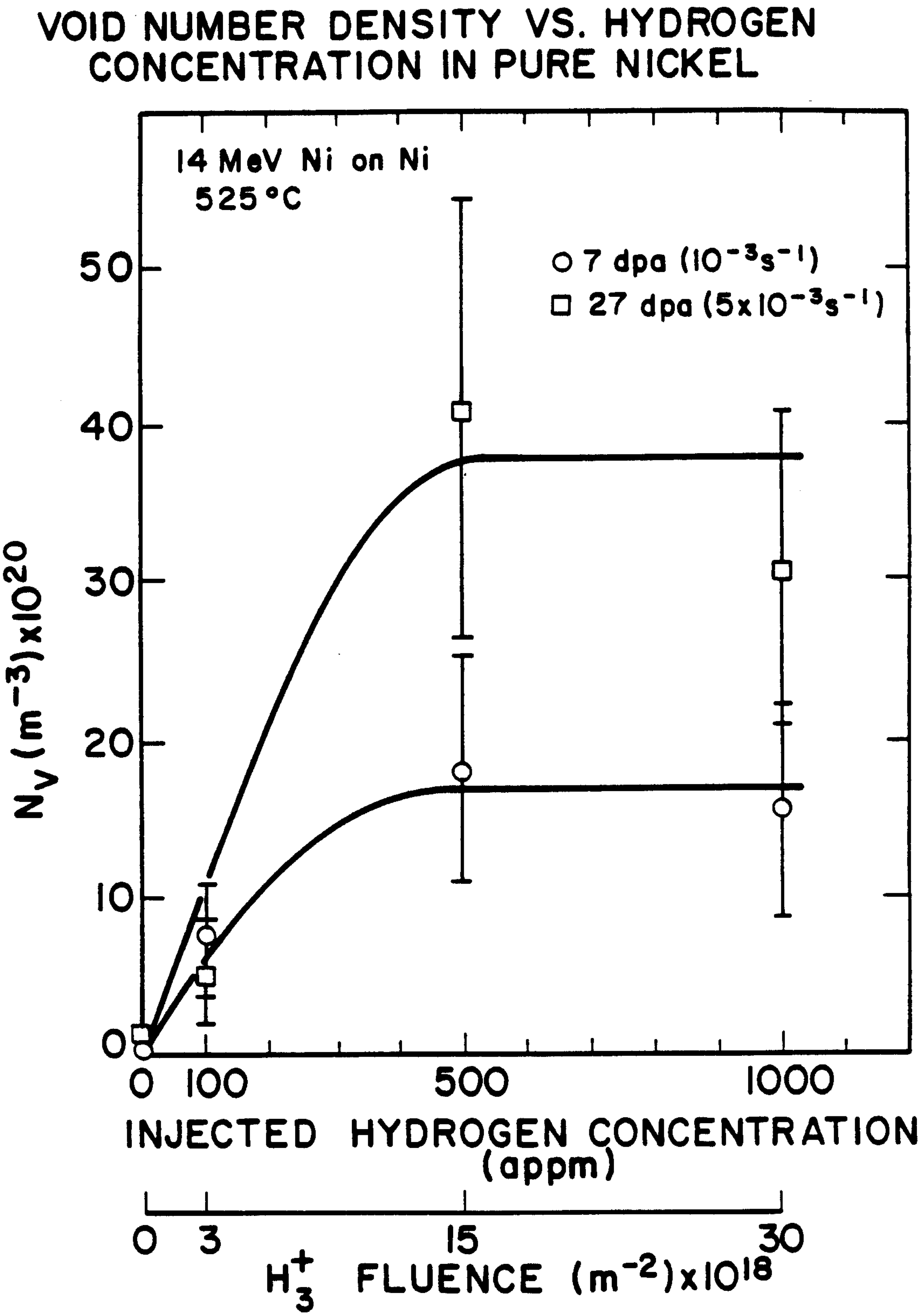


Figure 3. Void number density vs. injected hydrogen concentration for nickel irradiated to a fluence of 2.5×10^{20} ions/m^2 at 525 C.

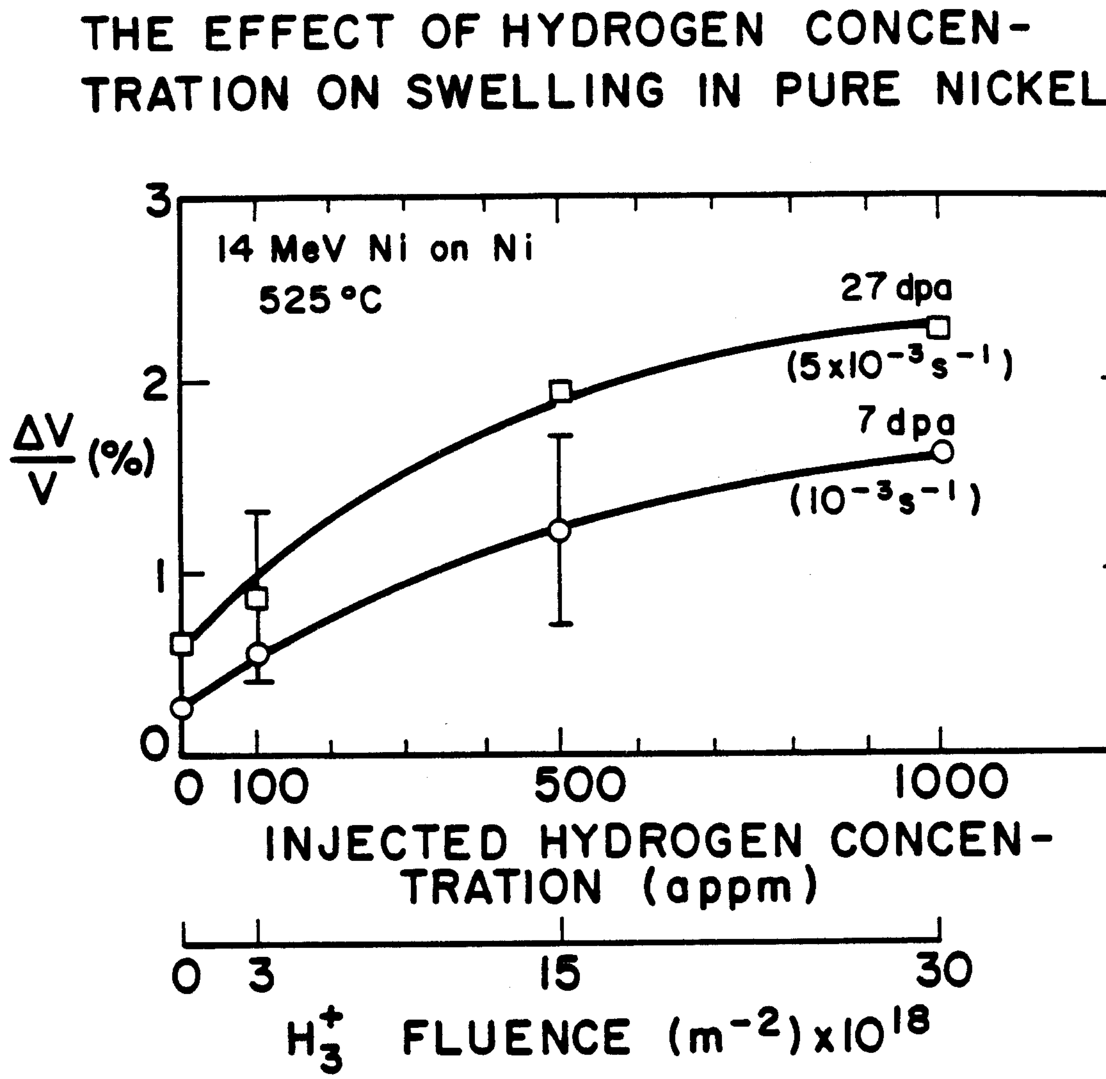


Figure 4. Swelling vs. injected hydrogen concentration for nickel irradiated to a fluence of $2.5x10^{20}$ ions/m^2 at 525 C.

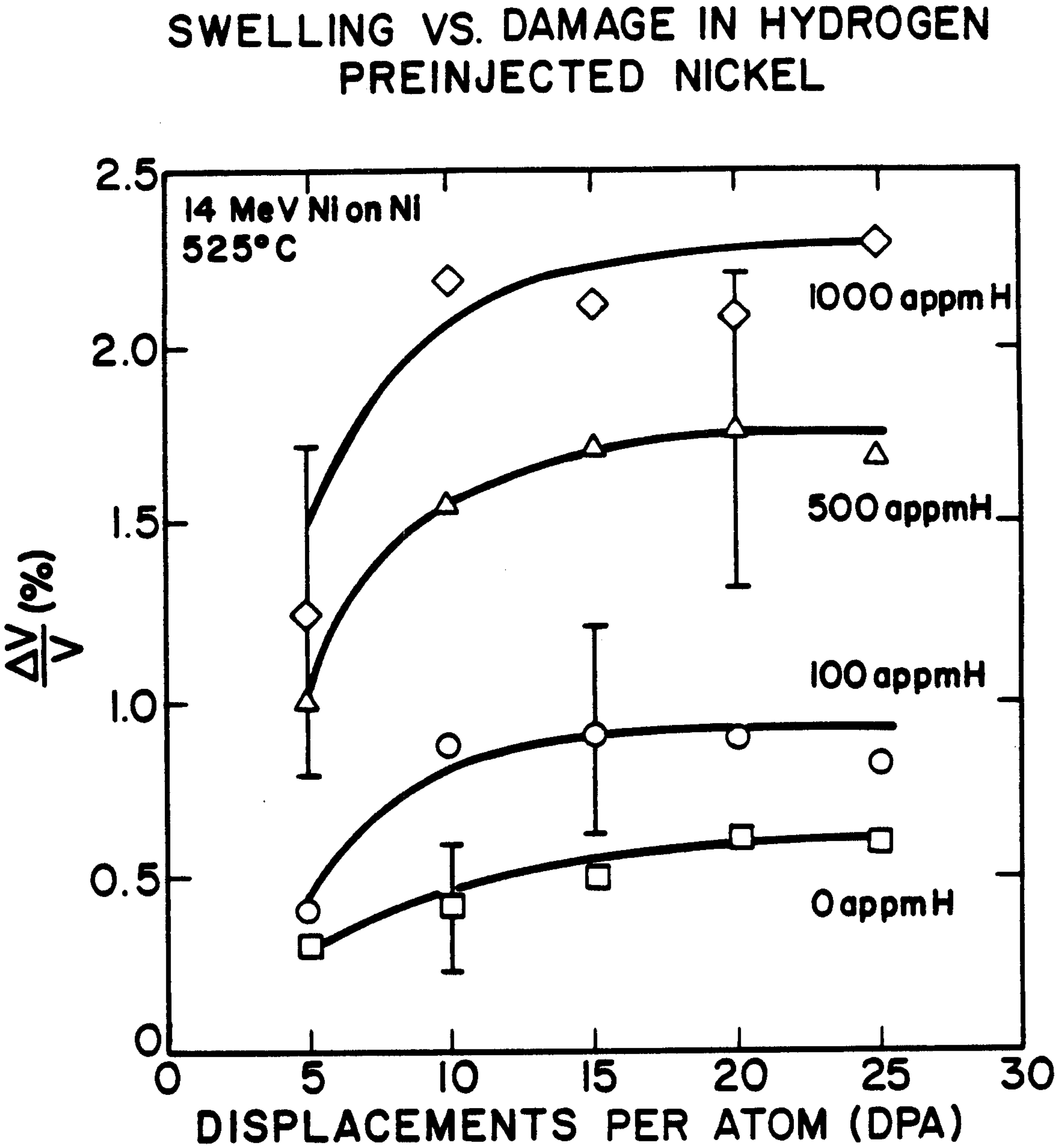


Figure 5. Swelling vs. damage for various injected hydrogen concentrations in nickel irradiated to a fluence of 2.5×10^{20} ions/m^2 at 525 C.