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THE ACTIVATION ENERGY FOR LOOP GROWTH IN Cu AND Cu-Ni ALLOYS

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Abstract

The apparent activation energy for the growth of interstitial dislocation loops in copper, Cu-1%Ni, Cu-2%Ni, and Cu-5%Ni during high voltage electron microscope irradiation was determined. The apparent activation energy for loop growth in all these materials can be taken to be 0.34eV±0.02eV. This value together with the corresponding value of 0.44eV±0.02eV determined earlier for Cu-10%Ni is discussed with reference to the void growth rates observed in these materials. The apparent activation energy for loop growth in copper (and in Cu-1%Ni that has a void growth rate similar to that in pure copper) is interpreted as twice the vacancy migration energy (indicating that divacancies do not play any significant role). For the materials with higher Ni content (in which the void growth rate is much lower than that in Cu and Cu-1%Ni) the measured apparent activation energy is interpreted to be characteristic of loops positioned fairly close to the foil surface and not of loops in "bulk material". From the present results in combination with the earlier results for Cu-10%Ni it is concluded that interstitial trapping is the most likely explanation of the reduced void growth rate in Cu-Ni alloys.

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1. INTRODUCTION

Barlow and Leffers (1977) have determined the apparent activation energy for the growth of interstitial dislocation loops in Cu-10%Ni during HVEM (high voltage electron microscope) irradiation to be 0.43eV±0.02eV. This result was suggested to reflect vacancy binding to clusters of Ni atoms with a binding energy of ~0.3eV. Leffers, Singh, and Barlow (1977) have pointed out that an alternative interpretation in terms of an interstitial binding energy of ~0.9eV to the Ni clusters is possible. Leffers et al. and Singh, Leffers, and Barlow (1978) used this vacancy or interstitial binding to explain the reduced void growth rate in Cu-Ni alloys: the clusters of Ni atoms were considered to act as recombination centres by trapping either vacancies or interstitials.

It is the aim of the present work to extend the investigation of point-defect binding in Cu-Ni by the addition of loop-growth experiments in Cu-Ni alloys with Ni contents below 10%. The new results will be interpreted (and the results on Cu-10%Ni reinterpreted) in the light of an investigation of the relation between the apparent activation energy for loop growth during HVEM irradiation and the activation energy for point-defect migration (Leffers and Singh 1978). The question whether interstitial or vacancy trapping is responsible for the reduced void growth in Cu-Ni allys will be considered in view of this new knowledge.

2. EXPERIMENTAL PROCEDURES

The materials used were copper of 99.999% purity and copper-nickel alloys containing 1, 2, and 5% nickel (by weight) produced from copper and nickel of 99.999% purity. Barlow and Leffers (1977) have given a more detailed description of the materials.

All materials were vacuum-annealed for 4 hours at 800°C before specimens for electron microscopy were made. The thin foil specimens were prepared from 3 mm discs by electropolishing (with jet) in a solution of 33% nitric acid in methanol at -20°C and 10V.

The experiments were made in the AEI EM7 microscope at Harwell operated at 1MV with a dose rate of about 10^{-2}dpa per second ($\sim 2 \cdot 10^{24}$ electrons m^{-2} s^{-1}). The nominal irradiation temperature was controlled to within ±2°C. The actual irradiation temperature is estimated to be within 10°C of the nominal temperature. During irradiation a vacuum of about $5 \cdot 10^{-7}$ torr was maintained in the specimen chamber.

The loop growth was recorded on cine film taken from the screen at a speed of one frame every 2 seconds. The screen was tilted 30° from the position perpendicular to the beam. This introduced some distortion in the images of the disloation loops; the loop growth rates quoted are corrected so that they correspond to a non-tilted screen (the correction depending upon the angle between the tilt axis of the screen and the direction in which the loop dimension is measured).

In Cu-10%Ni Barlow and Leffers (1977) measured the temperature dependence of the growth rate of the "rectangular" loops (with Burgers vector $a[100]$, habit plane (100), and line vectors [011] and [01$\bar{1}$]) emitted from one specific dislocation climb source. In the present work we could in practice only use rectangular loops emitted from climb sources in Cu-5%Ni and Cu-2%Ni: there were very few rectangular loops and climb sources in Cu-1%Ni and none in Cu. In Cu-1%Ni and in Cu we therefore had to use Frank loops for the measurements. The use of these two different procedures (based on rectangular loops and Frank loops, respectively) is justified by the finding of Barlow and Leffers that there is no significant difference between the apparent activation energy for growth of rectangular loops and Frank loops.

In Cu-5%Ni and Cu-2%Ni we compared the activation-energy plots obtained from different climb sources close to one another (foil thickness ~0.5 μm, {100} approximately parallel to the foil), each operating at the different temperatures. Normally the points fell on the same line, i.e. the conditions (e.g. position in the foil) were similar for most climb sources in a given area. In Cu-5%Ni and Cu-2%Ni new loops were continually emitted from the slimb sources in the temperature range 325--500°C (particularly when the area to be investigated was initially irradiated in the temperature range 400-500°C). In Cu-1%Ni and Cu, on the other hand, there was not much loop nucleation after the initial stage of irradiation. It was therefore necessary, as the first stage of each experiment, to necleate a number of loops in a certain area by irradiation at about 250-300°C; we then recorded the growth of these loops at various temperatures (~250-400°C). This procedure had the disadvantage that we could not keep irradiating exactly the same area because the loop would grow out to the surface and disappear. Thus, one activation--energy plot refers to loops in different (closely spaced) areas, which tends to introduce somewhat more experimental scatter than in the case of Cu-5%Ni and Cu-2%Ni.

The exact procedure adopted for the determination of the apparent activation energy for loop growth was the following:

The growth of the loops in a small area (of the order of a few μm in Cu-5%Ni and Cu-2%Ni, somewhat larger in Cu-1%Ni and

Cu) with foil thickness ~0.5 μm was recorded at different temperatures. The dose rate was kept constant in a given experiment, and the loop for which the growth rate was to be recorded was always brought to the central part of the beam. At a given temperature the great majority of the loops in a given area had growth rates close to one another; we used the average values of these growth rates. The logarithms of the average growth rates ($\dot{L}$) were plotted versus 1/kT (k being the Boltzmann constant and T the irradiation temperature in K). The apparent activation energy was found as the absolute slope of the line through those points. The best line through the points was found by a least-square fit that also gave the standard error on the slope.

In Cu-5%Ni and Cu-2%Ni we measured the growth along one of the edges of rectangular loops in foils with {100} approximately parallel to the foil, which corresponds to a climb direction approximately parallel to the foil surface (as reflected in a constant growth rate with time for a given loop, cf. fig. 1). In Cu-1%Ni and Cu we measured the growth along the stacking-fault fringes of the Frank loops, which again corresponds to a growth direction parallel to the foil surface; the growth rate in this direction was constant with time, cf. fig. 2, whereas the growth rate for a given loop in the direction perpendicular to the stacking-fault fringes decreased with time - as reflected in an increasing elongation with increasing size. The selected micrographs in figs. 3 and 4 of rectangular loops and Frank loops in Cu-5%Ni and in Cu, respectively (taken from the cine film) illustrate the behaviour of the two types of loops: the shape of the rectangular loops does not depend on size, whereas the Frank loops become increasingly elongated as they grow in size.

3. EXPERIMENTAL RESULTS

Plots of $\ln\dot{L}$ (logarithmic loop growth rate) versus 1/kT for the various experiments are shown in fig. 5. The lines for 0.5 μm foil of Cu-10%Ni from Barlow and Leffers (1977) are also shown (without individual points). The apparent activation energies E (absolute values of the slopes from the present experiments and from those of Barlow and Leffers on Cu-10%Ni (calculated in the same way as the present results) are listed in table I with the corresponding standard deviations.

Table I shows that the apparent activation energies for loop growth for all the materials with Ni contents of 5% or less can be represented by one value of the order of 0.34eV, whereas the activation energy in Cu-10%Ni is significantly different (~0.44eV*).

4. DISCUSSION

Kiritani, Yoshida, Takata, and Maehara (1975) found an apparent activation energy for loop growth in Cu of ~0.30ev ±0.02eV. Theoretically they deduced that the apparent activation energy should be 2 times the vacancy migration energy E_V^M. The best estimate of E_V^M seems to be ~0.7eV (Bourassa and Lengeler (1976), Wright and Evans (1966), and Antesberger, Sonnenberg, and Wienhold (1978) found E_V^M values of 0.72eV, 0.71eV, and 0.70eV, respectively), which means that the apparent activation energy measured by Kiritani et al. was lower than 1/2 E_V^M. Kiritani et al. acribed this discrepancy to the presence of divacancies with lower migration energy than that of monovacancies. However,

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* The value of 0.43eV quoted by Barlow and Leffers was estimated directly from the plot

Cu-1%Ni) is just about big enough to account for some reduction in swelling via vacancy or interstitial trapping (cf. Mansur, Yoo, and Coglan 1977), but it cannot account for the drastic reduction observed. In the cases of Cu-5%Ni and Cu-2%Ni there is also a very pronounced decrease in swelling, but the measured E values are not significantly different from those in Cu and Cu-1%Ni. This must mean that the present E values especially for Cu-5%Ni and Cu-2%Ni, but also for Cu-10%Ni, are not representative for the interior of the thicker foils where the voids are found. This whole problem is being investigated in more details.

If the effect of the Ni clusters is to trap vacancies, even a slight binding/trapping would be detected as a slight increase in E: loop growth in copper is governed by vacancy migration (Kiritani et al. 1975), and vacancy binding will cause an immediate increase in E. The thin foils investigated in the present work should then be unrepresentative of the void--relevant thicker foils to the extent that the Ni clusters (the presence of which are demonstrated by the operation of the climb sources, cf. Barlow and Leffers (1977)) should not produce any vacancy binding in the thin foils.

If, on the other hand, the effect of the Ni clusters is to trap interstitials, such binding/trapping would not change E until it has become so strong that E_I^M is bigger than E_V^M, i.e. until the binding energy reaches a magnitude of ~0.6eV. In the case of interstitial trapping the thin foils of Cu-5%Ni and Cu-2%Ni would thus only have to be unrepresentative of thicker foils in the quantitative way that E_I^M is still smaller than E_V^M - and not in the qualitative way that there is no binding.

Of these two possibilities the latter, i.e. that the Ni clusters present in Cu-5%Ni and Cu-2%Ni do produce (interstitial) binding also in the thin foils, but not enough to make E_I^M bigger than E_V^M, seems far more likely than the former that the Ni clusters do not produce any binding in thin foils.

We therefore consider the present results to support the suggestion that the reduction in swelling in Cu-Ni is produced by interstitial trapping at the Ni clusters - with the addition that the concentration of Ni clusters (or some other parameter for the Ni clusters) in the thin foils of Cu-5%Ni and Cu-2%Ni investigated in the present work is insufficient to produce the interstitial binding energy of ~0.6eV necessary to change the apparent activation energy for loop growth.

REFERENCES

Antesberger, G., Sonnenberg, K., and Wienhold, P., 1978, J. Nucl. Mat. 69 & 70, 660.

Antesberger, G., Sonnenberg, K., Wienhold, P., Coltman, R.R., Klabunde, C.E., Williams, J.M., and Chaplin, R.L., 1975, Fundamental Aspects of Radiation Damage in Metals, edited by M.T. Robinson and F.W. Young (U.S. ERDA Conference No. 751006) p. 561.

Barlow, P., and Leffers, T., 1977, Phil. Mag. 36, 565.

Bourassa, R.R., and Lengeler, B., 1976, J. Phys. F, 6, 1405.

Kiritani, M., Yoshida, N., Takata, H., and Maehara, Y., 1975, 5 J. Phys. Soc. Japan, 38, 1677.

Leffers, T., and Singh, B.N., 1978, Risø-M-2142 (to be published).

Leffers, T., Singh, B.N., and Barlow, P., 1977, Risø-M-1937.

Mansur, L.K., Yoo, M.H., and Coglan, W.A., 1977, Trans. Am. Nucl. Soc. 27, 332.

Singh, B.N., Leffers, T., and Barlow, P., 1978, Proceedings of f the Fifth International Conference on High Voltage Electron Microscopy, Kyoto, p. 581.

Wright, P., and Evans, J.H., 1966, Phil. Mag., 13, 521.

Table I

Apparent activation energy for loop growth (E) from the different experiments.

Material	E(eV)
Cu	0.35±0.02
Cu-1%Ni	0.34±0.03
Cu-2%Ni	0.34±0.01
Cu-2%Ni	0.36±0.01
Cu-5%Ni	0.34±0.02
Cu-5%Ni	0.33±0.02
Cu-10%Ni*	0.41±0.01
Cu-10%Ni*	0.45±0.02
Cu-10%Ni*	0.45±0.02
Cu-10%Ni*	0.44±0.02

*from Barlow and Leffers (1977)

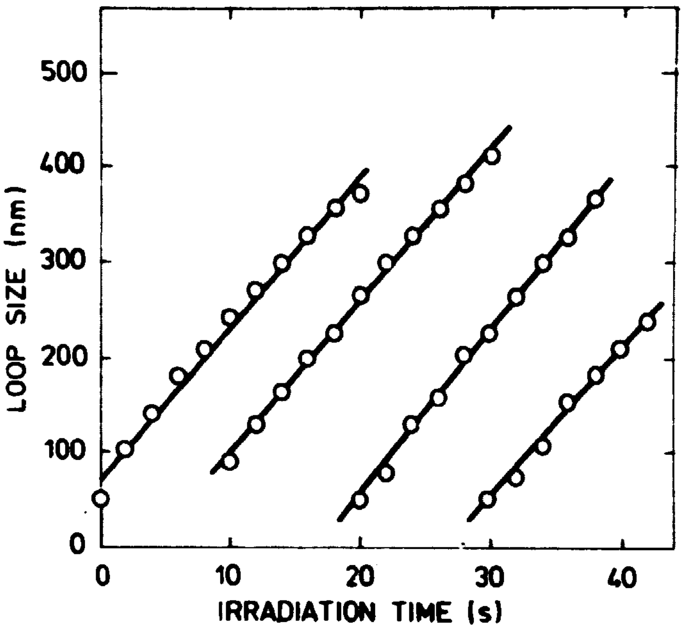

Fig. 1. The growth of four rectangular loops in Cu-5%Ni at 421°C corrected for the effect of the tilted screen. The starting points are arbitrarily plotted with intervals of 10 seconds.

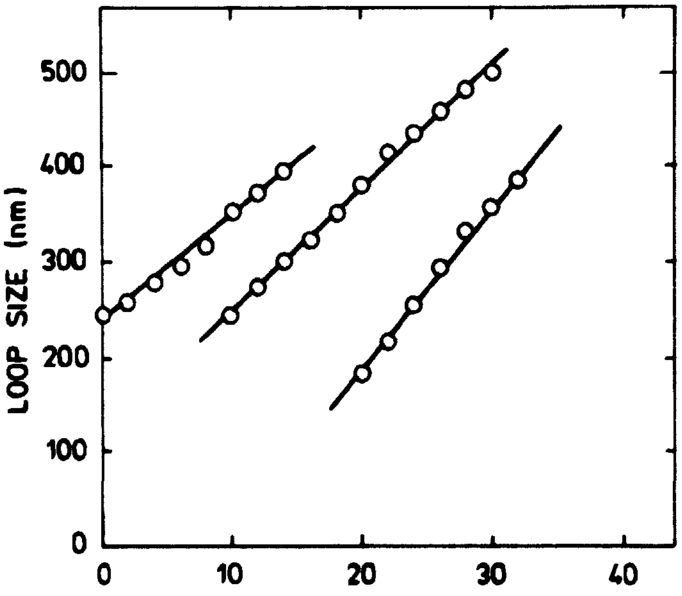

Fig. 2. The growth along the stacking-fault fringes of three Frank loops in copper at 400°C corrected for the effect of the tilted screen. The starting points are arbitrarily plotted with intervals of 10 seconds.

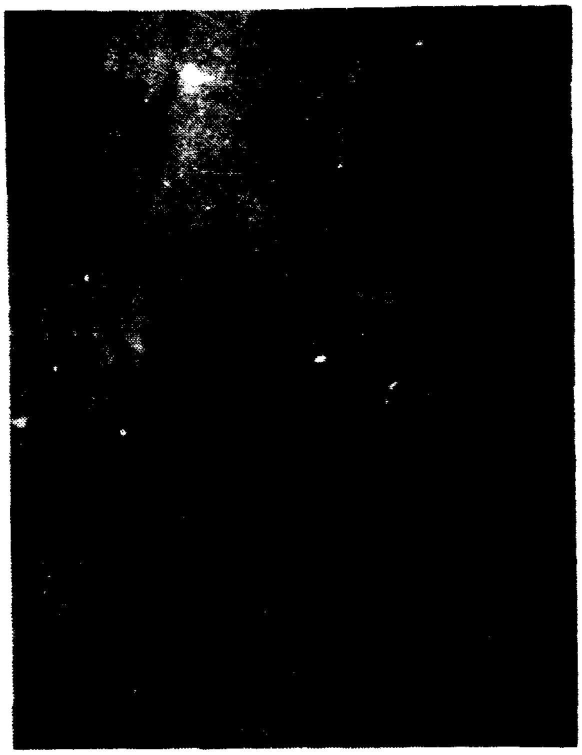

Fig. 3. A number of rectangular loops in Cu-5%Ni irradiated at 480°C. The shape of the loops (distorted by the tilt of the screen) does not depend on loop size. The distance between two markers along the tilt axis of the screen (horizontal) corresponds to o.35 μm.

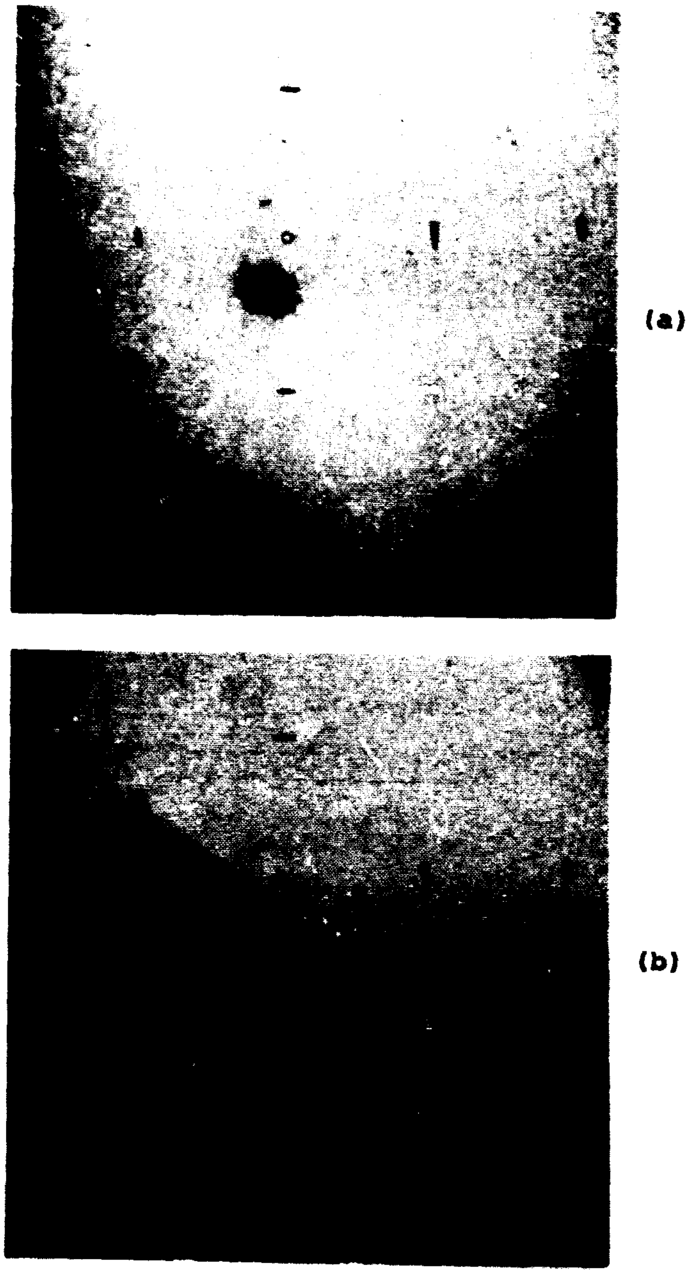

Fig. 4. Two Frank loops in copper irradiated at 355°C at different stages of their growth (the time elapsed between (a) and (b) is 58 seconds). The loops become increasingly elongated along the stacking-fault fringes with increasing size. The distance between two markers along the tilt axis of the screen (horizontal) corresponds to 0.35 μm.

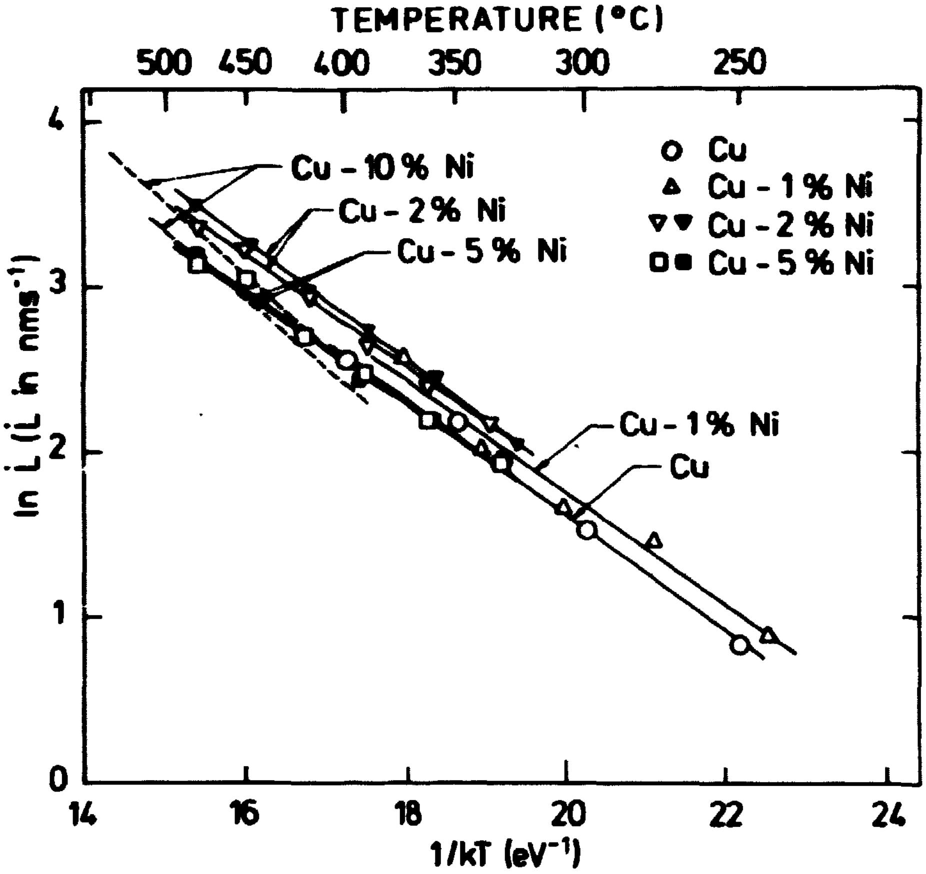

Fig. 5. Logarithmic loop growth rates versus 1/kT for the various experiments. The best line for each experiment is drawn according to a least-square fit. The corresponding lines for 0.5 µm foil of Cu-10%Ni from Barlow and Leffers (1977) are also shown (dotted lines without points).