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HELIUM BUBBLE FORMATION IN Cu, Ni AND Cu-Ni ALLOYS

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Abstract

Copper, nickel, and Cu-Ni alloys have been irradiated with 200-400 keV ^{3}He ions at a constant homologous temperature of $T/T_m = 0.65$. The samples have been analyzed by TEM to compare helium bubble size and density. Visible bubbles were observed in all samples. The pure copper and the Cu-Ni alloys were found to contain similar bubble densities and sizes after irradiation. The pure nickel samples contained helium bubbles of smaller size and higher density as compared to the alloys.

Introduction

The Cu-Ni system has been the subject of many experimental investigations over the past decade [1-6]. It is one of the few alloy systems which forms a complete solid solution over the entire composition range, although there is some evidence for a miscibility gap under certain conditions [1-3]. Its radiation damage properties are of considerable interest in that Cu-Ni alloys have been shown to be very resistant to void formation over a wide range of damage levels and temperatures [4-6]. Recently, some Cu alloys have been considered for use as high field magnet inserts in both tandem mirror [7-8] and tokamak [9] fusion reactors. The irradiation conditions under which this alloy system has been investigated in the past are shown in Fig. 1. Voids have been easily produced in pure Cu and pure Ni but there has been no published evidence of void formation in Cu-Ni alloys during neutron, ion, or electron bombardment. The cause of the suppressed void formation in the Cu-Ni alloys has remained uncertain.

In order to investigate some of the properties of the Cu-Ni system, we have undertaken a study of helium bubble formation. The purpose of this investigation was twofold. The first objective was to determine if helium bubble formation is possible in the Cu-Ni alloys which are resistant to void formation. A significant difference in the bubble parameters of the alloys as compared to the pure copper and nickel would indicate that there is some fundamental difference in the way that cavities are formed in the Cu-Ni alloys. Secondly, we wished to determine if there was something unusual about the formation or migration energies of vacancies in the alloys compared to the pure metals.

Experimental Procedure

Copper, nickel and Cu-Ni alloys (Cu-20 Ni, Cu-50 Ni, Cu-80 Ni) were obtained from J.L. Brimhall of Pacific Northwest Laboratory in the form of 3 mm disks. These samples were polished and then annealed at a homologous temperature of 0.75 T_m prior to implantation with helium. The TEM discs were irradiated with 200-400 keV ^{3}He ions at the Oak Ridge National Laboratory Van de Graaf facility to obtain a helium concentration of 200 appm at a depth of approximately 0.7 μm. Figure 2 shows the implanted helium profile for nickel, which was calculated using the program E-DEP-1. The incident He flux was on the order of 5 x 10^{11} He/cm^2-s and the total damage level created during the implantation was less than 10^{-3} dpa. The samples were implanted at a constant homologous temperature T/T_m = 0.65 to ensure equivalent thermal vacancy concentrations in each alloy. The temperatures ranged from 608°C for pure copper to 849°C for the pure nickel specimens. The premise of constant thermal vacancy concentration at a given homologous temperature assumes that E_f^v varies monotonically with alloy concentration, and experimental evidence indicates that this is a valid assumption [10].

All samples were held at the implantation temperature for one hour following the irradiation. The relatively high homologous temperature of T/T_m = 0.65 was used to ensure that there was no survival of displacement damage structure from the ^{3}He irradiation.

A thickness of 0.7 μm was removed from the front surface of the TEM samples using an electrolyte of 67% CH_3OH/33% HNO_3 at -20°C and a voltage of 3-4 volts. The depth removed was checked by measuring the amount of fringe shift at the step height between the polished and unpolished region of the foil with an interference microscope. The foils were then back-thinned by jet polishing with the same electrolyte at a temperature of -45°C and a voltage of 9-12 V. The samples were examined in a JEOL 200 CX electron microscope.

Experimental Results

A low density of He bubbles was observed in the pure Cu and pure Ni, and in all three of the alloys. Typical micrographs of the helium bubbles are shown in Figs. 3 and 4 for the pure elements and the alloys, respectively. The bubble parameters for the Cu, Ni and Cu-Ni alloys are summarized in Table 1. The helium bubbles for all samples investigated were found to be at, or in the vicinity of, a strain field such as a dislocation or grain boundary. No large systematic difference was determined between the bubble parameters at grain boundaries versus dislocations. Figure 5 shows the variation of bubble density and size as a function of alloy composition. There is essentially no dependence of these parameters on alloy composition, with the exception that the bubble distribution in the pure nickel sample is substantially more dense and of smaller size. An appreciable density of these small ($d \lesssim 5$ nm) bubbles was also observed in the other samples. For the alloy and pure copper samples the small bubbles accounted for less than 5% of the total visible helium bubble volume.

The concentration of implanted helium in the visible bubbles was calculated using a newly developed high-density equation of state for He [11]. The gas concentration in the visible bubbles as a function of alloy composition is shown in Fig. 6. As can be seen, a substantial fraction of the implanted helium can be accounted for by TEM. For this calculation, the solid-vapor surface energy was assumed to vary linearly among the alloys from 1.8 J/m^2 for copper to 2.3 J/cm^2 for nickel [12]. The helium atom per vacancy ratio for the visible bubbles was calculated to be on the order of 1/2 by utilizing the

Table 1. Bubble Parameters for Cu, Ni and Cu-Ni Alloys

Material	T_{irr} (°K)	Bubble Density x 10^{20} (m^{-3})	Average Visible Bubble Size (nm)
Cu	881	1.0	18
Cu-20 Ni	912	2.5	18
Cu-50 Ni	983	1.2	17
Cu-80 Ni	1071	0.83	25
Ni	1122	$\gtrsim$ 11	7

previously mentioned equation of state for helium, and assuming equilibrium bubbles.

Discussion

Helium bubbles have been found to form readily in Cu-Ni alloys at the homologous temperature of $T/T_m = 0.65$. The observed bubble parameters of the alloys are similar to those in pure Cu -- no significant differences have been found. This behavior is not unexpected as there are theoretical and experimental indications that helium tends to undergo spontaneous precipitation when implanted in metals, even when implanted at below damage threshold energies [13,14]. The fact that nothing unusual was observed in the bubble parameters for the Cu-Ni alloys indicates that there is apparently nothing anomalous about the vacancy formation energy for these alloys -- otherwise, one would expect a significant difference in the bubble parameters of the alloys compared to those bubbles formed in pure metal (e.g., bubble size and/or bubble density). This conclusion is in agreement with published experimental results [10].

The bubble parameters for the pure Ni samples were found to be substantially different from those of the pure Cu and alloy samples. The addition of copper to nickel causes a decrease in the bubble number density and an increase in the bubble size. The cause of this effect was not determined in this study.

In summary, we have found nothing concerning helium bubble growth in the Cu-Ni system at a homologous temperature of $T/T_m = 0.65$ which would explain the suppression of void swelling in the alloys. It would be worthwhile to study helium bubble growth in this alloy system at a lower homologous temperature and at lower helium concentrations. Also, the use of subthreshold energy implantation methods would be relevant as this would eliminate the contribution of implantation-produced vacancies.

Acknowledgments

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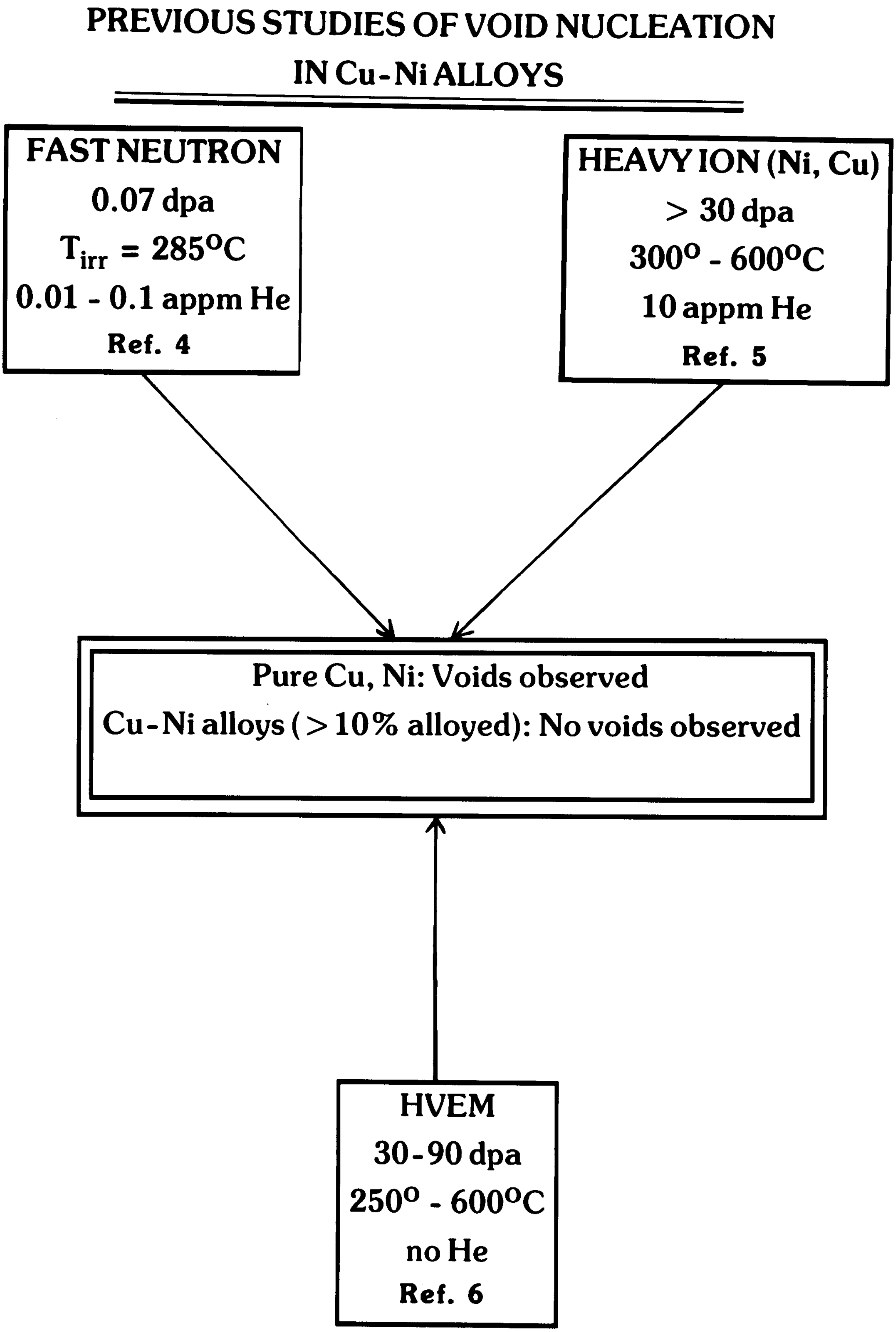


Fig. 1. Summary of previous studies of void formation in Cu-Ni alloys.

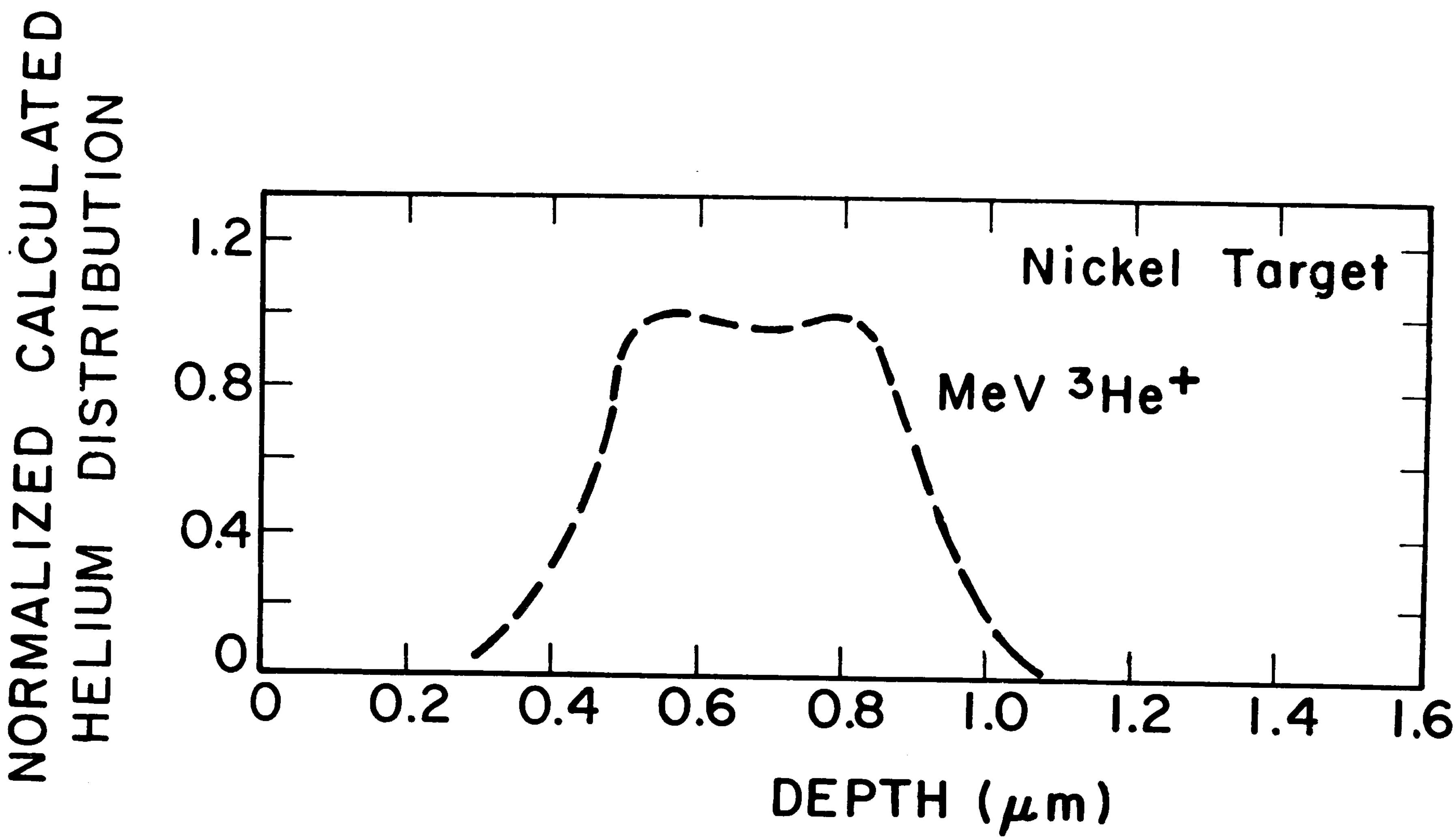
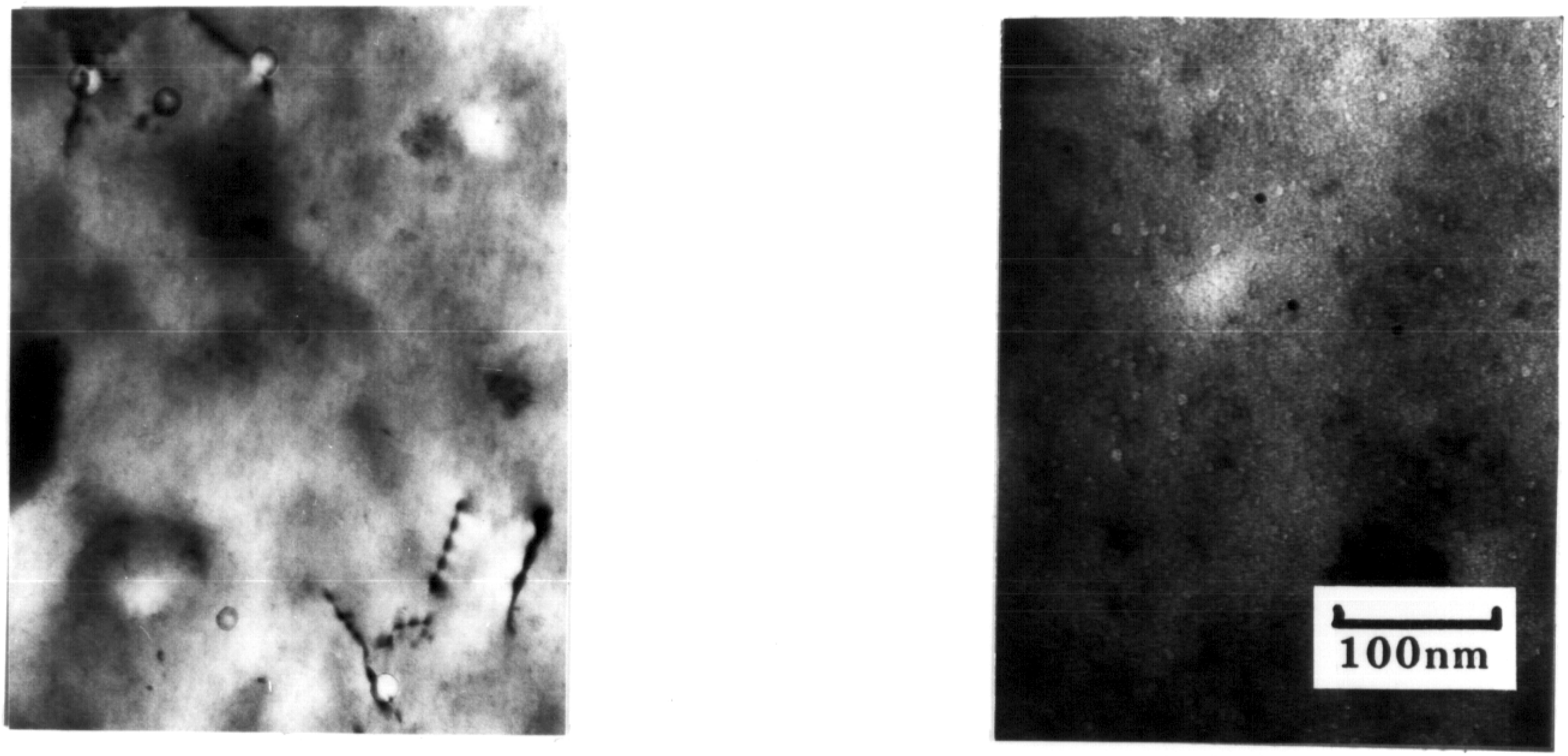


Fig. 2. Calculated implanted helium profile for the Cu-Ni alloys used in this study.

HELIUM BUBBLES IN PURE COPPER AND NICKEL



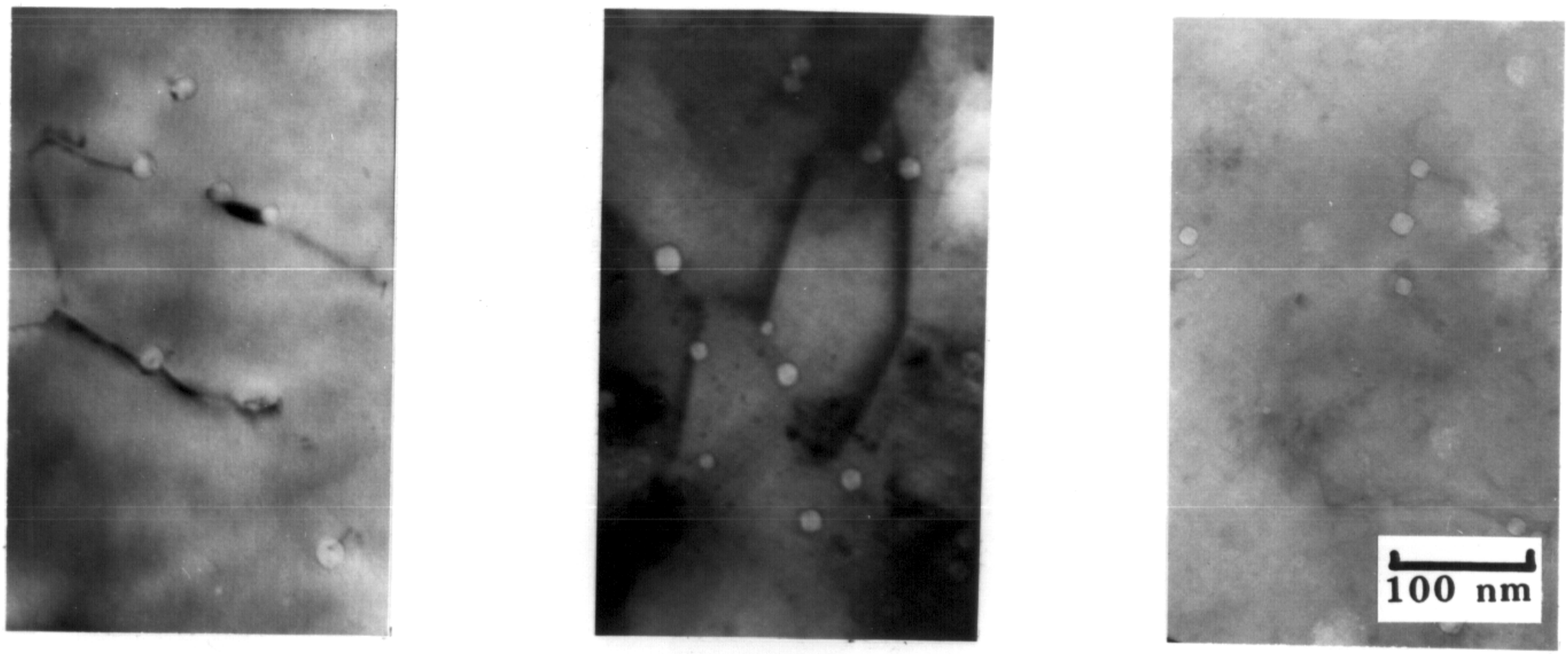
COPPER

NICKEL

200 appm He, 0.65T_m

Fig. 3. Helium bubbles observed in pure copper and nickel implanted with 200 appm He at the homologous temperature $T/T_m = 0.65$.

HELIUM BUBBLES IN Cu-Ni ALLOYS



Cu-20Ni   Cu-50Ni   Cu-80Ni

200 appm He, 0.65 T_m

Fig. 4. Helium bubbles observed in Cu-Ni alloys implanted with 200 appm He at the homologous temperature $T/T_m = 0.65$.

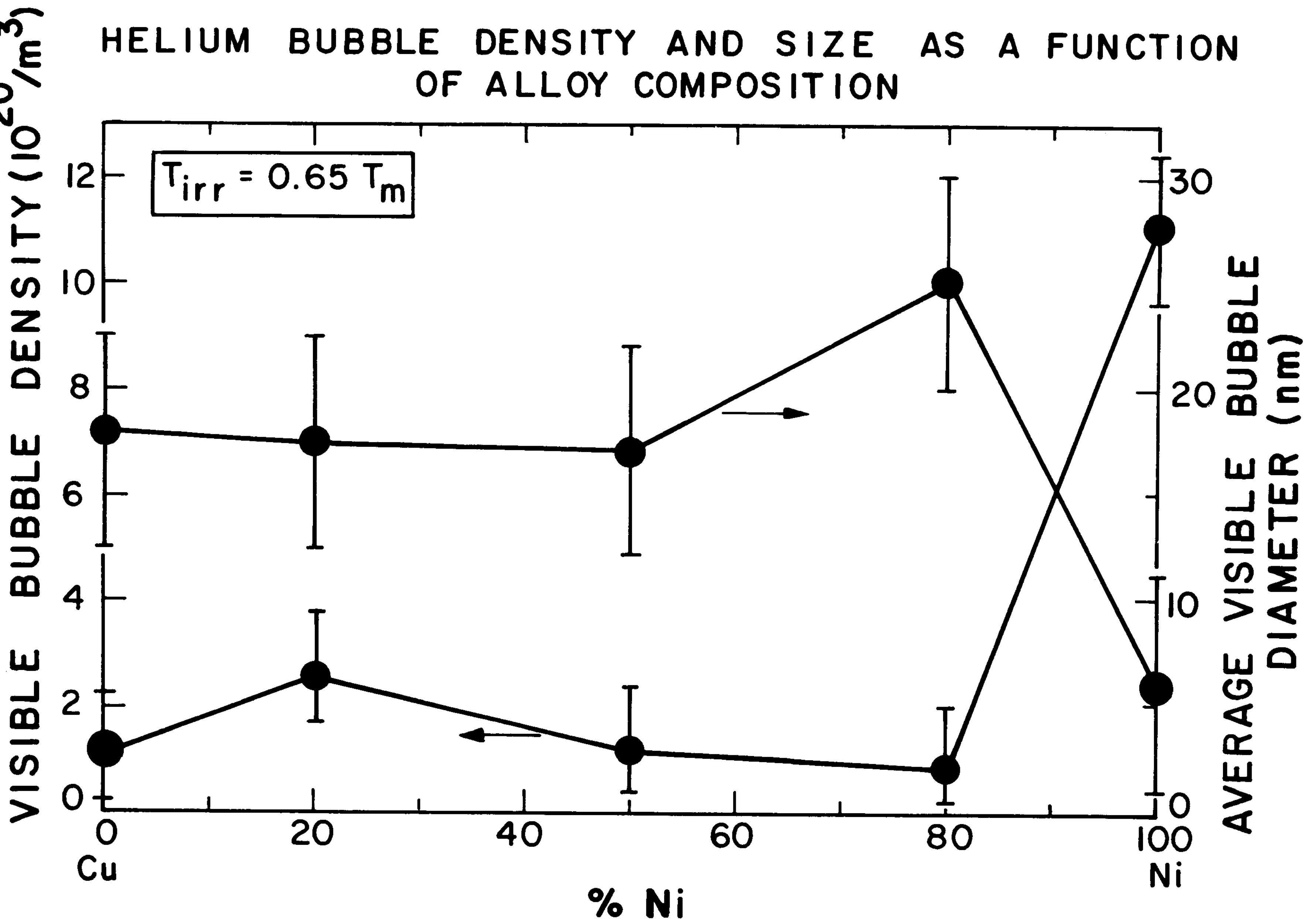


Fig. 5. Variation in the visible bubble density and average bubble size with alloy composition.

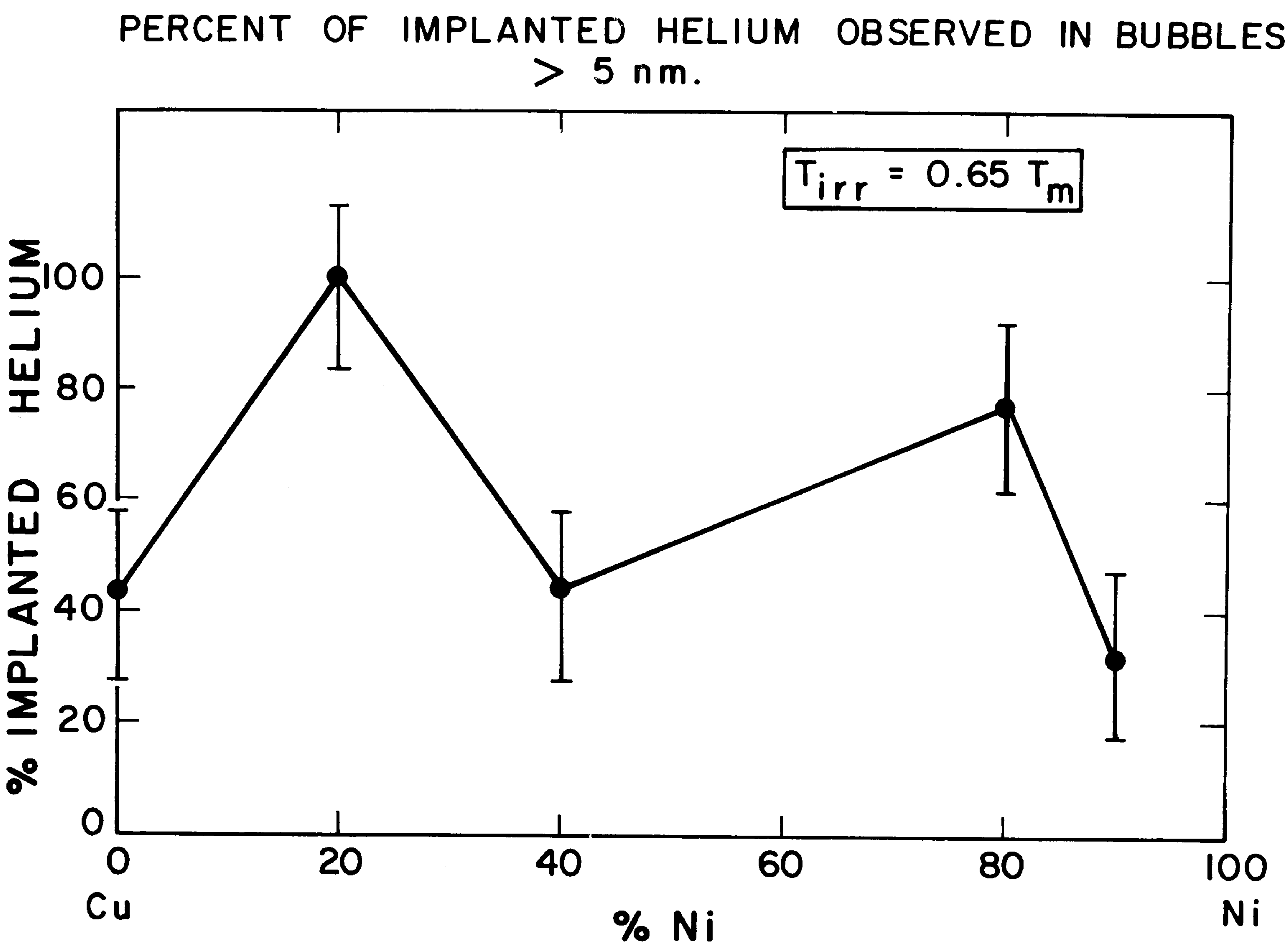


Fig. 6. Percent of helium observed in bubbles for the Cu-Ni alloys. A recently developed high-density equation of state is used for helium [11].