Gas Effects on Void Formation in 14 MeV Nickel Ion Irradiated Pure Nickel

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UWFDM-680

Presented at the 2nd International Conference on Fusion Reactor Materials, 14-17 April 1986, Chicago, IL.

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Fusion Technology Institute
University of Wisconsin
1500 Engineering Drive
Madison, WI 53706

http://fti.neep.wisc.edu

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Fusion Technology Institute, Nuclear Engineering Department
University of Wisconsin-Madison, 1500 Johnson Drive
Madison, WI 53706-1687 USA

Abstract

Nickel samples with various helium and oxygen concentrations have been irradiated with 14 MeV Ni ions at 500°C to a fluence of 8 x 10^{19} ions/m^2 (2 dpa at a 1-μm depth). Helium atoms with energy varying from 200 to 700 keV were pre-injected at room temperature. The samples with low oxygen content were obtained by a hydrogen reduction treatment and high vacuum outgassing. The density and the average diameter of voids were determined by TEM examination of cross-section specimens.

In the as-received Ni which was cold-worked, a heterogeneous void distribution was observed that could be attributed to the heterogeneous oxygen distribution. In the specimen irradiated after vacuum annealing at 800°C for one hour, voids are uniformly distributed.

Small amounts of helium (10 appm) enhanced the void nucleation remarkably in both high (180 appm) and low (75 appm) oxygen content samples, while larger amounts of helium (30 appm) reduced the observable void density in the high oxygen content sample. The void density is much lower and the void size is much larger in the low oxygen content samples compared to those observed in the high oxygen content samples.

1. Introduction

Nickel has long been selected as a model material in radiation-induced void swelling studies to better understand the void formation mechanism, and, therefore, the complex analysis problems imposed by phase changes are avoided [1-3]. The effect of helium, produced either by the transmutation reaction in neutron irradiated Ni or by pre- or co-implantion in ion irradiations of pure Ni, has been studied extensively [4-6]. However, the effect of oxygen on void formation in irradiated pure Ni has been neglected. Oxygen is the most common residual gas in metals and has been shown to promote void formation both by theoretical analysis [7,8] and by some experiments on other materials [9-13]. Therefore, it seems that any quantitative approach of gas effects on void formation would be incomplete without considering the effect of residual oxygen in the material. In the present experiments, we have investigated the effects of both helium and oxygen on void formation in ion irradiated pure Ni.

2. Experimental Procedure

The Ni used in this study was Marz grade (99.995 wt.% pure) foil from the Materials Research Corporation. Foil samples with 1 cm x 0.5 cm dimensions were mechanically polished with 0.3 μm alumina abrasive. Six samples with different pre-irradiation treatments were irradiated with 14-MeV Ni^{3+} ions at the University of Wisconsin Heavy-Ion Irradiation Facility. One sample was irradiated in the as-received state (cold worked), while three were annealed at 800°C for 3.6×10^3 s in a vacuum of 4×10^{-5} Pa to remove the cold-worked structure. Two of the annealed samples were then injected with either 10 appm or 30 appm helium before Ni-ion irradiation. The remaining two samples (with a thickness of about 0.25 mm) were first heated at 1000°C in flowing dry hydrogen for 4.3×10^4 s to reduce the oxygen content, and then annealed at

150°C in a vacuum of 6.6×10^{-7} Pa for 1.8×10^3 s to remove residual hydrogen. Analyses performed by Los Alamos National Laboratory, using a vacuum fusion technique, indicated that the oxygen content in the Ni foil was reduced from the original 180 appm to 75 appm after such treatment. One foil with reduced oxygen content was also pre-injected with 10 appm helium before Ni-ion irradiation.

The helium atoms with energy varying from 200 to 700 keV were pre-injected in the Ni samples at room temperature using the University of Wisconsin 700 kV Accelerator. This produced a zone with relatively uniform helium concentration extending from the sample surface to a depth of about 1 μm, which is separated from the injected interstitials introduced during the 14 MeV Ni self-ion irradiation.

All the samples were finally irradiated at 500°C with a flux of 3×10^{16} $Ni^{3+}/m^2/s$ to a fluence of 8×10^{19} Ni^{3+}/m^2. The displacement damage as a function of depth for 14 MeV Ni ions on pure Ni was calculated using the Brice code [14] and is plotted in Fig. 1. According to this calculation, Ni-ion irradiation in our study would produce a displacement level of 2 dpa at 1 μm or about 8 dpa at the damage peak. It should be noted that a displacement efficiency factor of k = 0.8 was used in the displacement damage calculation to be consistent with previous ion bombardment studies, even though k = 0.3 is probably a more appropriate factor for neutron or ion irradiations [15,16].

After irradiation, the specimens were prepared in cross-section [3] for observation by Transmission Electron Microscopy (TEM), which allows the entire damage region to be studied in one single foil. TEM analysis was performed on a JEOL 100B and a JEOL TEMSCAN-200CX electron microscope.

3. Results and Discussion

After irradiation, a heterogeneous void distribution was observed in the cold-worked (as-received) Ni containing 180 appm oxygen, as can be seen from Figs. 2 and 3. In Fig. 3 (a), it can be seen that voids formed preferentially around a very occasional impurity particle. An Energy Dispersive X-ray Spectroscopy (EDXS) analysis on the particle showed only the presence of Ni (elements with $Z < 10$ can not be detected by EDXS). It is believed that the particle was an oxide. If that is the case, then the high void density around the particle might be attributed to the higher local free oxygen content because of the re-solution of Ni oxide during irradiation. Voids were also found to form preferentially on dislocations and along grain boundaries as shown in Figs. 3 (b) and (c), where the oxygen concentration could also be higher than in the bulk material. The average void density was determined to be 6×10^{19} m^{-3} at the depth of 1 μm and 2×10^{20} m^{-3} at the damage peak. The corresponding average void diameter was 35 nm at the 1-μm depth and 30 nm at the damage peak.

Figure 4 shows the entire damage region of the three annealed specimens which contain 180 appm oxygen and varying levels of helium in the first micrometer region. The variation of void density and average diameter with depth is shown in Fig. 5. It is clear from Fig. 4 (a) that the heterogeneous void distribution in the irradiated, cold-worked Ni (Figs. 2 and 3) is not a feature of irradiated, annealed Ni specimens. Also, compared to the cold-worked specimen, the void density is much higher, and voids are much smaller in diameter in the annealed material. This change might be explained by homogeneous distribution of oxygen atoms due to thermal diffusion during annealing.

The effect of pre-injected helium in the annealed, high oxygen content (180 appm) Ni is shown by comparison of the micrographs and curves presented in Figs. 4 and 5, respectively. Pre-injection of 10 appm helium enhanced the void nucleation remarkably, while pre-injection of 30 appm helium reduced the void density. Compared to the sample without helium pre-injection, pre-injection of 10 appm helium increased the void density at 1 μm (2 dpa) by about three times, but pre-injection of 30 appm helium reduced the void density by a factor of 2.6. The suppression of void formation by pre-injected helium was previously reported for higher helium levels than used in this study [6], and the suppression was considered to be the result of copious nucleation of sub-microscopic cavities in the implanted region. To verify this explanation, the region pre-injected with 30 appm helium was observed very carefully at a magnification of 200,000X for sub-microscopic cavities in addition to the voids. Sub-microscopic cavities were not resolved. Another possibility is that large levels of pre-injected helium tend to suppress void nucleation by increasing the vacancy-interstitial recombination rate, because helium has a high possibility of trapping a vacancy [17] and these trapped vacancies function as recombination sites when their density is high.

The variation of the void distribution and the dependence of the void parameters along the depth in two irradiated low-oxygen content (75 appm) specimens are shown in Fig. 6 and Fig. 7, respectively. The sample shown in Fig. 6 (b) had ~ 10 appm helium injected in the first micrometer region. For the purpose of comparison, the void parameter curves for the sample with high oxygen content (180 appm) and zero helium is also presented in Fig. 7. Also, it should be noted that the void density is drawn on a logarithmic scale in Fig. 7 (a) because the void density in the low oxygen content samples is at least one order of magnitude lower than that in the high oxygen sample except in the helium pre-injected region. On the other hand, the average void size in the low oxygen content samples is much greater. In the helium pre-injected region we have a high density of small diameter voids, but, compared to the 10 appm helium region in the sample with high oxygen content (Fig. 5), the void density in the same region of the low oxygen content sample is much lower and the average void size is larger. Another notable difference is that toward the end of the helium range (1 μm) in the low oxygen sample, the voids get progressively larger and the void density becomes progressively smaller. There is a void denuded zone in the range of 1-2 μm because fewer gas atoms are available for void nucleation. This phenomenon was not seen in the high oxygen content sample that was pre-injected with the same amount of helium because there were oxygen atoms available to stabilize the void embryos.

In addition to the results shown above, Ni samples with different intrinsic oxygen contents and with pre-injected oxygen have also been irradiated to higher fluences. The results, which confirmed the observations of this study, are being presented separately [18].

4. Conclusions

The effects of residual oxygen and pre-injected helium on void formation in pure Ni have been studied by cross-section technique following 14 MeV Ni ion irradiation to a fluence of 8 x 10^{19} ions/m^2 at 500°C. The following conclusions can be drawn:

(a) The residual oxygen plays an important role in promoting void formation in ion irradiated, pure Ni. Lowering the oxygen content from 180 appm to 75 appm reduces the void density and increases void size remarkably.

(b) Voids tend to form in regions with high dissolved oxygen contents. The heterogeneity in void distribution observed in cold-worked Ni could be attributed mainly to the heterogeneous oxygen distribution.

(c) Pre-injection of a small amount (10 appm) of helium enhances the void nucleation significantly in both high (180 appm) and low (75 appm) oxygen content Ni, but the void density in the low oxygen content sample is still much lower compared to that in the high oxygen content sample after the same amount of helium pre-injection.

(d) Pre-injection of a relatively large amount (30 appm) of helium reduced the observable void density in the Ni foil containing 180 appm oxygen.

(e) The experimental results in this study support the theoretical model which predicts that gas is necessary for voids to form in Ni [8,18].

Acknowledgment

The authors wish to thank L.E. Seitzman for his many valuable discussions, D.J. Pertzborn for performing the helium pre-injection, R.C. Schmidt and other members of the radiation damage group at the University of Wisconsin for their assistance during Ni-ion irradiations. This research is supported by the U.S. Department of Energy, Office of Fusion Energy.

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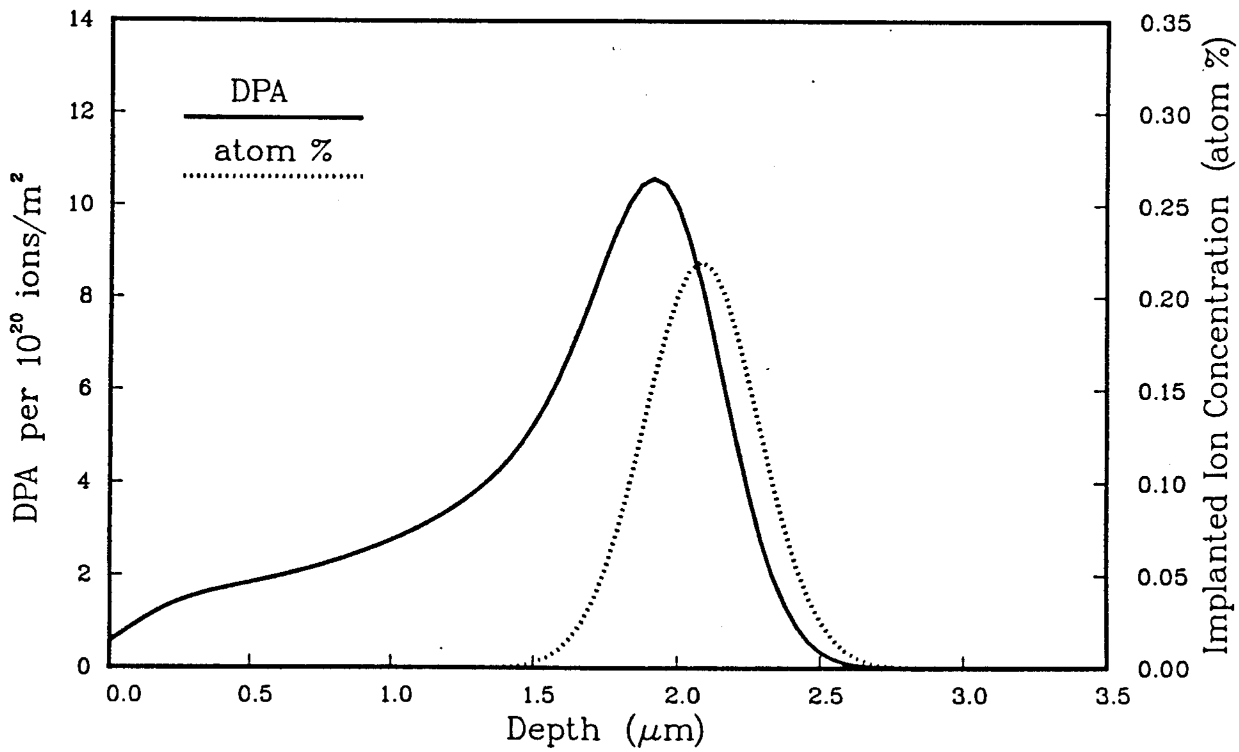


Fig. 1. Displacement damage and implanted ion concentration versus depth from the irradiated surface for 14 MeV Ni-ion irradiated pure Ni calculated using the Brice code. The damage efficiency (k) used is 0.8.

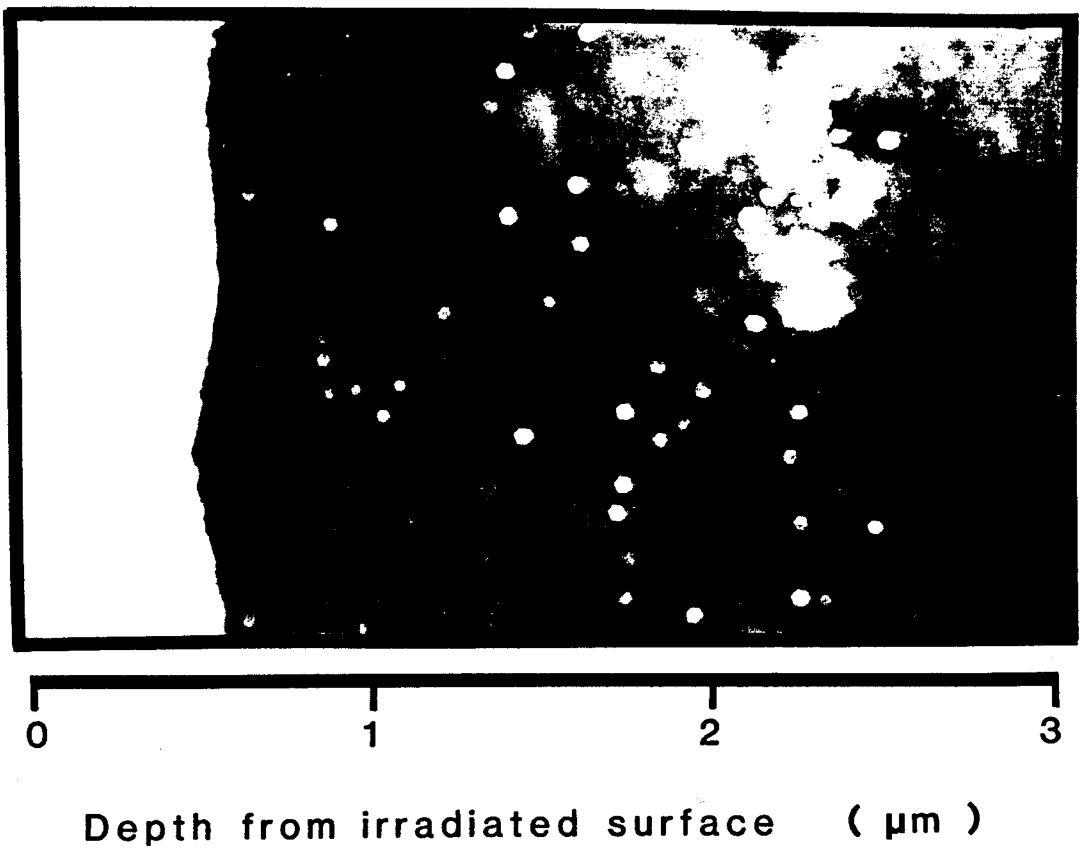


Fig. 2. TEM micrograph which shows the cross-section of 14 MeV Ni-ion irradiated, cold-worked pure Ni (the first half micron was lost during sample preparation). The oxygen content is 180 appm in the sample.

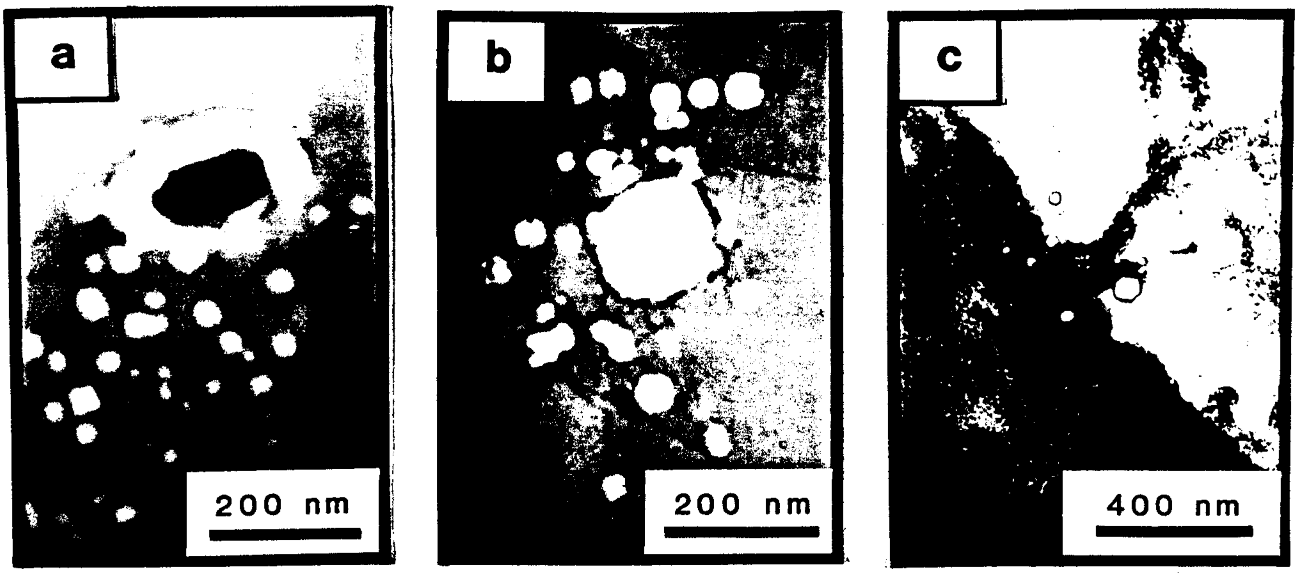


Fig. 3. Heterogeneous void distribution in 14 MeV Ni-ion irradiated cold-worked pure Ni containing 180 appm oxygen. Voids formed preferentially around an impurity particle (a), on dislocations (b), and along the grain boundary (c).

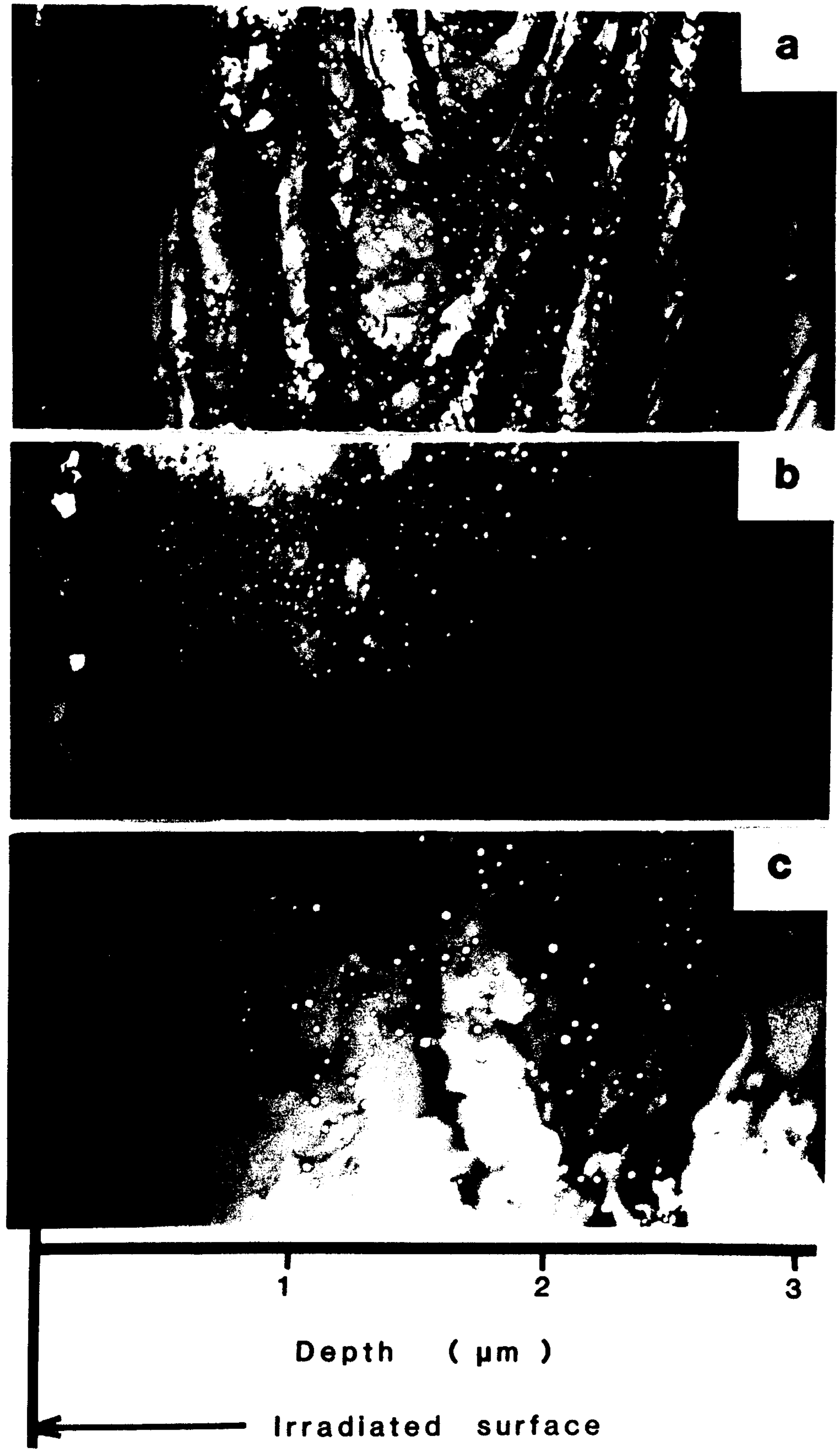
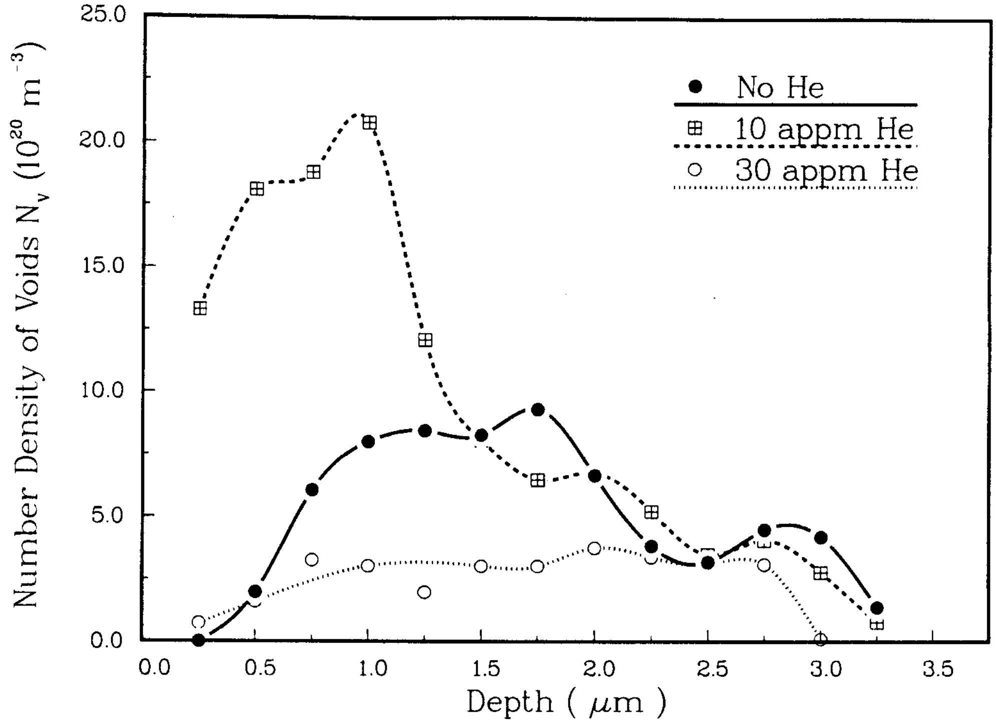
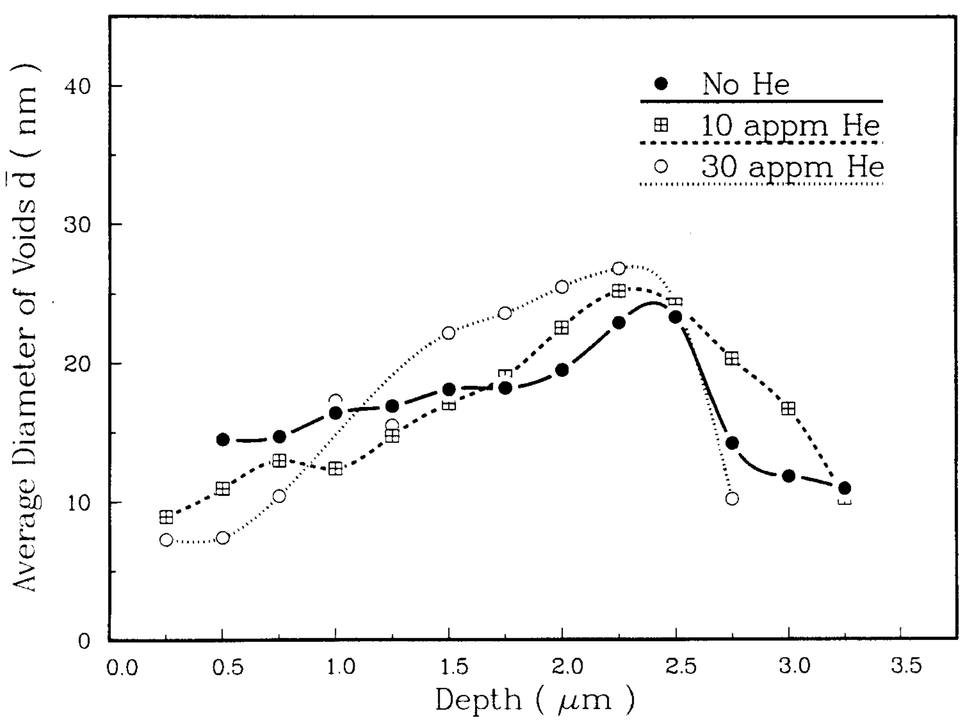


Fig. 4. Cross-section TEM micrographs which span the entire damage region of 14 MeV Ni-ion irradiated pure Ni containing 180 appm oxygen. The samples were annealed at 800°C, 4×10^{-5} Pa for one hour before helium pre-injection and Ni irradiation.
(a) Without helium pre-injection;
(b) With 10 appm helium pre-injected in the first micrometer;
(c) With 30 appm helium pre-injected in the first micrometer.



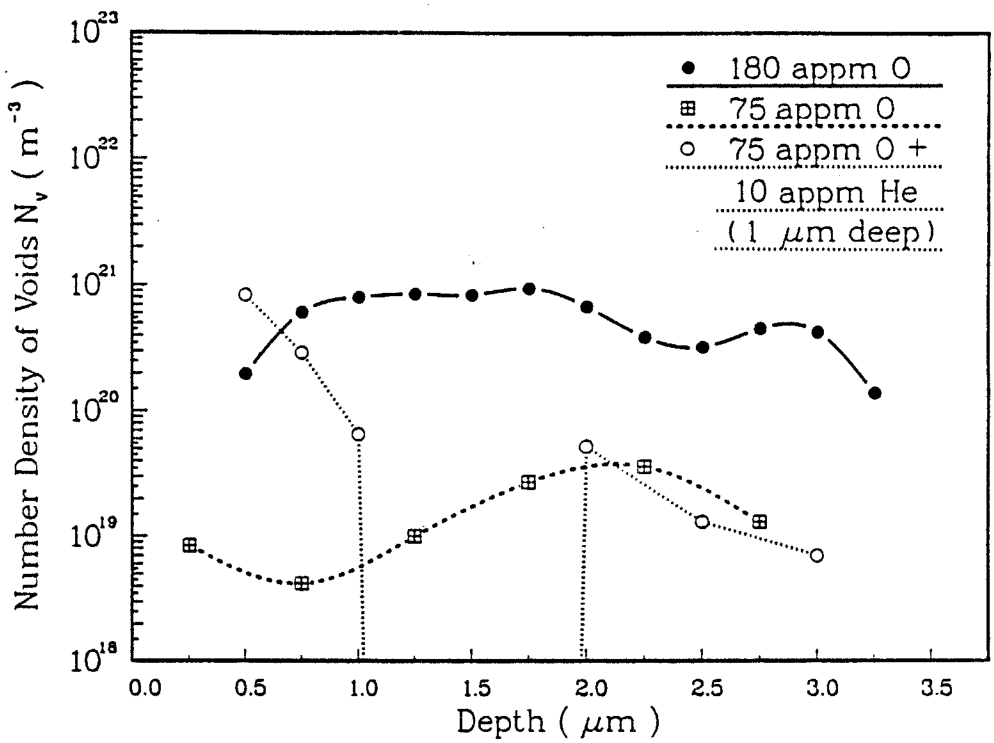
(a)



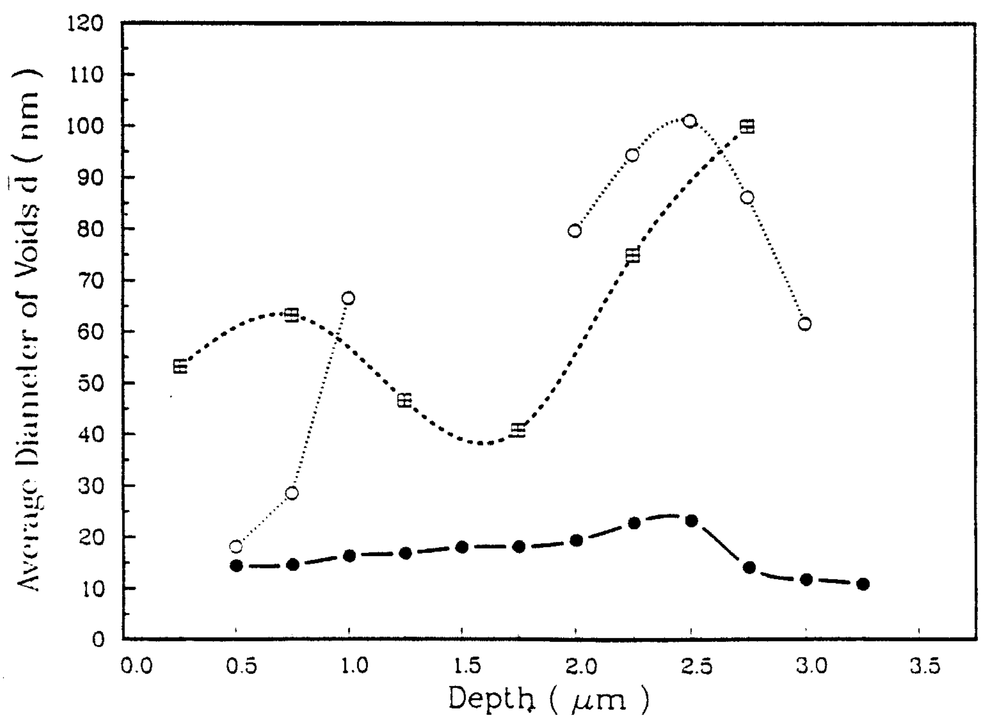
(b)

Fig. 5. Number density of voids (a) and average diameter of voids (b) versus depth for the irradiated high oxygen content (180 appm) samples as shown in Fig. 4.

Fig. 6. Cross-section TEM micrographs of 14 MeV Ni-ion irradiated low oxygen content (75 appm) pure Ni samples without (a) or with (b) 10 appm helium pre-injected in the first micrometer region.



(a)



(b)

Fig. 7. Number density of voids (a) and average diameter of voids (b) versus depth for the irradiated low oxygen content (75 appm) samples with or without 10 appm helium pre-injected in the first micrometer region. The curve for the high oxygen content (180 appm) zero helium sample shown in Fig. 5 is repeated here for comparison.