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Void Formation and Growth in Copper-Nickel
Alloys During Irradiation in the High Voltage
Electron Microscope

by

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May 1977

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Abstract

The formation and growth of voids during irradiation in a high voltage electron microscope were studied in copper and Cu-Ni alloys. For each composition the range of irradiation temperatures from 250°C to 550°C was covered. The development of the irradiation-induced dislocation structure was also studied. At irradiation temperatures up to 450°C the void swelling decreased rapidly with increasing Ni content and became practically zero for Cu-10%Ni. The decrease in swelling was produced mainly by decreased void growth (and not by decreased void number density). At 550°C the void swelling increased with increasing Ni content up to 5%, whereas for Cu-10%Ni the swelling became practically zero; again the changes in swelling with Ni content were mainly determined by changes in void growth.

The reduction in void swelling and growth due to alloying is ascribed to vacancy or interstitial trapping at submicroscopic Ni precipitates, i.e. to the precipitates acting as recombination centres. The increase in void swelling and growth with increasing Ni content, on the other hand, is ascribed to dislocation climb sources which emit loops and hence produce a fairly high dislocation density at a temperature where there are only few dislocations in pure copper or Cu-Ni with lower Ni content.

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1. INTRODUCTION

It is well documented that alloying elements may reduce void swelling (e.g. Nelson, Hudson, Mazey, Walters, and Williams 1972, Appleby and Wolff 1973, Hudson, Francis, Mazey, and Nelson 1973, Michel and Moteff 1974, Hudson and Ashby 1975, Sorensen and Chen 1975, Bloom, Stiegler, Rowcliffe, and Leitnaker 1976).

In the copper-nickel system Brimhall and Kissinger (1972) and Mazey and Menzinger (1973) (using neutron and ion irradiation, respectively) found swelling reduction or total swelling suppression for a very wide composition range (2-98% Ni). The investigations of Brimhall and Kissinger and Mazey and Menzinger were of a preliminary nature. They covered the whole composition range of Cu-Ni alloys with five compositions. Brimhall and Kissinger used only one irradiation temperature (285°C) and one (quite low) dose ($2.4 \cdot 10^{23}$n/m^2). Mazey and Menzinger carried out the irradiations at various temperatures for each composition, but the temperature range covered was fairly narrow, and the irradiation conditions were not the same for the various temperatures.

According to the published phase diagrams (e.g. Hansen 1958) the Cu-Ni system is a single-phase system which should rule out any swelling-reduction mechanism involving precipitates. However, as pointed out by Mazey and Menzinger, there are a number of indications that in spite of this apparent complete solid solubility, there is thermodynamically a miscibility gap in the Cu-Ni system (e.g. Schüle, Spindler, and Lang 1975). Also, it is well established that irradiation can cause phase instability and induce precipitation. The observation of dislocation climb sources in electron-irradiated Cu-Ni (Leffers and Barlow 1976, Barlow and

Leffers 1977) demonstrates the existence of submicroscopic precipitates or clusters in Cu-Ni at Ni contents as low as 1-2%.

It is the aim of the present work to investigate in detail the void swelling in Cu-Ni alloys. The Ni content was varied in the range 0-10%; the upper limit of 10% is determined by the fact that at this Ni content the swelling becomes practically zero at all irradiation temperatures. The irradiation temperature was varied in the range 250-550°C. The irradiations were done in a high voltage electron microscope (HVEM).

For the proper understanding of the influence of alloying elements on swelling it is important to separate the effects of alloying on void nucleation and on void growth. In the present work we emphasize such a separation of the effects of changes in alloy composition and irradiation temperature on void number density (nucleation) and on void size (growth). We also underline the importance of the observed differences in the irradiation-induced dislocation structure for the swelling process.

2. EXPERIMENTAL PROCEDURE

The materials investigated were Cu and Cu-Ni alloys containing 1, 2, 5, and 10% Ni (by weight). The alloys were produced from Cu and Ni of 99.999% purity by melting in an argon atmosphere. The material was rolled to 0.25 mm plate from which discs were punched. The discs were annealed in vacuum for 4 hours at 800°C. Thin foils for electron microscopy were prepared from the discs by electropolishing in a solution of 33% nitric acid in methanol at -30°C and 10 V.

The irradiation was done in the AEI EM7 microscope at Harwell operated at 1 MV with dose rates in the range 0.9-1.4·10^{-2} dpa per second ($-2·10^{24}$ electrons/m^2/s). The nominal irradiation temperature was controlled within $\pm$ 3°C. The actual irradiation temperature would be within 10°C of the nominal temperature. During irradiation a vacuum of $-5·10^{-7}$ torr was maintained in the specimen chamber. The exact average dose rate within a circle of radius 0.4µm at the centre of the irradiated area was measured before each experiment. The reduction in dose rate at a distance of 0.5 µm from the centre would be about 25%.

The thickness of the irradiated areas was about 1 µm at the lower irradiation temperatures (250-350°C). At the higher irradiation temperatures it was necessary to increase the thickness to produce voids; at 550°C the thickness was -2 µm. During irradiation the developments in void structure with dose were followed by taking a number of micrographs. The micrographs would cover at least two different orientations of the foil. The thickness of the layer with voids could then be calculated from the maximum relative displacement between two voids and the tilting angle. The void density C_v was calculated from the number N of voids in a certain area A (about 1 µm^2 in the central part of the irradiated area) and the thickness t of the layer with voids:

$$C_v = N/(A·t) \tag{1}$$

The mean diameter d_v was calculated from

$$d_v = (\sum_{i=1}^{N} d_i^3/N)^{1/3} \tag{2}$$

where d_i is the measured diameter of void i. The swelling $\Delta V/V$

is given by

$$\Delta V/V = C_v \cdot \pi/6 \; d_v^{\,3} \tag{3}$$

The void micrographs were made in diffraction conditions with the dislocations out of contrast. Micrographs were also made in orientations with dislocation contrast, so that the development of the dislocation structure could be followed.

The dislocation flux (which is defined as the product of the dislocation density ρ and the average velocity v_d of the dislocations in their climb and/or glide motion) was calculated from

$$\rho \cdot v_d = 2N_d/t \tag{4}$$

where N_d is the number of times (per unit of time) that a given point in the thin foil was passed by a dislocation, t is the foil thickness, and 2 is a geometrical correction factor allowing for the fact that the dislocation motion is observed in plane projection (Makin 1974). In practice the measurements were made on a cine-film taken from the screen (with one frame every two seconds).

3. RESULTS

In this section we shall first describe the variation of swelling, void density, and void size with composition and irradiation temperature. Later in the section the observations on the dislocation structure developed during irradiation are described. The reproducibility of all the experimental results quoted has been checked by independent experiments.

3.1. Swelling

Fig. 1 gives characteristic values for the void swelling

as a function of alloy composition for the various irradiation temperatures. The swelling corresponds to doses of 90 dpa, 45 dpa, and 30 dpa for the irradiation temperatures $250^{\circ}C$, $350^{\circ}C$, and $450-550^{\circ}C$, respectively; as shown below (figs. 2-4) the swelling values quoted for specific doses in fig.1 give a reasonable representation of the swelling rates. Fig.1 shows that, at all temperatures, the swelling is reduced to practically zero at a Ni content of 10%; in the earlier work (Brimhall and Kissinger 1972, Mazey and Menzinger 1973) no voids were observed in Cu-20%Ni. For irradiation temperatures up to $450^{\circ}C$ the swelling decreases with increasing Ni content. For irradiation temperatures $500^{\circ}C$ and $550^{\circ}C$ the swelling reaches a maximum for 2% and 5% Ni, respectively.

It should be noted that the difference between the swelling results for $250^{\circ}C$, $350^{\circ}C$ and $450-550^{\circ}C$ has been reduced, because the swelling results for different irradiation temperatures are given at different doses (and not at the same dose). However, such a difference in dose was found to be necessary: for the lower irradiation temperatures and higher Ni contents high doses were necessary to produce a measurable swelling, whereas for the higher irradiation temperatures too high doses were to be avoided in order to keep the voids reasonably small compared to the foil thickness.

Fig. 2 and 3 show the swelling as a function of irradiation time at $350^{\circ}C$ and $500^{\circ}C$ for the various alloy compositions up to 5% Ni. There is no clear indication of an incubation period*

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* The term "incubation period" refers to the intersection of the abscissa axis and a straight line through the points registered in a given experiment; the use of this term does not necessarily imply that such a linear extrapolation beyond the lowest point registered reflects the physical swelling behaviour.

except for Cu-5%Ni at 500°C. In order to evaluate the possible effect of incubation periods in Cu-5%Ni at other irradiation temperatures we have in fig. 4 plotted swelling versus time for all irradiation temperatures except 250°C (at 250°C the points are even closer to the abscissa axis than they are at 350°C). A comparison of fig. 1 and fig. 4 shows that fig. 1 does give a reasonable representation of the swelling rates in Cu-5%Ni for all irradiation temperatures in spite of the fact that the lines in fig. 4 do not intersect the abscissa axis at time zero.

Fig. 5 gives the temperature at which swelling has its peak value as a function of alloy composition; the data are extracted from fig. 1. There is a monotonic increase in peak--swelling temperature with increasing Ni content.

3.2. Void density

Fig. 6 shows the temperature dependence of the maximum void number density, C_V^{max}, for Cu and Cu-Ni alloys. It should be noted that the maximum in the void number density generally occurs at doses lower than those for which the swelling results are given in fig. 1. In fig. 6 no C_V^{max} value is given for Cu-10%Ni at 350°C, 450°C, and 500°C for the following reasons: at 350°C C_V^{max} varied enormously from experiment to experiment, and in some experiments no voids were observed; at 450°C and 500°C we were not able to produce voids in the normal irradiation experiments (see 3.3.).

Since there is no clear difference between the C_V^{max} values (at a given temperature) for Ni contents of 2, 5, and 10%, the temperature dependence of C_V^{max} for all these three Ni contents is represented by one common (dotted) curve.

3.3. Void size

The mean void diameters for specific doses at the various irradiation temperatures are presented as a function of Ni content in fig. 7. The specific doses selected are 90 dpa, 45 dpa, and 30 dpa at $250^{\circ}C$, $350^{\circ}C$, and $450-550^{\circ}C$, respectively (as in fig. 1). In general, the size decreases with increasing Ni content for irradiation temperatures up to $500^{\circ}C$.

As already mentioned, the results for Cu-10%Ni at $350^{\circ}C$ were rather scattered and consequently no point is given. The dotted curve between 5 and 10% Ni at $350^{\circ}C$ represents an upper limit; it is based on the observation that in none of the experiments the mean void diameter in Cu-10%Ni was found to be bigger than that in Cu-5%Ni. For Cu-10%Ni irradiated at $450^{\circ}C$ and $500^{\circ}C$ no voids were observed in the normal experiments, and hence no points are shown. In order to investigate whether voids could grow at all in Cu-10%Ni at $450^{\circ}C$ and $500^{\circ}C$ we preirradiated Cu-10%Ni specimens at $250^{\circ}C$ for 2 hours and then raised the irradiation temperature to $450^{\circ}C$ and $500^{\circ}C$. This led to void formation and growth at $500^{\circ}C$. The dotted curve between 5 and 10% Ni is based on the size of these voids. At $450^{\circ}C$ we were not able to produce visible voids.

For irradiation at $550^{\circ}C$ there is a clear maximum in the mean void diameter for a Ni content of 5%. No voids were observed in pure copper and in Cu-1%Ni at $550^{\circ}C$. This is consistent with observations in independent irradiation experiments in which voids were first grown at $500^{\circ}C$ in Cu-1%Ni to a size larger than 100 nm. When the specimen temperature was raised (while still irradiating) to $550^{\circ}C$, the voids ceased to grow, and after a period of continued irradiation they exhibited a tendency to shrink. This (i.e. zero growth) provides justification

for the zero size points for Cu and Cu-1%Ni at 550°C in fig. 7.

Figs.8 and 9 show the mean void diameter as a function of irradiation time at 350°C and 500°C for Ni contents up to 5%. The curves confirm that the particular choice of doses in fig. 7 (corresponding to irradiation times of approximately 120 min., 60 min., and 40 min. for 250°C, 350°C, and 450-550°C, respectively) gives a reasonable representation of the void growth rates.

Fig. 10 shows void micrographs for irradiation at 350°C for the various compositions up to 5% Ni content. For 0,1 and 2% Ni the doses are of the order of 45 dpa as in figs. 1 and 7; for 5% Ni it has been necessary to go to a substantially higher dose.

3.4. Relation between void density, void size, and swelling

The swelling results in fig. 1 can be described in terms of void number density and mean void diameter on the basis of figs. 6 and 7*.

In the temperature range up to 450°C, where swelling decreases monotonically with increasing Ni content, the decrease in swelling is due to decreasing void diameter. The only exceptions are the decreases in swelling when going from 0 to 1% Ni at 350°C and 450°C, which are caused by decreasing void

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* The results in fig. 6 refer to an earlier stage of irradiation than those in figs.1 and 7, but the number densities corresponding to figs.1 and 7 are fairly close to those in fig.6.

density (the diameter remains almost unchanged). A preirradiation experiment was made to further investigate these decreases in swelling caused by decreases in void density: a specimen of Cu-1%Ni was irradiated at 250°C for 30 min. and then irradiated at 450°C (i.e. the temperature at which the decrease is most pronounced). The preirradiation increased the void number density to approximately the level observed in copper in the normal 450°C experiment. The increase in void density did not produce any significant change in void size (the mean void diameter for a given dose was approximately the same as that in Cu and Cu-1%Ni in the normal 450°C experiments), i.e. the swelling increased to the level found in copper in the normal 450°C experiment (see fig. 1). Thus the reduction in swelling due to the decrease in void number density from Cu to Cu-1%Ni does not represent a reduction in the basic swelling capacity as the reductions due to decrease in void size do: if the void density in Cu-1%Ni is increased to the level characteristic of copper, the swelling becomes equal to the swelling in copper.

At 500°C the increase in swelling from 0 to 1% Ni is due to the simultaneous increase in void density and void diameter (mainly the former); the increase in swelling from 1 to 2% Ni is due to the increase in void density that more than counter-balances the decrease in void diameter. When the Ni content is increased from 2 to 5%, the void density and void diameter both decrease.

At 550°C the changes in mean void diameter dictate the changes in swelling, the void density remaining almost constant for the compositions which produce voids (2% Ni and more).

Thus the conclusion of this section is that the changes in swelling with alloy composition, particularly in the composition

/temperature range with the characteristic decrease in swelling with increasing Ni content, are mainly governed by the changes in mean void diameter.

3.5. Dislocations and "black dots"

The formation and growth of voids are closely linked to the dislocation structure. We have tnerefore also followed the development of the dislocation structure in the irradiation experiments.

For Ni contents of 2% and more, high dislocation densities (up to approximately 10^{14} m^{-2}) developed for irradiation temperatures up to 450-500°C (cf. Barlow and Leffers 1977). The early stage of development of the dislocation network at 350°C for Ni contents of 0, 1, 2, and 5% is shown in fig. 11. The micrographs (taken in dislocation contrast with fairly thin specimens (~0.7 μm) after a dose of about 8 dpa) illustrate the increase in dislocation density and the drastic increase in the density of "black dots" as the Ni content increases from 1 to 2%; this is exactly the change in composition for which there is a drastic decrease in void diameter (fig. 7). From 2 to 5% Ni there is a moderate increase in the densities of dislocations and black dots. In the specimens with high black--dot density, i.e. for Ni contents of 2% and more, the majority of the black dots remain black dots or at least take very long time to develop into loops (Barlow and Leffers 1977). The corresponding series of dislocation micrographs taken at 450° C (not to be shown) reflects the same behaviour, i.e. an increase in dislocation density and a drastic increase in black-dot density when going from 1 to 2% Ni.

The increase in dislocation density with increasing Ni

content at $350^{\circ}C$ and $450^{\circ}C$ does not lead to an increase in dislocation flux. The average dislocation velocity decreases so much that the net effect is a decrease in flux as shown in fig. 12 (results from Barlow, Ph.D. thesis to be published).

4. DISCUSSION

As described in 3.4, the changes in void swelling with changes in composition in the Cu-Ni alloys investigated are mainly determined by changes in void size or void growth rate. For instance there is a clear trend that the decrease in swelling caused by increasing Ni content (at a given irradiation temperature) mainly comes via reduced growth. In general, changes in the number of voids nucleated do not contribute significantly to the reduction in swelling with increasing Ni content; on the contrary, there is a trend that the void density increases with increasing Ni content.

Thus, the mechanisms for the reduction in swelling due to alloy additions which can only operate via reduction in the number of voids nucleated (e.g. trapping at solute atoms of the gas atoms necessary for nucleation (Hudson et al. 1973)) are ruled out. All the other mechanisms suggested operate via a reduction in the vacancy surplus, as a result of either a) decrease in the preferential (biased) absorption of self-interstitials at the dislocations (Kuhlmann-Wilsdorf 1973, Norris 1975), b) vacancy and/or self-interstitial trapping at solute atoms (Smidt and Sprague 1973, Michel and Moteff 1974, Koehler 1975, Schilling and Schroeder 1975), or c) vacancy and/or self-interstitial trapping at coherent precipitates (Nelson et al. 1972, Appleby and Wolff 1973, Mazey, Bullough, and Brailsford

1976). A reduction in vacancy surplus in accordance with any of these mechanisms would lead to reduced void growth (even though they are often imagined to exert their influence on swelling mainly via a reduction in the number of voids nucleated).

In 4.1 the observed reduction in void size or growth with increasing Ni content will be discussed in terms of reduced vacancy surplus and the different mechanisms suggested for this reduction in vacancy surplus. Even though the general trend is for the void size to decrease with increasing Ni content, there is at 550°C a clear increase in void size with Ni content up to a Ni content of 5%; this will be discussed in 4.2. The different regions for the dependence of void size on Ni content are shown schematically in fig. 13.

The dependence of the observed void number densities on Ni content and irradiation temperature will be discussed in 4.3.

Throughout the discussion we shall assume that any local variation in Ni content due to irradiation-induced segregation is small in comparison with the differences in Ni content between the different Cu-Ni alloys.

4.1. Decrease in void size with increase in Ni content

The average dislocation velocity was found to be very low under the typical conditions of reduced void growth (3.5). This might be considered to suggest an explanation of the reduced void growth and swelling in terms of reduced net absorption of interstitials at the dislocations because of reduced bias. This is, however, not necessarily a correct interpretation: low swelling has, logically, to be accompanied by low dislocation climb rates (or dislocation flux) disregarding the cause of the reduced swelling.

If reduced bias due to the Ni addition had been the cause
of the reduced void growth and swelling in the void-relevant
thicker foils, the reduction in bias should be approximately
proportional to the reduction in swelling rate, i.e. the bias in
Cu-10%Ni should be reduced by orders of magnitude compared to
pure copper. However, according to climb-rate experiments on
thinner foils this does not seem to be the case. In the thinner
foils, where the disappearance of point defects is dominated by
recombination and diffusion to the surface, a reduction in bias
by a certain factor would lead to a reduction in climb rate by
approximately the same factor. Actually, dislocations climb quite
readily in thinner foils of Cu-10%Ni (Barlow and Leffers 1977);
there is nothing like orders of magnitude reduction in climb
rate relative to pure copper. Thus, the experimental observations
are not in favour of the possibility that the reduced void growth
and swelling is caused by reduced bias.

Furthermore, the theoretical arguments for reduction in
bias as the cause of reduced swelling (Kuhlmann-Wilsdorf 1973,
Norris 1975) only refer to oversized substitutional atoms
(which, by attaching themselves to the dislocations, should
cause a reduction in the dilatational stresses), whereas Ni
atoms in copper are undersized.

We are thus led to suggest that the reduction in vacancy
surplus with increasing Ni content that produces the reduced
void growth can only arise as a result of trapping of vacancies
or interstitials leading to enhanced recombination. It is of
course implicit in this hypothesis that there is a binding between
vacancies or interstitials and Ni atoms or clusters of Ni atoms.

Barlow and Leffers (1977) found an activation energy for
interstitial loop growth in thin foils of HVEM-irradiated Cu-10 Ni
higher than that found in pure copper. The difference was

interpreted in terms of vacancy binding with a binding energy of ·0.3eV (the vacancy migration energy being ·1eV).A vacancy binding energy of this magnitude was considered to be unlikely for binding to individual Ni atoms, but reasonable for binding to the Ni clusters that had to be present to account for the dislocation climb sources in the thin foils.

Smidt and Sprague (1973) considered vacancy trapping in a solid-solution alloy with vacancy formation energy (E_V^F) 1.5 eV and vacancy migration energy (E_V^M) 1.0 eV. They found that a vacancy/solute-atom binding energy of 0.4 eV (corresponding to 0.25 E_V^F or 0.4 E_V^M) will produce a significant reduction in vacancy surplus in a 0.3 at% solid solution alloy. In copper 0.25 E_V^F and 0.4 E_v^M are both $\sim$ 0.3 eV (Bourassa and Lengeler 1976). This means that a vacancy binding energy of 0.3 eV in the Cu-10%Ni alloy is, according to Smidt and Sprague, of the correct magnitude to cause a significant reduction in vacancy surplus and hence in void growth.

It must be recognised, however, that the interpretation of the difference in activation energy for loop growth between Cu-10%Ni and pure copper in terms of vacancy binding is not the only possibility. The difference may also, as discussed in the following, be interpreted in terms of interstitial binding.

Kiritani, Yoshida, Takata, and Maehara (1975) derived the following equation for the loop growth rate $\dot{L}$:

$$\dot{L} = 2a(Z_{IL} - Z_{VL})(PM_V/Z_{IV})^{1/2} \qquad (5)$$

where a is a constant for a given loop crystallography, Z_{IL} and Z_{VL} are the numbers of capture sites for interstitials and vacancies around each atom site in the dislocation core, P is

the production rate of Frenkel pairs, M_V is the vacancy mobility, and Z_{IV} is the capture-site number for vacancy/interstitial recombination. In this equation, based on the assumption $M_V \ll M_I$ where M_I is the interstitial mobility, the activation energy is determined by the vacancy mobility. If, instead, it is assumed that the interstitials are the slowly moving point defects $(M_V \gg M_I)$, the growth-rate equation (5) would become:

$$\dot{L} = 2a(Z_{IL} - Z_{VL})(PM_I/Z_{IV})^{1/2} \tag{6}$$

This equation, together with equation (5) for pure copper, could also account for the increase in activation energy in Cu-10%Ni as compared with pure copper. Such an interpretation would imply an interstitial binding energy of ~ 0.9 eV (derived from an interstitial migration energy of ~ 1 eV). An interstitial binding energy of 0.9 eV is unlikely for individual Ni atoms (Koehler 1975); it would have to reflect interstitial binding to the Ni clusters present in the Cu-Ni alloys. An interstitial binding energy of this magnitude would be more than sufficient to produce a very significant reduction in void growth (Koehler 1975). The interstitial binding energies quoted in literature for Cu-Ni are too scattered to be of any real use in judging the importance of interstitial trapping for the reduced void growth.

Thus, the reduction in void growth produced by Ni addition must be caused by the Ni clusters acting as recombination centres - either because the clusters trap vacancies, or because they trap interstitials. On the basis of the present results it is not possible to decide which of these two mechanisms is operative. If one accepts the vacancy-trapping mechanism, one would then have to accept that some Ni clusters mainly trap vacancies

(acting as recombination centres) and others mainly trap interstials (acting as dislocation climb sources), cf. Barlow and Leffers (1977). In the case of interstitial trapping, on the other hand, one would have to accept the very high interstitial binding energy of $\sim$ 0.9 eV.

The results in fig. 7 (summarized in fig. 13) reflect a clear trend for the reduction in void size in Cu-Ni relative to pure copper to become less pronounced with increasing irradiation temperature for a given alloy composition (at the highest temperatures the situation may even change so that the voids grow bigger than in pure copper). This is consistent with the suggestion that the reduction in void growth is caused by point-defect trapping: if there is a given binding energy in a given alloy, the extent of trapping will decrease with increasing temperature. Furthermore, figs. 7 and 13 show that the region in which the void size decreases with increasing Ni content is extended to higher irradiation temperatures as the Ni content increases, which is also consistent with point-defect trapping: if the binding energy increases with increasing Ni content, the temperature corresponding to a given extent of trapping will increase with increasing Ni content.

In the thin foils investigated by Barlow and Leffers (1977) the dislocation loops emitted from the climb sources started as "black dots". These black dots seemed to be Ni precipitates made visible by attached loop nuclei. In the void-relevant thicker foils of alloys containing 2% or more nickel irradiated at 350°C and 450°C (i.e. under conditions typical for the region with reduced void growth) a high density of black dots that did not grow to form loops was observed (see 3.5 and also Barlow and

Leffers). This shows that formation of Ni precipitates and re-
duction in void size appear together, which supports the con-
clusion that the recombination centres are Ni clusters and not
individual Ni atoms. The density of black dots in fig. 11c and
11d is of the order of $5 \cdot 10^{20}$ m^{-3} (i.e. approximately equal to
the void density). The density of recombination centres necessary
to explain the reduced void growth and void swelling would be
substantially greater than the void density. Thus we have to
assume that, apart from the precipitates made visible by attached
loop nuclei, there is a substantially greater number of invisible
precipitates. This corresponds to the conclusion of Barlow and
Leffers that the number of Ni precipitates made visible by attached
loop nuclei (in the early stage of loop emission from a climb
source) is insufficient to explain the observed point-defect
trapping. Mazey et al. (1976) have also suggested that invisible
precipitates (in Al-Mg) were responsible for swelling suppression
by point-defect (vacancy) trapping.

In the bulk material used in their neutron-irradiation ex-
periments Brimhall and Kissinger (1972) observed a population
of black dots and few or no dislocations in Cu-2%Ni and Cu-20%
Ni. This indicates that the population of black dots, which is
also observed in the present work, is a characteristic feature
of irradiated Cu-Ni alloys with reduced swelling. The high dis-
location densities observed in the present work, but not in the
work of Brimhall and Kissinger, are probably surface effects on-
ly to be found in thin film experiments like HVEM irradiation.
The dislocation climb sources operating in the surface layers
(because the vacancy surplus produced by the growth of the in-
terstitial loops can be eliminated by a net vacancy flux to the

surface) produce a high dislocation density. These dislocations rearrange themselves, mainly by glide, to produce a dense dislocation network throughout the foil thickness.

4.2. Increase in void size with increase in Ni content

For irradiation at 550°C the void size and the void swelling (being zero at 0 and 1% Ni) increase with Ni content up to 5% Ni.

We assume that for Ni contents up to 5% the vacancy or interstitial binding energy is insufficient to cause any significant reduction in void growth at 550°C. Thus, in the absence of sufficiently effective recombination centres, these alloys should, as far as void growth is concerned, behave like pure copper. However, in pure copper and Cu-1%Ni one condition for void growth is not fulfilled at 550°C: there is no loop formation and hence no dislocation network or dislocation flux. The dislocation climb sources in alloys with 2% Ni and more still emit dislocation loops at 550°C. These dislocations provide a high dislocation flux (see fig. 12) which makes void growth possible even though the dislocation density is fairly low. In Cu-10%Ni the binding energy is obviously sufficiently high to cause sufficient point-defect trapping and thereby reduce void growth even at 550°C.

It should be underlined that the change from a situation with no dislocations and no voids in alloys with 0 and 1% Ni to a situation with dislocations and voids in alloys with higher Ni content is not just the effect of the increase in homologous temperature due to the increase in melting point: the difference in homologous temperature between Cu-1%Ni and Cu-2%Ni is only about 2°C.

The results indicate that in copper the cut-off in swelling at high temperature is due to the absence of dislocations. When dislocations are provided - in the present work from the climb sources introduced by the addition of nickel - void growth (and void nucleation) continues at least up to $550^{\circ}C$.

4.3. Void number density

The experimental results quoted in fig. 6 show that, at all irradiation temperatures, the void number density in Cu-2%Ni and Cu-5%Ni is higher than that in Cu-1%Ni. At $250^{\circ}C$ the number density in Cu-10%Ni is also higher than that in Cu-1%Ni. At $350^{\circ}C$, $450^{\circ}C$, and $500^{\circ}C$ we found it difficult or impossible to produce visible voids in straight-forward irradiation experiments in Cu-10%Ni (see 3.2 and 3.3), which may reflect either a real suppression of void nucleation or experimental difficulties due to the very low void growth rate (note that there are no difficulties with void nucleation in Cu-10%Ni at $550^{\circ}C$).

It may seem surprising that the number of voids that have nucleated (and grown to become visible) increases when the Ni content changes from 1% to higher values, whereas the void growth rate generally decreases for the same change. If one considers the vacancy surplus to be the main driving force for void nucleation, one would expect the decrease in vacancy surplus with increasing Ni content, discussed in 4.1, to reduce void nucleation, just as it reduces void growth. It should be underlined, however, that void nucleation takes place in the early stages of irradiation, during which the vacancy surplus may be quite different from that in the later stages that determine the subsequent void growth.

A simple extrapolation of the void number densities in Cu-Ni would suggest that the void number density in pure copper at a given irradiation temperature is smaller than that in Cu-1%Ni - which is the case at 250°C and 500°C, but not at 350°C and 450°C. This lack of a simple relation between the void number densities in Cu-Ni and in pure copper originates from the difference in temperature dependence of void number density between Cu-Ni and pure copper (fig. 6). With the present limited understanding of the void nucleation process it seems impossible to explain this complicated variation of void density with alloy composition and irradiation temperature.

5. CONCLUSIONS

The addition of sufficient amounts of nickel to copper reduces void swelling to zero or practically zero at all irradiation temperatures. The nickel content needed to produce a given reduction in swelling increases with increasing irradiation temperature.

For certain combinations of irradiation temperature and alloy composition the nickel addition leads to an increase in swelling.

When nickel additions lead to reduced swelling, the effect is exerted mainly by a reduction in void growth rate and not by a reduction in the number of voids nucleated.

It is suggested that the reduction in void growth rate is caused by vacancy or interstitial trapping at GP-zone-like nickel precipitates, i.e. that the nickel precipitates act as recombination centres.

ACKNOWLEDGEMENTS

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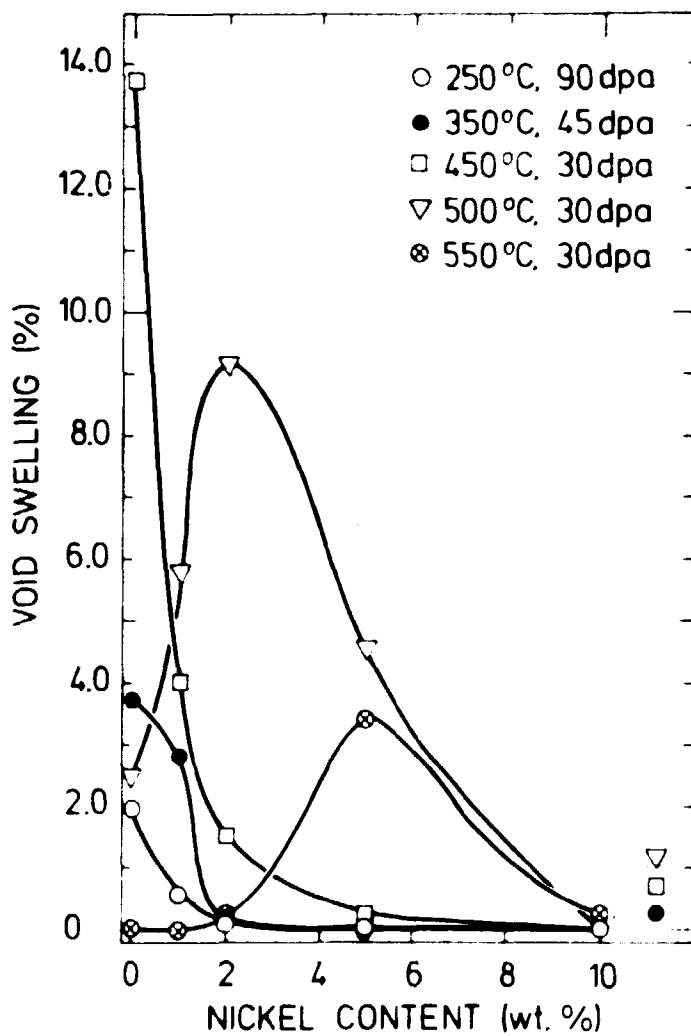
Fig. 1. Void swelling at specific doses as a function of Ni content for the different irradiation temperatures.
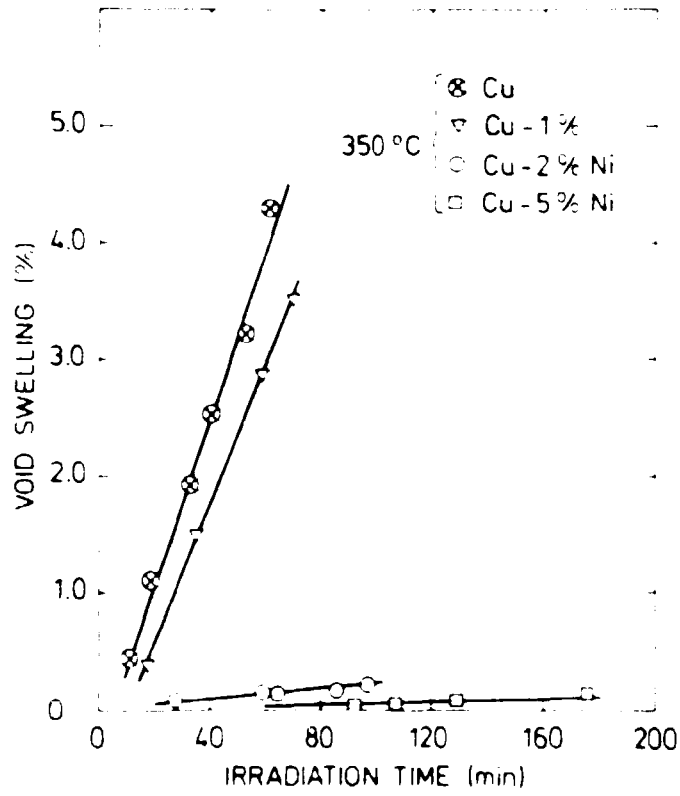
Fig. 2. Void swelling at 350°C versus irradiation time for different alloy compositions.
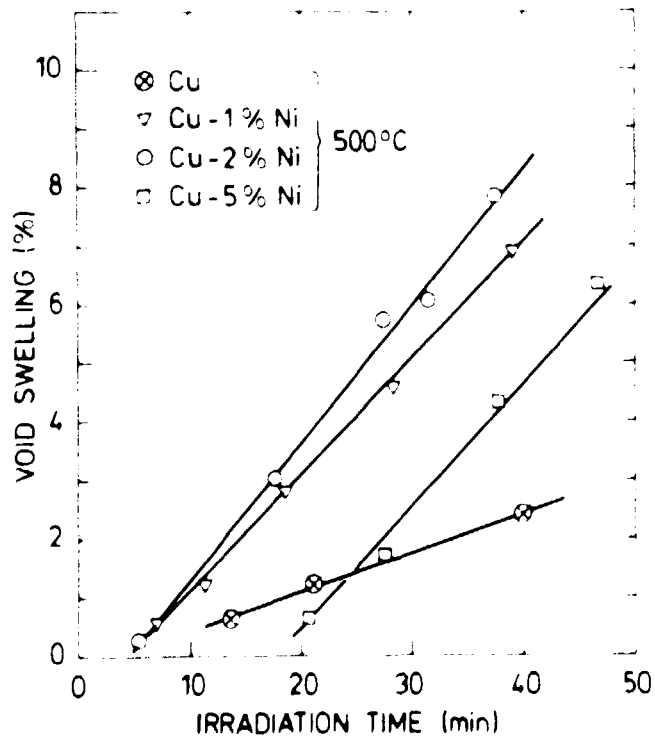
Fig. 3. Void swelling at 500°C versus irradiation time for different alloy compositions.
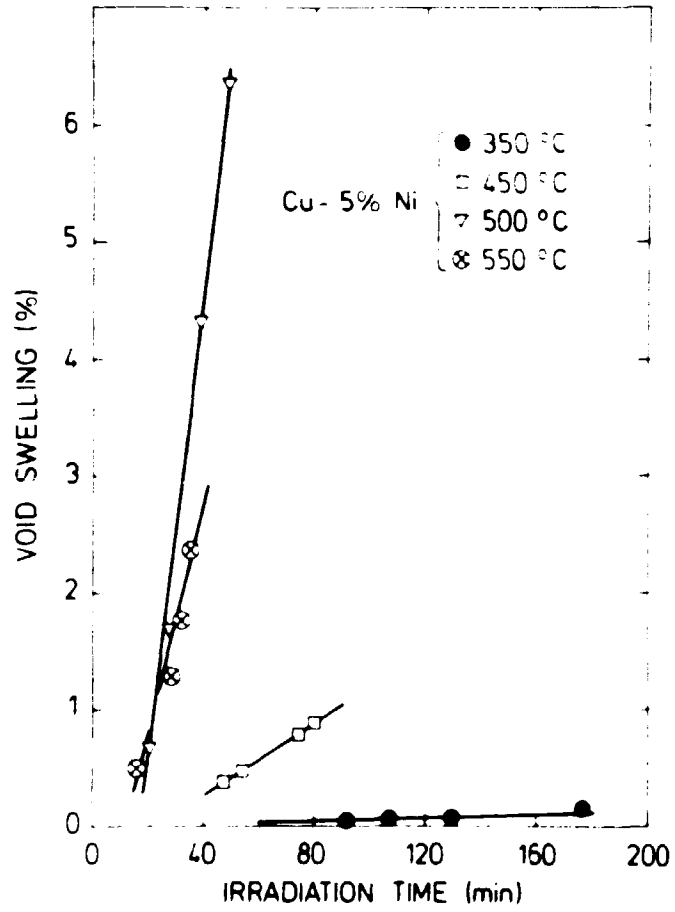
Fig. 4. Void swelling in Cu-5%Ni versus irradiation time for different irradiation temperatures.
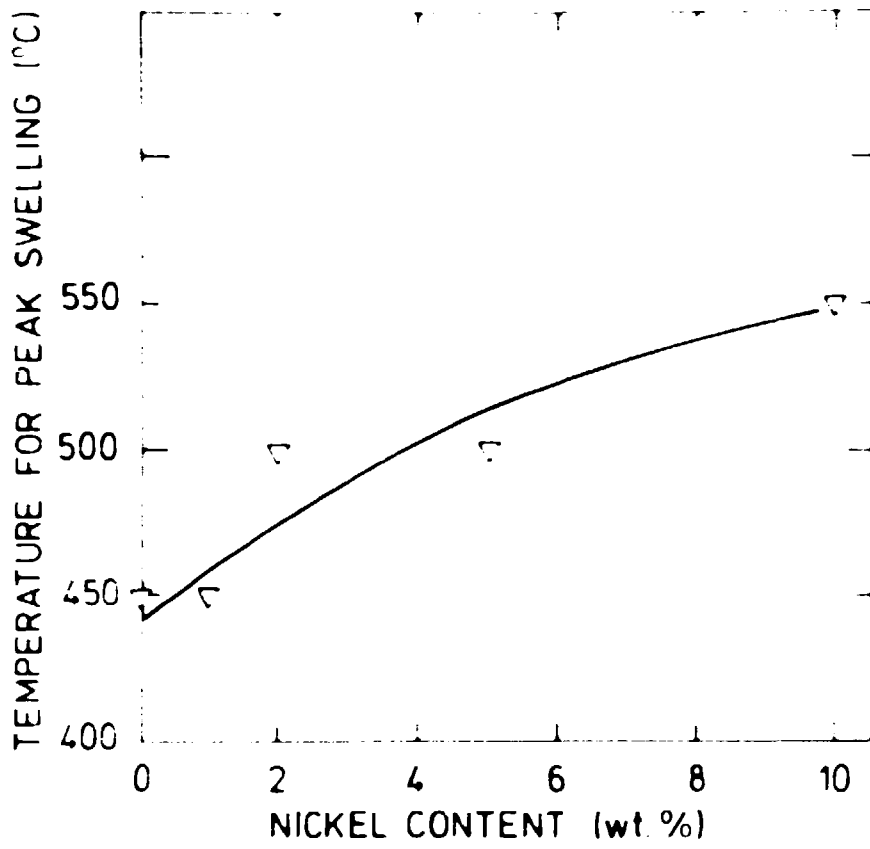
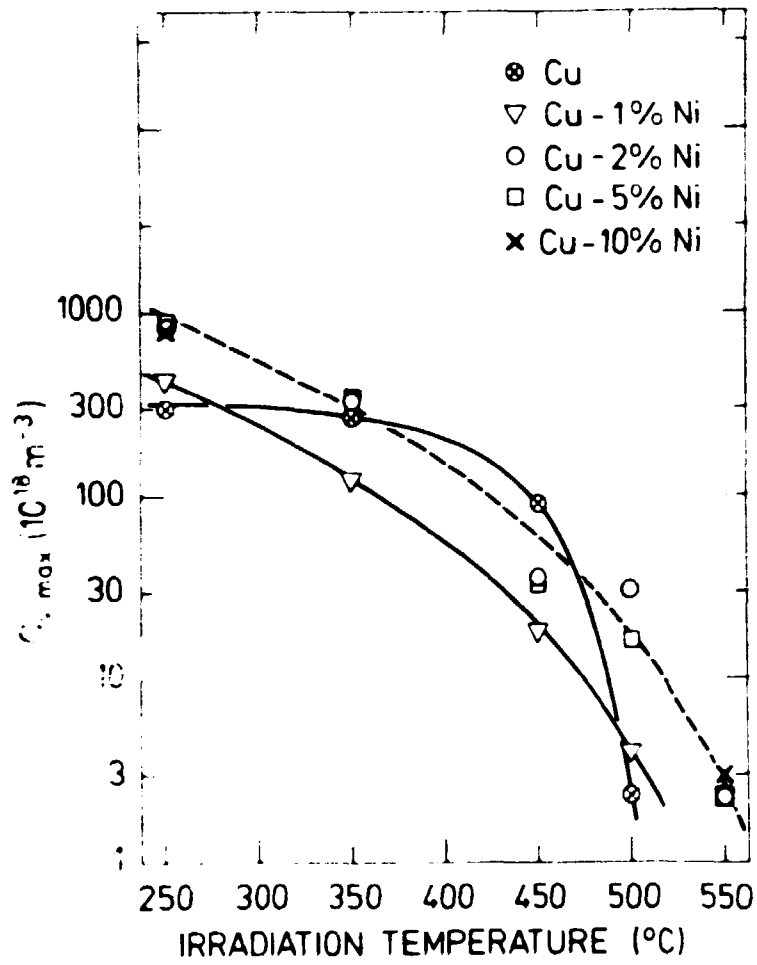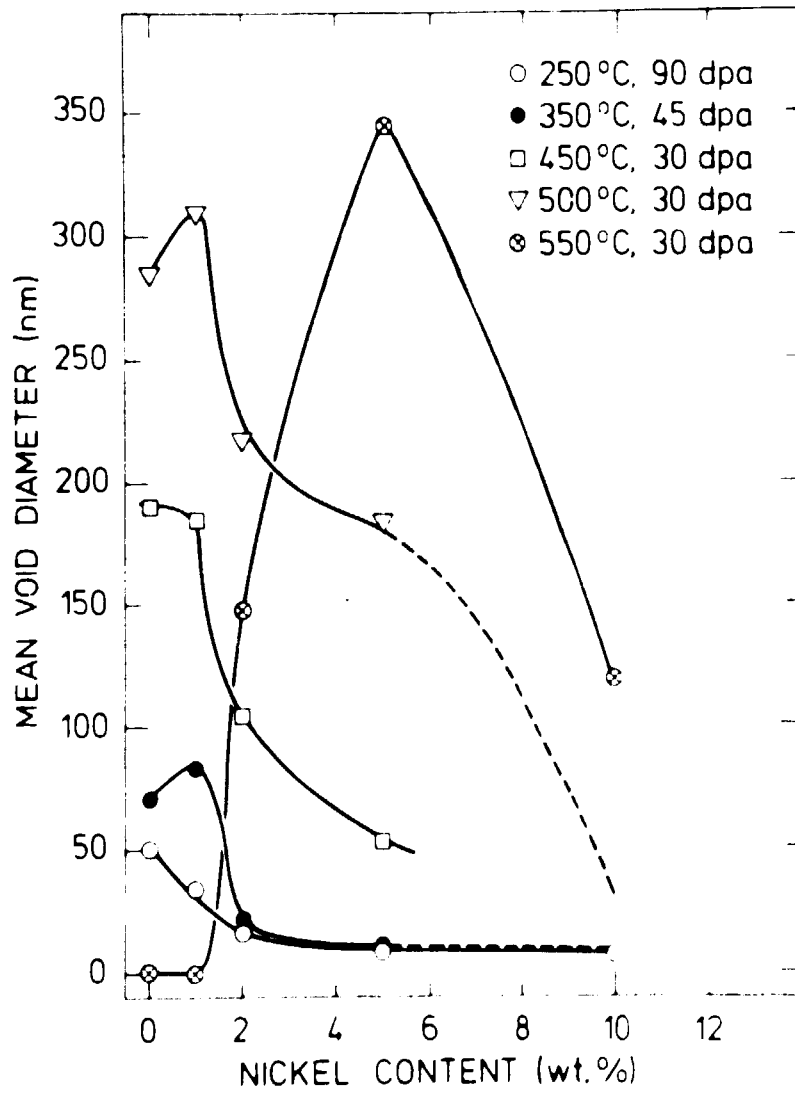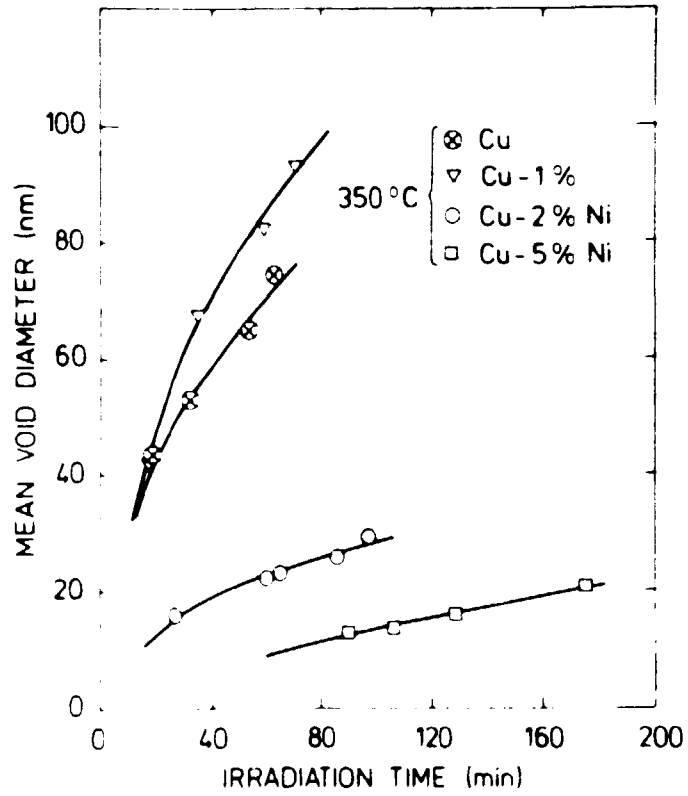Fig. 5. Peak swelling temperature as a function of Ni content.

Fig. 6. Maximum void number density versus irradiation temper-
ature for the different alloy compositions. The dotted
curve represents the alloys with Ni contents of 2% and
more.

Fig. 7. Mean void diameter at specific doses as a function of Ni content for the different irradiation temperatures.

Fig. 8.   Mean void diameter at 350°C versus irradiation time
for different alloy compositions.
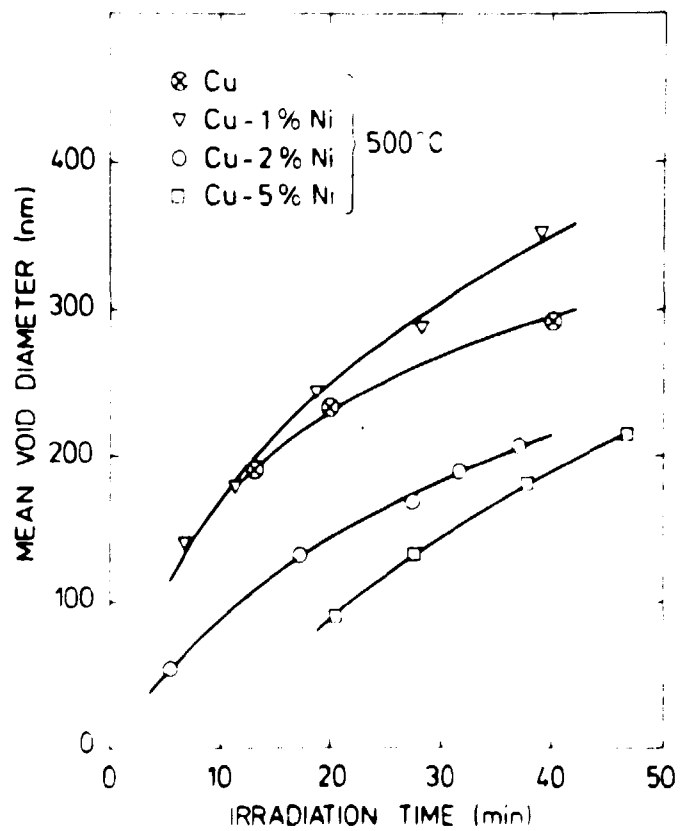
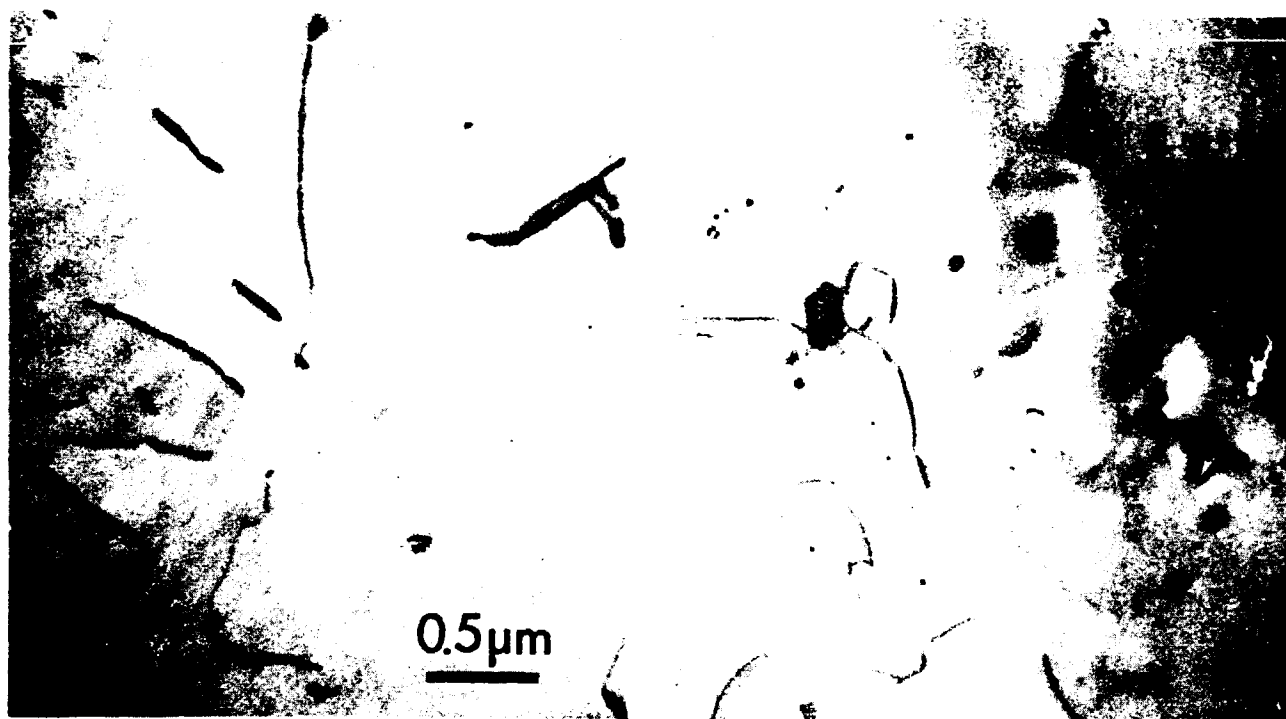Fig. 9.   Mean void diameter at 500°C versus irradiation time
for different alloy compositions.
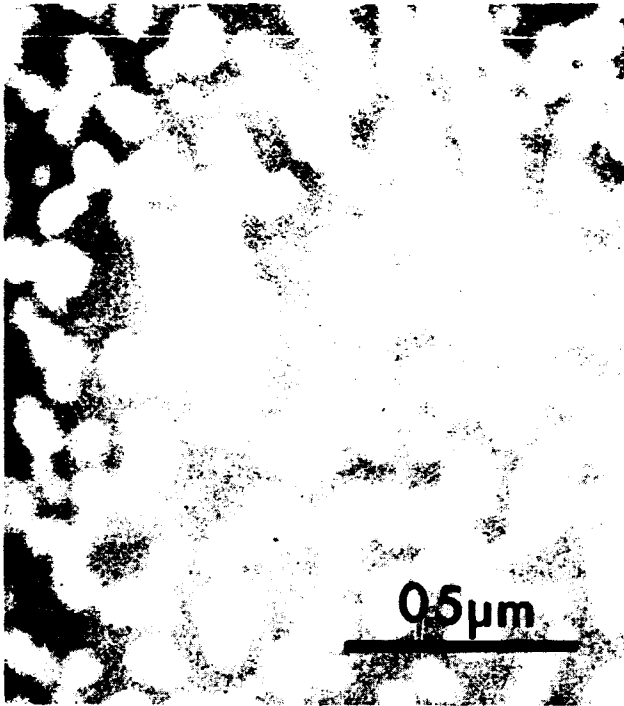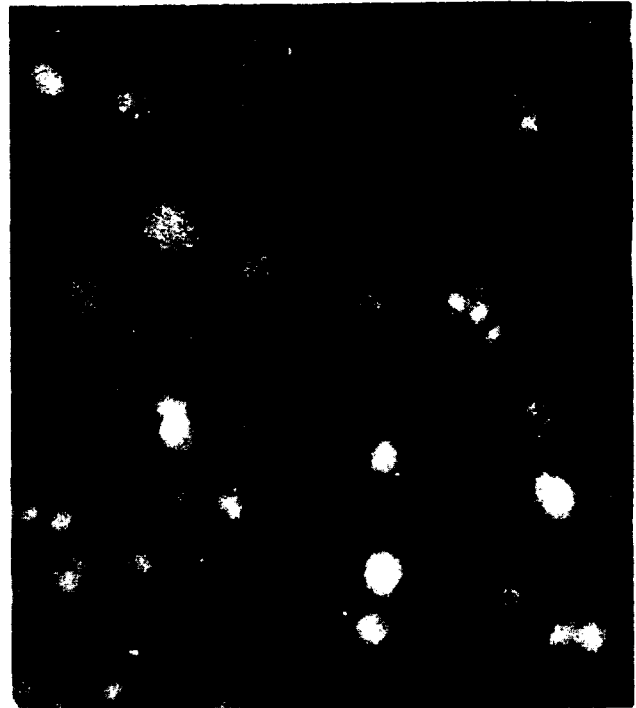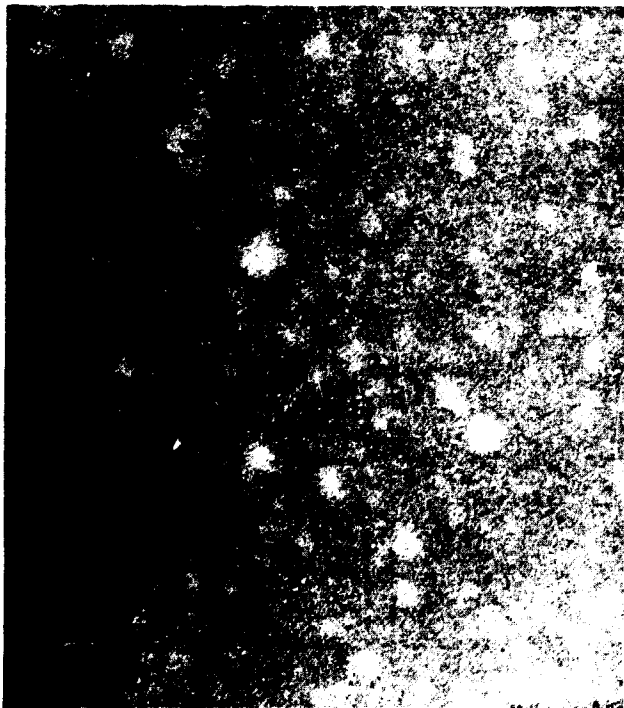
Fig. 10. Void micrographs for copper and Cu-Ni alloys irradiated at 350°C. a) pure copper (57 dpa); b) Cu-1Ni (53 dpa); c) Cu-2Ni (55 dpa); d) Cu-5Ni (57 dpa).

Fig. 11.   Micrographs in dislocation contrast for copper and
            Cu-Ni alloys irradiated at 350°C. a) pure copper
            (6 dpa); b) Cu-18Ni (7 dpa); c) Cu-2.Ni (9 dpa);
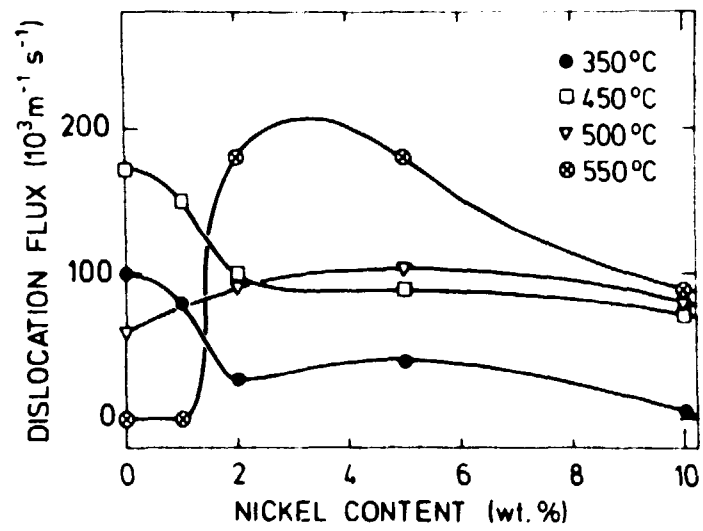   d) Cu-58Ni (7 dpa).

Fig. 12.   Dislocation flux **versus** Ni content at different
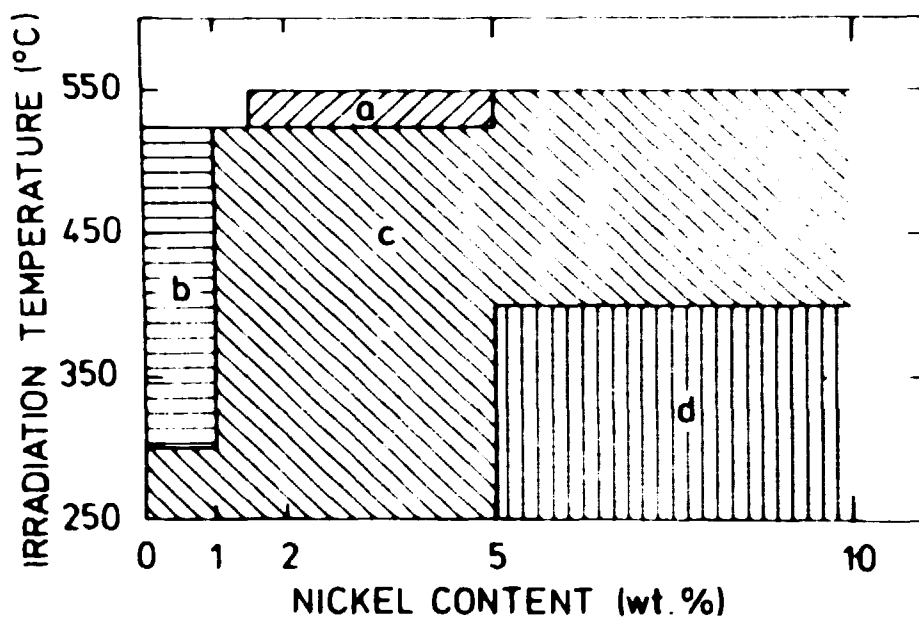   irradiation temperatures.

Fig. 13. The two-dimensional alloy-composition/irradiation-
-temperature space subdivided into different zones
according to the dependence of void size or void
growth on increases in Ni content. Zone a: size
increases with increasing Ni content. Zone b: only
moderate changes in size with change in Ni content.
Zone c: size decreases with increasing Ni content.
Zone d: size drastically reduced already at a Ni
content of 5%, further reduction with increasing Ni
content difficult to measure.