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ABSTRACT

14 MeV Cu ions have been implanted into a pure Ni specimen at 500°C to a dose of 6 x 10^{20} ions/m^2. TEM and AEM analyses were performed in cross section to investigate the effect of implanted Cu on the formation of defect clusters. The TEM result has been compared with that obtained in another Ni specimen which was irradiated with 14 MeV Ni ions to the same damage level at the same temperature. While voids formed throughout the entire damage range in the Ni ion irradiated sample, they mainly appeared at the near surface region and at the peak damage depth in the Cu ion implanted specimen. A high density of dislocation loops formed in the region where implanted Cu ions were detected by AEM. The AEM result of the implanted Cu concentration profile has been compared with a Monte Carlo calculation.

INTRODUCTION

Materials in future D-T fusion reactors will be subject to intense displacement damage from high energy fusion neutrons. The excess vacancy and interstitial concentrations produced by irradiation will result in the formation of voids and/or dislocation loops that will greatly alter the material performance. Heavy ion irradiation, which can give a displacement rate several orders higher than that presently available in neutron irradiations, has proven to be a very useful tool in the study of radiation induced defect cluster formation for fusion reactor material research [1].

Previous studies have revealed that Ni-Cu alloys are very resistant to void formation [2-7], although voids do form readily in both pure Ni and pure Cu. In the present study, the effect of a small concentration of implanted Cu on void formation has been studied by performing both transmission electron microscopy (TEM) and analytical electron microscopy (AEM) on a 14 MeV Cu ion implanted (irradiated) Ni cross section specimen. Since both the displacement damage and the Cu concentration resulting from the Cu ion implantation are depth dependent, TEM and AEM analyses in cross section permit a direct correlation of the microstructural and microchemical evolutions for the entire damage range within the same specimen.

EXPERIMENTAL PROCEDURE

Marz grade (99.995 wt.% pure) Ni foil from the Material Research Corporation was used in this study. Two foils with 1 x 0.5 x 0.025 cm dimensions were mechanically polished with 0.05 μm alumina abrasive. They were first heated at 1000°C in flowing dry hydrogen for 4.3 x 10^4 s to reduce the oxygen content, and then annealed at 150°C in a vacuum of 6.6 x 10^{-7} Pa for 1.8 x 10^3 s to remove residual hydrogen since it has been shown that both oxygen and hydrogen greatly promote void formation in Ni [8-10]. Analyses performed by the Los Alamos National Laboratory indicated that the oxygen content in the Ni foil was reduced from the original 180 appm to 75 appm after such treatment. The samples were irradiated with either 14 MeV Cu^{3+} or 14 MeV Ni^{3+} ions at the University of Wisconsin Heavy-Ion Irradiation Facility at 500°C with a dose rate of 1.5 x 10^{16} ions/m^2 s. The sample irradiated with Cu ions received a total dose of 6 x 10^{20} ions/m^2 and that irradiated with Ni ions received a total dose of 6.5 x 10^{20} ions/m^2, so that the damage level at 1 μm depth equals 25 dpa (~ 75 dpa at peak damage depth) in both samples according to the Monte Carlo calculations performed using the TAMIX code [11]. The depth profiles of displacement damage and ion concentration, calculated for 14 MeV Cu ion irradiated Ni at the ion dose of 10^{20} ions/m^2, are shown in Figure 1.

After irradiation, the samples were electroplated with Ni on both sides and prepared in cross section for TEM and AEM analyses. The details of the sample preparation procedure have been reported earlier [12,13]. The cross sectioned samples were then examined in a JEM 200CX II TEMSCAN microscope operating at 200 keV. The radiation induced defect clusters in both samples were observed in TEM mode. The implanted Cu concentration in the Cu ion irradiated sample was studied by AEM in STEM mode with a beam spot size of < 20 nm using a TN-2000 energy dispersive X-ray spectroscopy (EDXS) system. During the AEM analysis, a series of points lying on a line normal to the interface between the plated and the irradiated Ni, i.e. parallel to the direction of incident ions and covering the entire damage range, was analyzed to provide the depth profile of the implanted Cu concentration. To prevent the interference of Cu signals from the brass sample holder, a graphite holder was used during the AEM study; this appeared to be very effective.

RESULTS AND DISCUSSION

TEM Observations

While voids were the only significant defects observed in the Ni ion irradiated specimen, and they formed throughout the entire damage range, both voids and high densities of dislocation loops were seen in the Cu ion irradiated specimen, with the voids mainly located both at the near surface region and at a depth which is about 2.75 to 3 μm from the irradiated surface. The TEM cross section micrographs which show the entire damage region for the two specimens are shown as Figure 2 and Figure 3 respectively. It should be noted that a surface layer ~ 0.3 μm thick was removed before Ni plating in the process of cross section sample preparation to assure good bonding at the interface, so the actual depth from the original irradiated surface is ~ 0.3 μm deeper than the depth from the interface indicated in the two cross section micrographs.

The density and average diameter of the voids and swelling in both samples, as well as the dislocation loop density in the Cu ion irradiated specimen, have been plotted against the depth from the irradiated surface (the ~ 0.3 μm removed layer was added), and they are shown in Figure 4. To reduce the uncertainties in the swelling calculation, the morphology of voids was studied by viewing them with the electron beam close to several main zone axes, as shown in Figure 5. The voids in both samples were determined to be bounded by {100} and {111} faces (cubic truncated by octahedra), with truncation parameters [14] between 0.5 and 0.6. The void dimensions in the <110> direction and the appropriate volume factor [14] were used in the determination of void volume. The average diameter reported in Figure 4(b) is the diameter of the sphere which has the equivalent volume as the average void volume. The dislocation loops observed in the Cu ion irradiated specimen are mostly perfect loops with the Burgers vector of a/2 <110>. Figure 6 shows the typical dislocation loop images at various depths in the Cu ion irradiated sample. At both the surface and at the end of damage range, some large loops (~ 50 nm in diameter) were observed. In between, a high density of smaller loops (≤ 10 nm) was found. The analysis of the interstitial/vacancy nature of the dislocation loops has only been performed on some of the larger loops (≥ 50 nm) using the technique outline by Edington [15]. Both interstitial and vacancy loops have been identified.

From Figure 4, it is very clear that there is a void suppressed region, which extends from the depth of 1 μm to the depth of about 3 μm, in the Cu ion irradiated specimen. Also in that region, high densities of dislocation loops are formed. The void density and swelling peak at the depth of 3 μm for the same sample is believed to be the correspondent of the damage peak, where the point defect production rate has a sharp increase. One may notice that comparing with the Monte Carlo calculation, the observed damage peak location is much deeper below the surface. Similar discrepancies have been noted for a long time in the cross section studies of 14 MeV heavy ion irradiated materials [10,12,16]; the most common explanation is that the electronic stopping data used in the range calculation is usually too high [10,12].

AEM Analyses

Because the implanted Cu content is very low and the Cu K_α (8.04 keV) and the Ni K_β (8.26 keV) are close to each other, great care must be taken to distinguish the copper. To detect the small Cu K_α signal which may be hidden in the Ni K_β peak, two regions of interest were selected, one covering the energy range between 7.9 and 8.4 keV (both Ni K_β and Cu K_α are included) and the other only covers the Ni K_α peak. The counting on each point was continued until a constant height (4096 counts) for the Ni K_α peak had been reached. The ratio of the two peak integrals (Cu K_α + Ni K_β divided by Ni K_α) was then calculated and plotted against the depth. Finally, the curve was normalized to Cu concentration versus depth by filling the total number of implanted Cu ions, which is known from the beam current during irradiation, into the area underneath the peak integral ratio curve. That normalized Cu concentration versus depth curve is shown in Figure 7 along with the Cu distribution curve calculated by the Monte Carlo method for the irradiation dose.

To determine the error bar, the measurement at several depths was repeated three times. Although the scattering of the data is relatively large, the increase of the Cu signal in the region 1.5 to 2.75 μm below the implanted surface is distinct. Comparing Figure 7 with Figure 4 (a), (c) and (d), it is quite clear that the implanted Cu suppressed void formation and promoted dislocation loop formation.

Several mechanisms have been proposed in the literature [2,3] to explain the void resistance in Ni-Cu alloys. The most plausible one seems to be the trapping of vacancies and interstitials at the boundaries of fine scaled clusters having compositions different from the matrix [7], because the clustering in the Ni-Cu alloys has been suggested both by experimental results and by thermodynamic considerations. However, since the Cu concentration in this study is so low, especially at the early stages of the irradiation, the validity of the trapping by clustering mechanism is doubtful. The detailed discussion about the void suppression mechanism for this case is beyond the scope of this paper and will be given elsewhere.

From Figure 7, one can also see a relatively large discrepancy between the calculated and measured Cu range and concentration in the Cu ion implanted Ni specimen. The measured values are lower and cover a wider region. That is partly due to the radiation enhanced diffusion, because the diffusion spreading was not considered in the Monte Carlo calculation. There is some evidence showing that the irradiation enhanced diffusion coefficient of Cu in Ni-Cu alloy could be increased by two orders of magnitude by ion bombardment below ~ 550 °C [17]. It should also be recognized that part of the Cu range discrepancy in Figure 7 may arise because the AEM data is obtained at the detection limits of the EDS system.

CONCLUSIONS

The TEM/AEM cross-section analysis method can be an effective tool for studying the effect of injected foreign interstitials in heavy ion irradiated materials, provided that the concentration of the implanted species is high enough to be detected.

A small amount of implanted copper promotes dislocation loop formation and suppresses void formation in ion irradiated nickel at 500°C.

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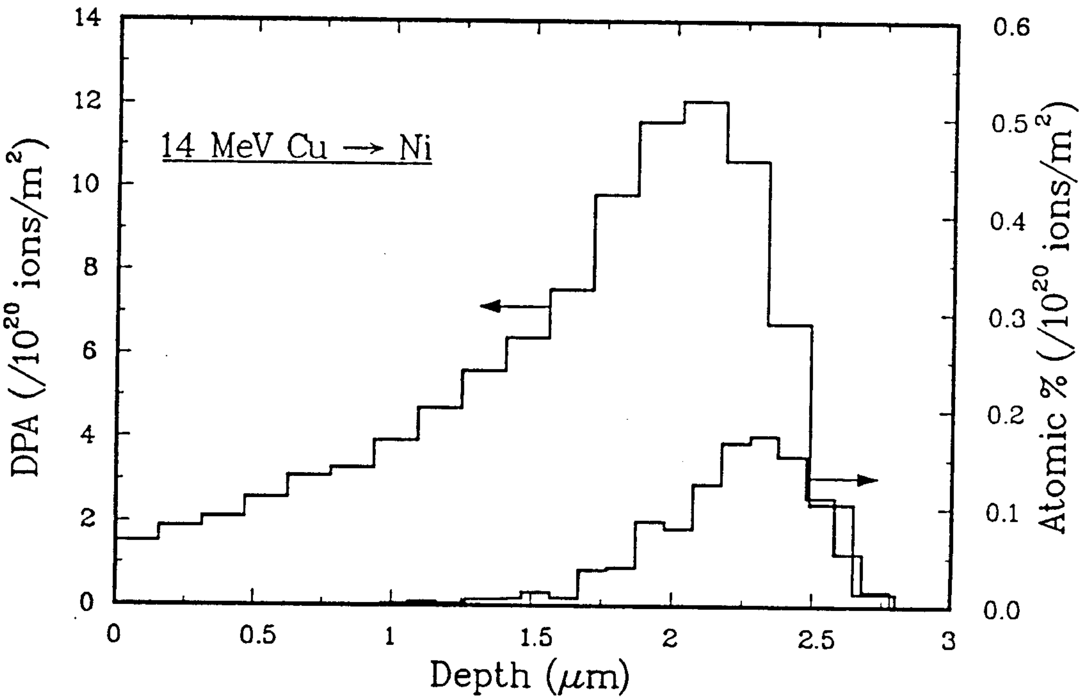

Figure 1. Depth profiles of displacement damage and implanted ion concentration in the 14 MeV Cu ion irradiated Ni, calculated by a Monte Carlo code, the TAMIX (1000 histories).

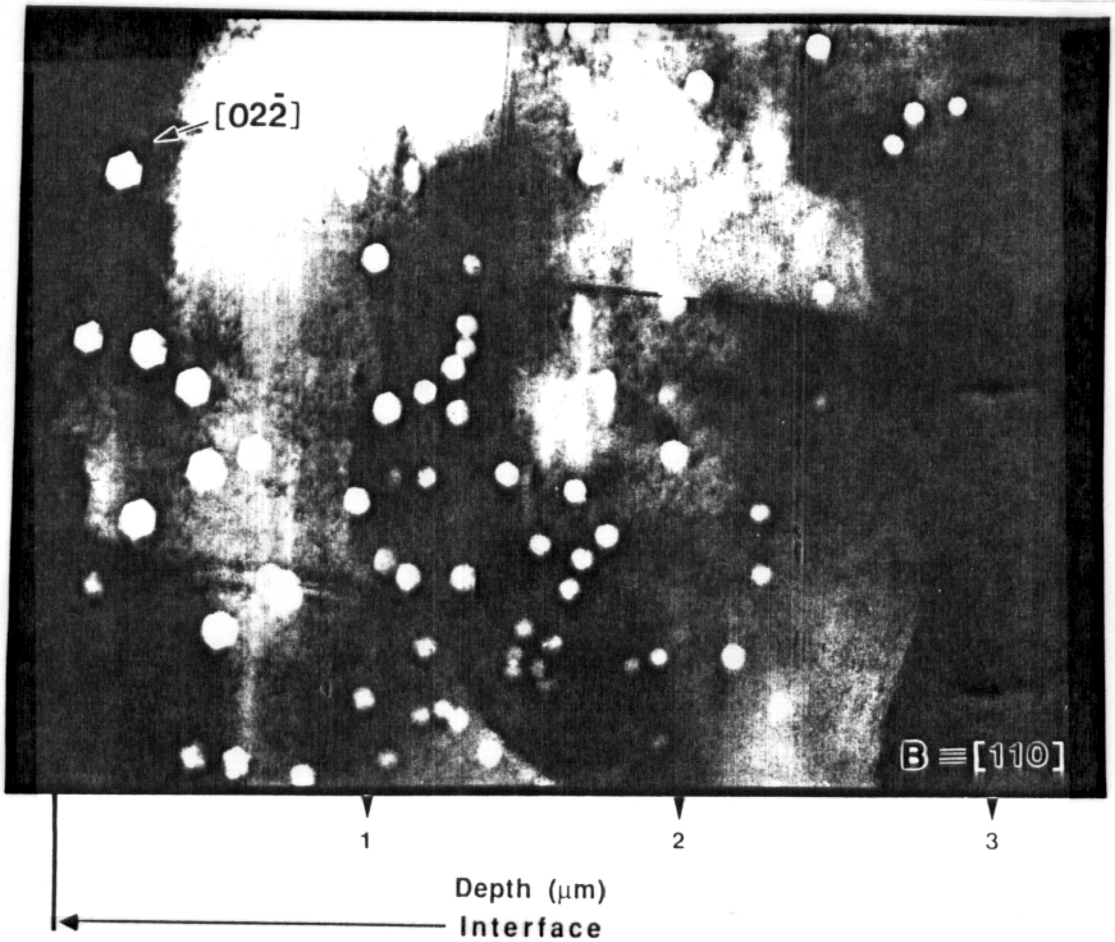

Figure 2. TEM cross-section micrograph showing void distribution in 14 MeV Ni ion irradiated Ni. Since about 0.3 μm was removed from the original surface, the actual depth from the irradiated surface should be the depth from the interface plus 0.3 μm.

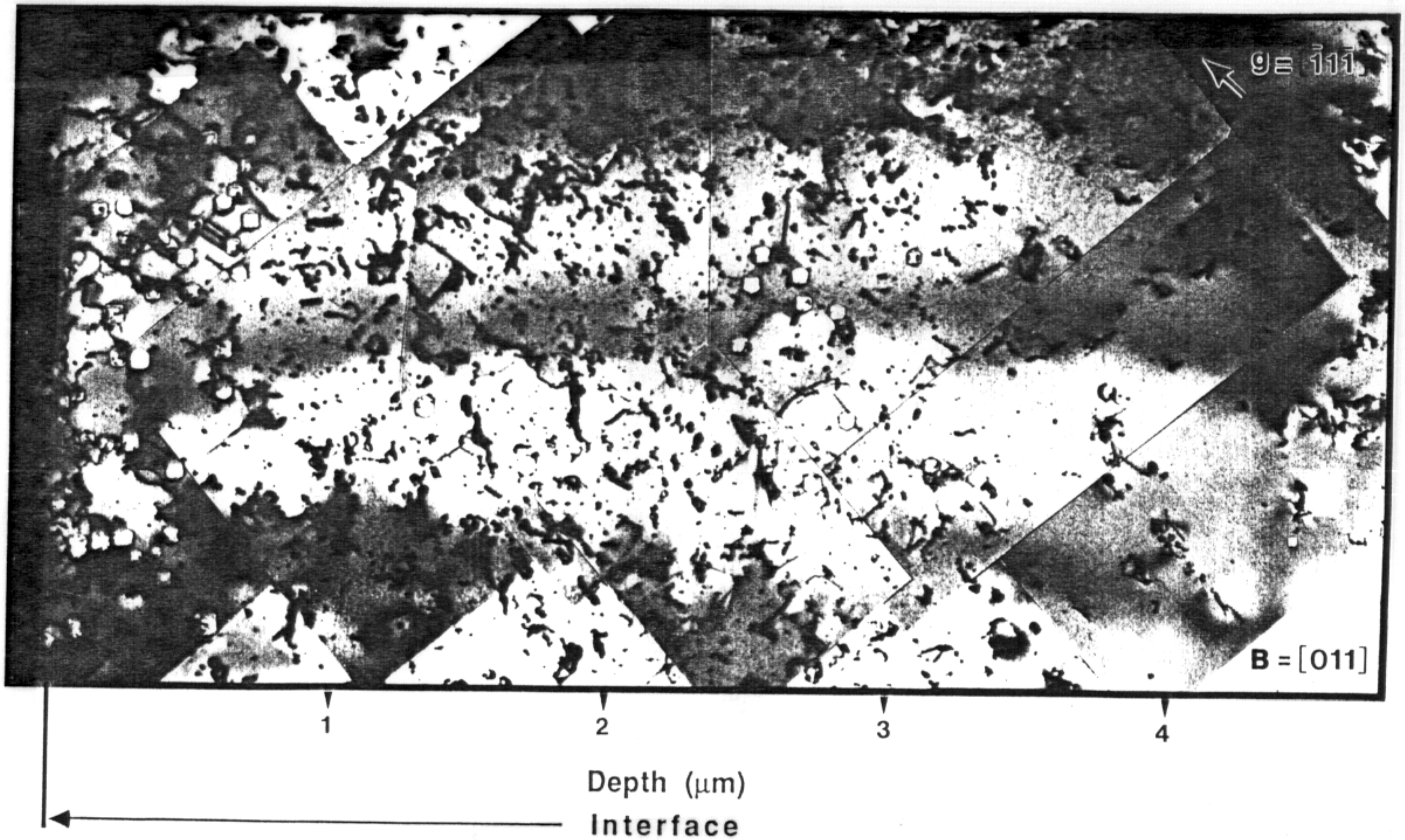

Figure 3. TEM cross-section micrograph showing void and dislocation loop distribution in 14 MeV Cu ion irradiated Ni. Since about 0.3 μm was removed from the original surface, the actual depth from the irradiated surface should be the depth from the interface plus 0.3 μm.

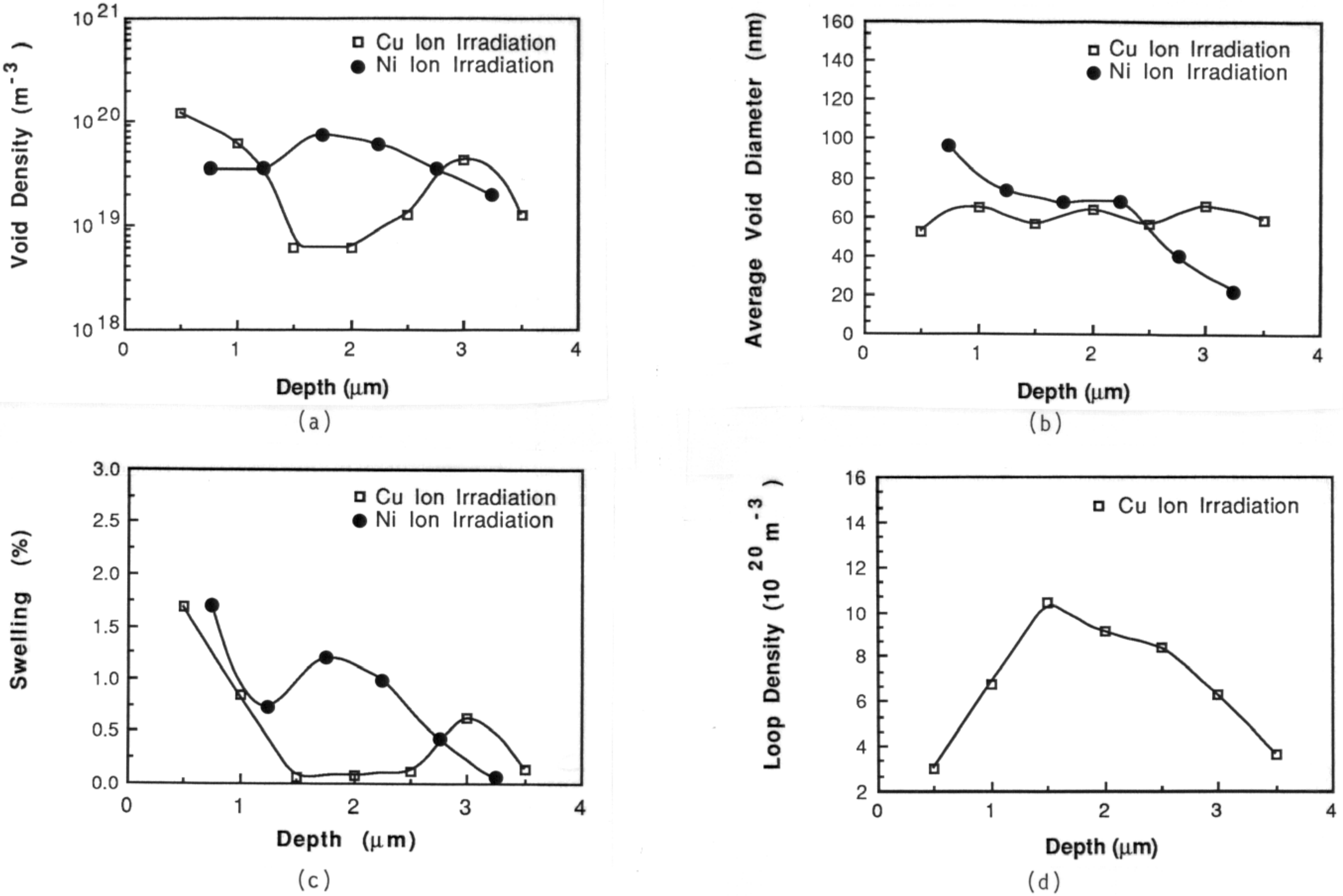

Figure 4. Comparison of (a) void density, (b) average void diameter, (c) swelling versus depth from the irradiated surface curves for Cu and Ni ion irradiated Ni, and (d) dislocation loop density distribution in Cu ion irradiated Ni.

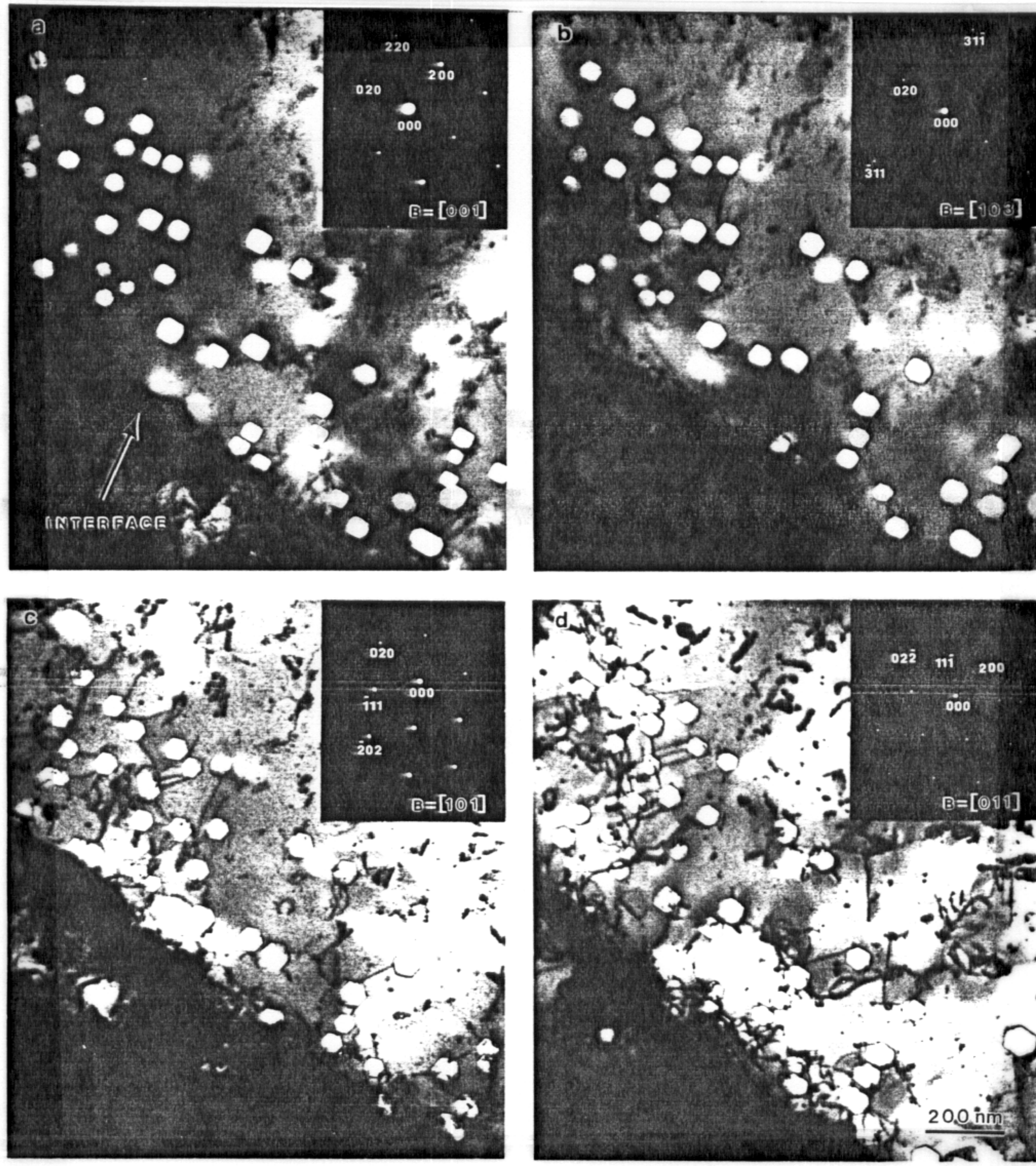

Figure 5. The morphology of voids in 14 MeV Cu ion irradiated Ni. (a) B = [001], (b) B = [103], (c) B = [101] and (d) B = [011]. The voids shown in (a), (b), (c) and (d) are the same group located near the interface.

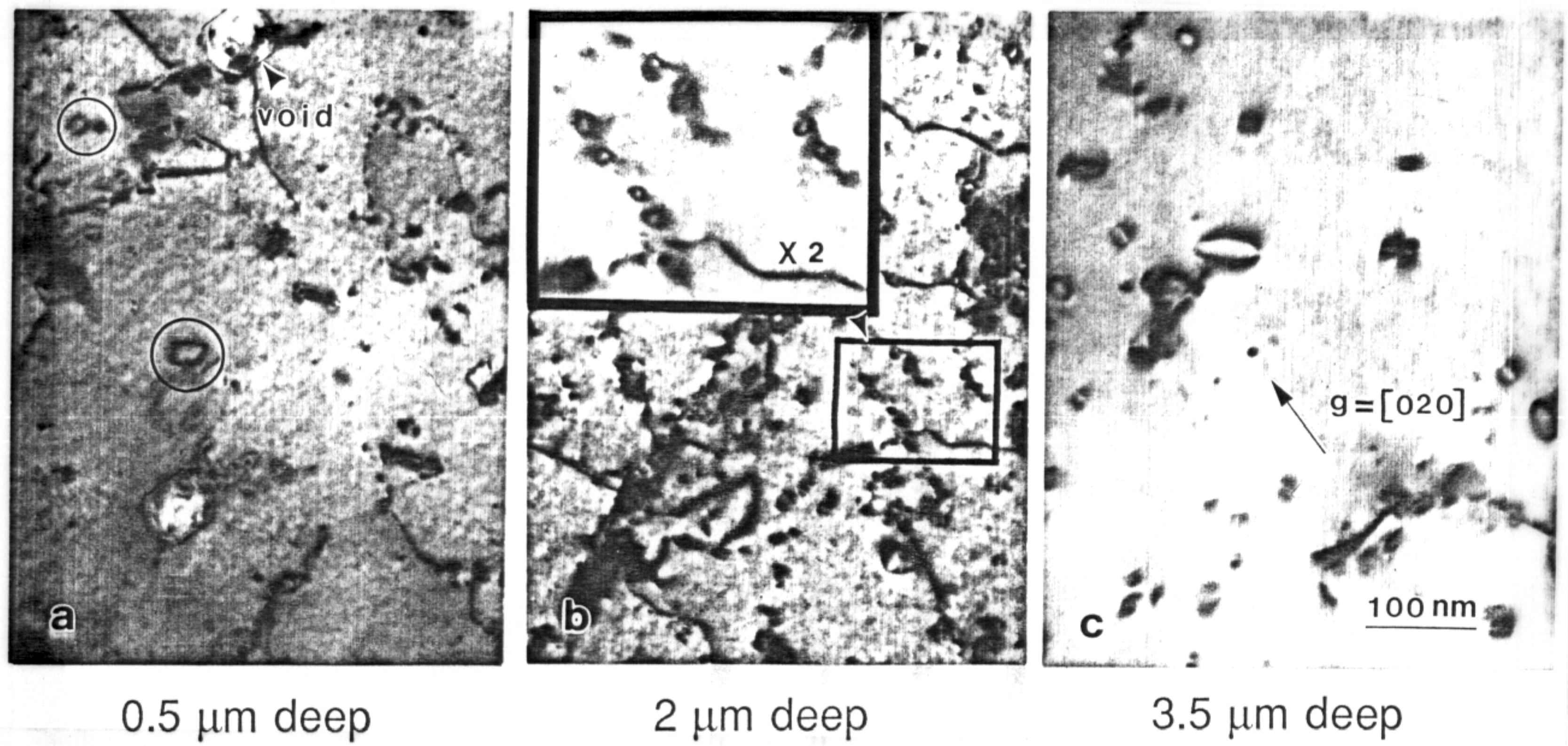

Figure 6. The dislocation loop images taken from the various depths in the 14 MeV Cu ion irradiated Ni. (a) 0.5 μm deep, (b) 2 μm deep and (c) 3.5 μm deep.

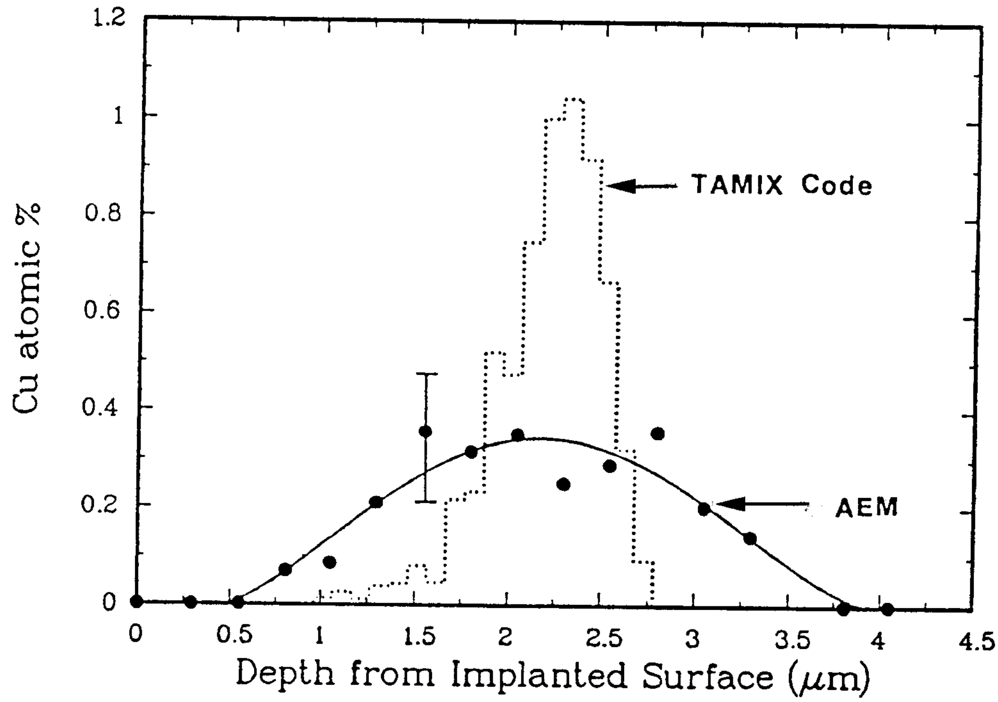

Figure 7. The distribution of implanted Cu in 14 MeV Cu ion irradiated Ni measured by AEM in cross section (normalized by ion implantation dose - 6 x 10^{20} ions/m^2). The calculated Cu distribution, using the TAMIX code for the same dose, is drawn in dotted line for comparison.