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A STUDY OF THE VACANCY LOOP FORMATION PROBABILITY IN Ni-Cu AND Ag-Pd ALLOYS.

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Abstract

The molten-zone model of vacancy loop formation from a displacement cascade predicts that the loop formation probability should scale with the melting temperature. To investigate this possibility the vacancy loop formation probability has been determined in a series of Cu-Ni and Ag-Pd alloys. The irradiations were performed at room temperature with 50 keV Kr^+ ions and the resulting damage structure was examined by using transmission electron microscopy. In the Cu-Ni alloy series, the change in loop formation probability with increasing Ni concentration was complex, and at low- and high- nickel concentrations, the defect yield did not change in the predicted manner. The defect yield was higher in the Cu-rich alloys than in the Ni-rich alloys. In the Ag-Pd alloy the change in the loop formation probability followed more closely the change in melting temperature, but no simple relationship was determined.

1. Introduction

The molecular dynamic computer simulations of low energy displacement cascades in Cu and Ni have shown that a portion of the cascade exists in a molten state[1-7]. Based on these simulations, it was proposed that the lower loop formation probability in Ni compared to in Cu (i.e., 0.33[8] in Ni compared to 0.5 in Cu[8]) was a consequence of the faster solidification rate of the molten zone in Ni[2-4]. The difference in the solidification rate was attributed to the higher melting temperature of Ni. Later, Flynn and Averback[9] suggested that strong coupling between the electron and phonon systems would increase the solidification rate and that this effect would be more important in Ni than in Cu.

To investigate the dependence of the loop formation probability on the melting temperature, the damage produced by room temperature 50 keV Kr^+ ion irradiations in a series of Cu- Ni and Ag-Pd alloys was studied. These alloy systems

were selected since solid solution alloys are formed over the entire composition range and the elements have a similar mass. Consequently , the cascade dynamics are expected to be similar in each alloy system. In addition, the melting temperature of the alloys extends over a wide range. In the Cu- Ni system, the melting temperature increases from 1356 K (Cu) to 1726 K (Ni) and in the Ag-Pd system it increases from 1234 K (Ag) to 1827 K (Pd). Thus, the melting temperature can be varied systematically so that the dependence of the loop formation probability on the melting temperature can be assessed. The results of this study are presented in this paper.

2 Experimental procedures

Electron transparent TEM discs made from Cu-Ni and Ag-Pd alloys were irradiated at room temperature with 50 keV Kr^+ ions to ion doses of either 2 or 5 x 10^{11} ions cm^{-2}. The need for accurate ion dosimetry was avoided by irradiating discs of each alloy system in the same batch. However, the values of the defect yield obtained from samples irradiated in different batches were within experimental error, indicating that the results from different ion irradiations could be compared. The defect yield and the defect image size were used to quantify the damage structure. The defect yield, which is defined as the fraction of displacement cascades that produce visible defects, was determined from images formed with a 200-type diffraction vector. The image size was measured as the length of the interface between the black and white lobe in black/white lobe images and as the maximum dimension in black dot images. Details of these procedures have been described elsewhere[10].

3. Results

The difference in the damage structure produced by the irradiation of the Cu-Ni alloys can be seen qualitatively in the series of micrographs presented in Figure 1. Clearly, the areal defect density and the defect size decrease with increasing Ni content. The change in the defect density as a function of alloy content is quantified in Figure 2 where the defect yield is plotted against the alloy composition. (Also shown in this figure is the change in melting temperature as the solute content increases). The defect yield changes with the alloy melting temperature but not in a simple manner. At Ni levels < 20 wt%, the defect yield and the melting temperature increase, which is contrary to the prediction of the molten zone model. For alloy concentrations between 20 and 90 wt % Ni, the defect yield decreases as the melting temperature increases. However, even in the 60 wt% Ni alloy the defect yield is less than in pure Ni.

The average and the maximum image sizes in the Cu-Ni alloys are reported in Table 1. In general, the average and the maximum defect size decreased with

increasing Ni content.

Micrographs comparing the vacancy loop population produced in the Ag-Pd alloys are presented in Figure 3. It is evident that the defect yield and the defect size decrease with increasing Pd content. This decrease of the defect yield is quantified in Figure 4 where the defect yield is plotted against the alloy composition. For comparison the change in the melting temperature with alloy content is also shown. From Figure 4 it can be seen that the defect yield decreases as the melting temperature increases.

The maximum and average defect size in the Ag-Pd alloys are listed in Table 1. As in the case of the Cu-Ni alloys, the maximum and the average loop size decrease as the alloy melting temperature increases.

An alternative way to express the different response of the alloys is to consider how the number of vacancies retained in a vacancy loop of average size changes. The number of vacancies retained in a loop of average diameter, $N_{v,obs}$, can be calculated for loops on {111} planes from

$$N_{v,obs} = \langle d^2 \rangle \pi / (a^2_o)(3)^{1/2} \qquad (1)$$

In this equation, $\langle d^2 \rangle$ is the root mean square of the loop diameter and a_o is the lattice parameter. The value of the lattice parameter for the different alloys was obtained from ref [11]. In determining this expression no account was taken of the distribution of loop Burgers vectors in the alloys. This assumption was made as only 50% of the loop population could be unambiguously assigned a Burgers vector. The number of vacancies retained in a loop of average size is plotted as a function of alloy content for the Cu-Ni and Ag-Pd alloys in Figure 5. In both systems, the number of vacancies in a loop decreases linearly with increasing melting temperature. The rate of change is greater in the Ag-Pd than in the Cu-Ni system because of the larger difference in the defect yield and in the size of vacancy loops.

Discussion

The model for the formation of a vacancy loop that evolved from the molecular dynamic computer simulations of displacement cascades, predicts that the loop formation probability is dependent on the melting temperature of the material[2-5]. To examine this dependency, the damage structure produced in a series of solid solution Cu-Ni and Ag-Pd alloys was examined. It was shown that the defect yield, the defect size and the number of vacancies retained in a vacancy loop of average size decreased with increasing melting temperature. In the Cu-Ni alloy series, the dependency of the defect yield (or the loop formation probability) on the amount of solute (melting temperature) was complex. For example, the

alloy series, the dependency of the defect yield (or the loop formation probability) on the amount of solute (melting temperature) was complex. For example, the defect yield increased when ≤20 wt% Ni was added, which is contrary to the predicted behavior. This increase in defect yield is consistent with the result of Stathopoulos et al[12] who observed a similar increase in a Cu-10wt%Ni alloy following an irradiation with 30 keV Cu^+ ions. Another incongruity occurs in the Ni-rich alloys, where the defect yield is lower in the alloys than in the pure material. A similar decrease was found by Robinson and Jenkins[13] in their study of the defect yield in Ni and solid solution Ni-Cr alloys. Following a room temperature irradiation with 80 keV Ni^+ ions, the defect yield decreased from 0.44 in Ni to 0.17 in a Ni-17wt%Cr alloy. In the systems where these incongruities occur the melting temperature change caused by the solute addition is +116° for 20% Ni in Cu, - 23° for 10% Cu in Ni and - 30° for 17wt% Cr in Ni. These exceptions to the predicted melting temperature dependency suggest that other solute effects play a role in determining the loop formation probability and that they dominate when the melting temperature difference between the pure material and the binary alloy is small. These other solute effects and their influence on the loop formation probability are discussed in another paper presented at this conference[14] .

Stathopoulos et al[12] studied the dependence of the loop formation probability on solute content in a series of binary Cu alloys. Single phase alloys of Cu-Al, Cu-Ge, Cu-Si, Cu-Zn, Cu-Ni and Cu-Be were irradiated at room temperature with 30 keV Cu^+ or W^+ ions. For both Cu^+ and W^+ ion irradiations, the defect yield increased as the solute content increased. The dependence of the size of the defects on the alloy content was complex, and was dependent on the alloying element, the amount of the element and on the ion species used. They explained the defect yield behavior by solute elements dechannelling ions and planarly channelled knock-ons. These effects will increase the number of defects created, which in turn would increase the defect yield. In all the alloys, except Cu-Ni, the addition of solute decreases the alloy melting temperature. Therefore, these experimental observations are qualitatively in agreement with the molten zone model for loop formation.

One difficulty with the present comparison in the Cu-Ni alloys is that the samples were irradiated at different temperatures relative to Stage III. In copper Stage III is peaked at 280 K whereas in nickel Stage III is at 350 K. Therefore at the irradiation temperature both vacancies and interstitials are mobile in copper, but only interstitials are mobile in nickel. The role that vacancy mobility plays in loop formation from a displacement cascade can be evaluated from diffuse x-ray scattering measurements[15]. This work showed that the number of vacancies in loops increased by 50 % in Cu and Ni on annealing through stage III. If we increase the number of vacancies in nickel by 50 %, the conclusions drawn above still hold but the quantitative results change. Moreover, since the x-ray

measurements were performed on bulk specimens, a smaller than 50% increase is expected in the current experiments where thin samples were employed and in which the nearby free surfaces provide a strong sink for freely migrating vacancies.

In the Ag-Pd alloys, the defect yield, the defect image size and the number of vacancies retained in a vacancy loop decreased with increasing alloy content as predicted by the molten zone model. The dependence on the melting temperature was simpler than in the Cu-Ni alloys and no anomalies were observed. This may be due to the large differences in the melting temperature of the Ag-Pd alloys dominating any solute effects. Complications arising from the any difference in the temperature of Stage III relative to the irradiation temperature could not be assessed as no data on Stage III recovery in Pd was found.

These experimental results on their own do not prove or disprove the molten zone loop formation model. However, when they are considered along with the explanation for other experimental observations (namely, the formation of vacancy loops at temperatures below Stage I [8,10,16,17,18] , the decrease in loop formation probability as the irradiation temperature is decreased [8,10,16, 17,18] and the changes in loop population[8,10,16,17] (i.e., loss, replacement of one loop by another with a different Burgers vector, loop coalescence) and the decrease in defect production rate as the ion dose is increased [8,10,16,17]), a strong case can be made for this loop formation mechanism. In light of the importance of vacancy loop formation to radiation damage processes (swelling, hardening, creep, etc) it may be time to reevaluate previous results in terms of this model.

5. Conclusions

The defect yield, the defect image size and the number of vacancies retained in a dislocation loop of average size has been determined in a series of Cu-Ni and Ag-Pd alloys irradiated at room temperature with 50 keV Kr^+ ions. The dependence of the defect yield on the alloy melting temperature was complicated by the observations in the Cu-Ni system where the defect yield increased rather than decreased with increasing melting temperature for Ni additions of < 20 %. Also it was found that the defect yield was lower in the Ni-rich alloys than in pure Ni. No complex dependencies of the defect yield on the melting temperature were found in the Ag-Pd system. In the Cu-Ni and Ag-Pd alloy systems, the number of vacancies retained in a loop of average size decreased with increasing melting temperature in a simple manner.

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TABLE 1. The average and maximum defect size in the different alloys. The Cu-Ni samples were irradiated with 50 keV Kr^+ ions to an ion dose of 5×10^{11} ions cm^{-2} and the Ag-Pd alloys to an ion dose of 2×10^{11} ions cm^{-2} .

MATERIAL	AVERAGE SIZE nm	MAX. SIZE nm	MATERIAL	AVERAGE SIZE nm	MAX. SIZE nm
Cu	3. 3	10	Ag	6. 01	16
Cu-20%Ni	3. 2	11	Ag-25%Pd	4. 80	12. 6
Cu-40%Ni	3. 0	9	Ag-50%Pd	4. 47	13. 9
Cu-60%Ni	2. 3	7	Ag-75%Pd	3. 87	10. 4
Cu-80%Ni	2. 4	7	Pd*	3. 06	11. 6
Cu-90%Ni	1. 9	5			
Ni**	2. 1	4. 8			

*Data from another irradiation
**Data from Robertson et al[16]

Figure Captions

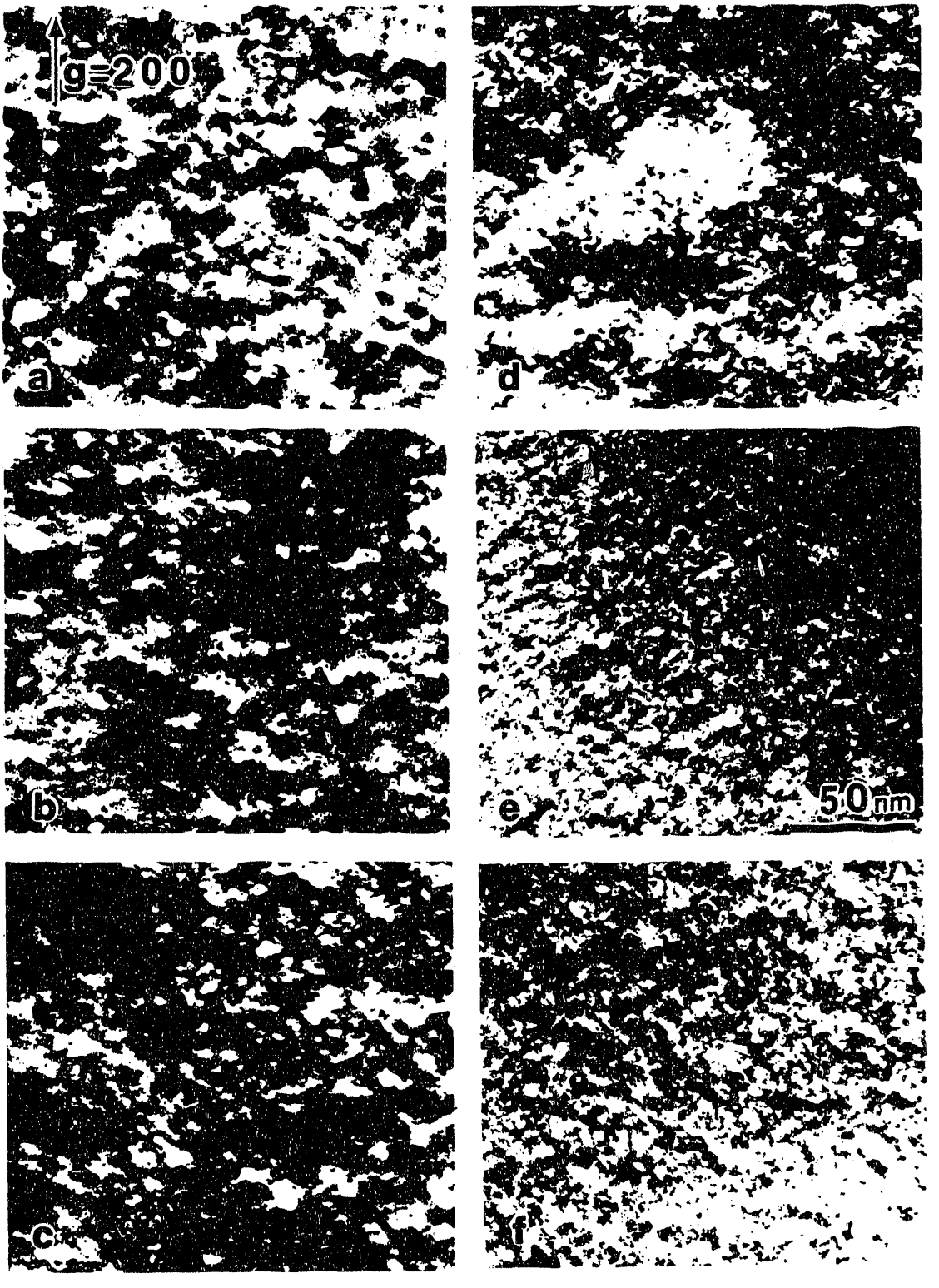
Figure. 1. Dark-field micrographs of the defect structure produced in the Cu-Ni alloy series by a room temperature irradiation with 50 keV Kr^+ ions to a dose of 5 x 10^{11}ions cm^{-2}. (a) Cu, (b) Cu-20%Ni, (c) Cu-40%Ni, (d) Cu-60%Ni, (e) Cu-80%Ni and (f) Cu-90%Ni. g = (002).

Figure 2. Defect density as a function of alloy melting temperature. The samples had been irradiated at room temperature with 50 keV Kr^+ ions to an ion dose of 5 x 10^{11} ions cm^{-2}. The solid line shows the alloy melting temperature.

Figure 3. Dark-field micrographs of the defect structure produced in the Ag-Pd alloy series by a room temperature irradiation with 50 keV Kr^+ ions to a dose of 2 x 10^{11}ions cm^{-2}. (a) Ag, (b), Ag-25%Pd, (c) Ag-50%Pd, (d) Ag-75%Pd and (e) Pd. The pure Pd sample was irradiated at a different time from the other alloys. g = (002).

Figure 4. Defect density as a function of alloy melting temperature for the Ag-Pd alloys. The samples had been irradiated at room temperature with 50 keV Kr^+ ions to an ion dose of 2 x 10^{11} ions cm^{-2}. The solid line shows the alloy melting temperature.

Figure 5. Number of vacancies retained in a dislocation loop as a function of alloy melting temperature. The Cu-Ni samples had been irradiated to a dose of 5 x 10^{11} ions cm^{-2} and the Ag-Pd alloys to a dose of 2 x 10^{11} ions cm^{-2} . The solid and dashed lines show the melting temperature for the Cu-Ni and Ag-Pd alloys, respectively



SMALIEKASKI AL FIGURE 1

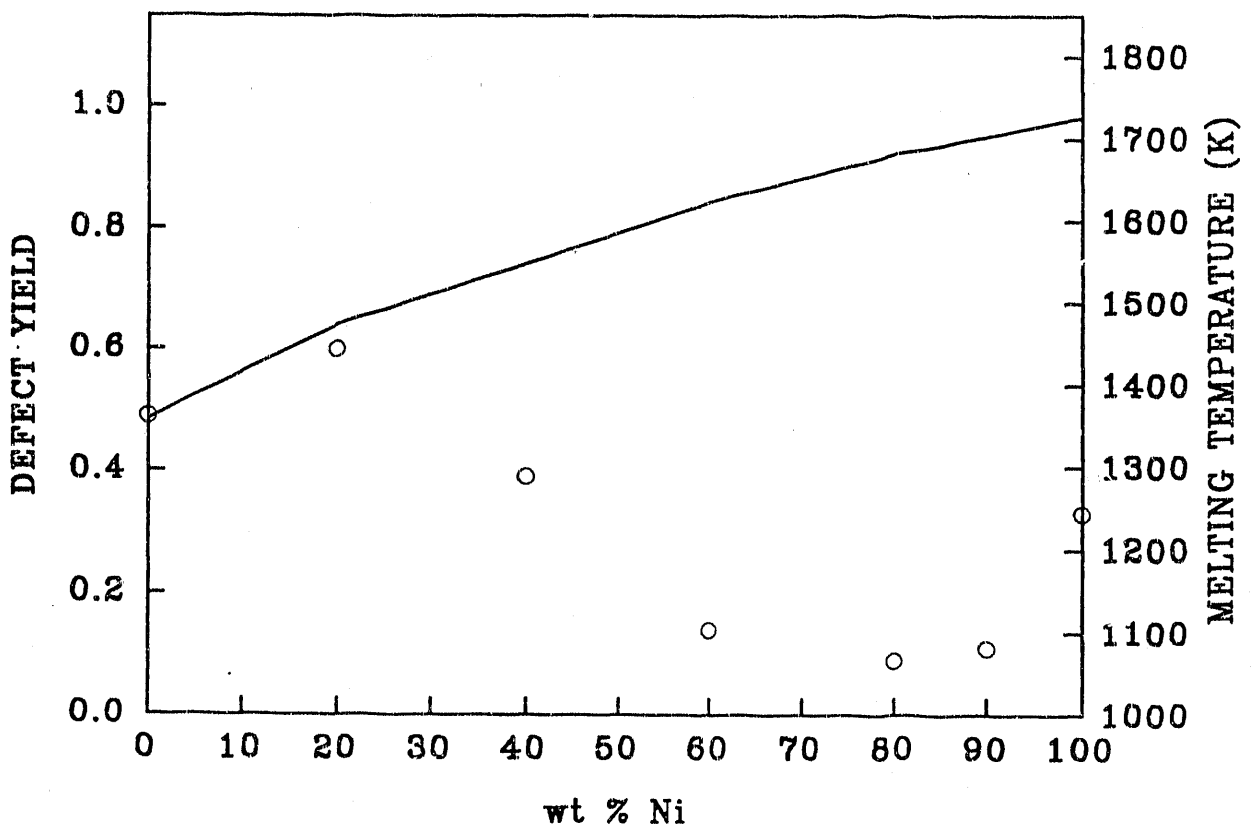
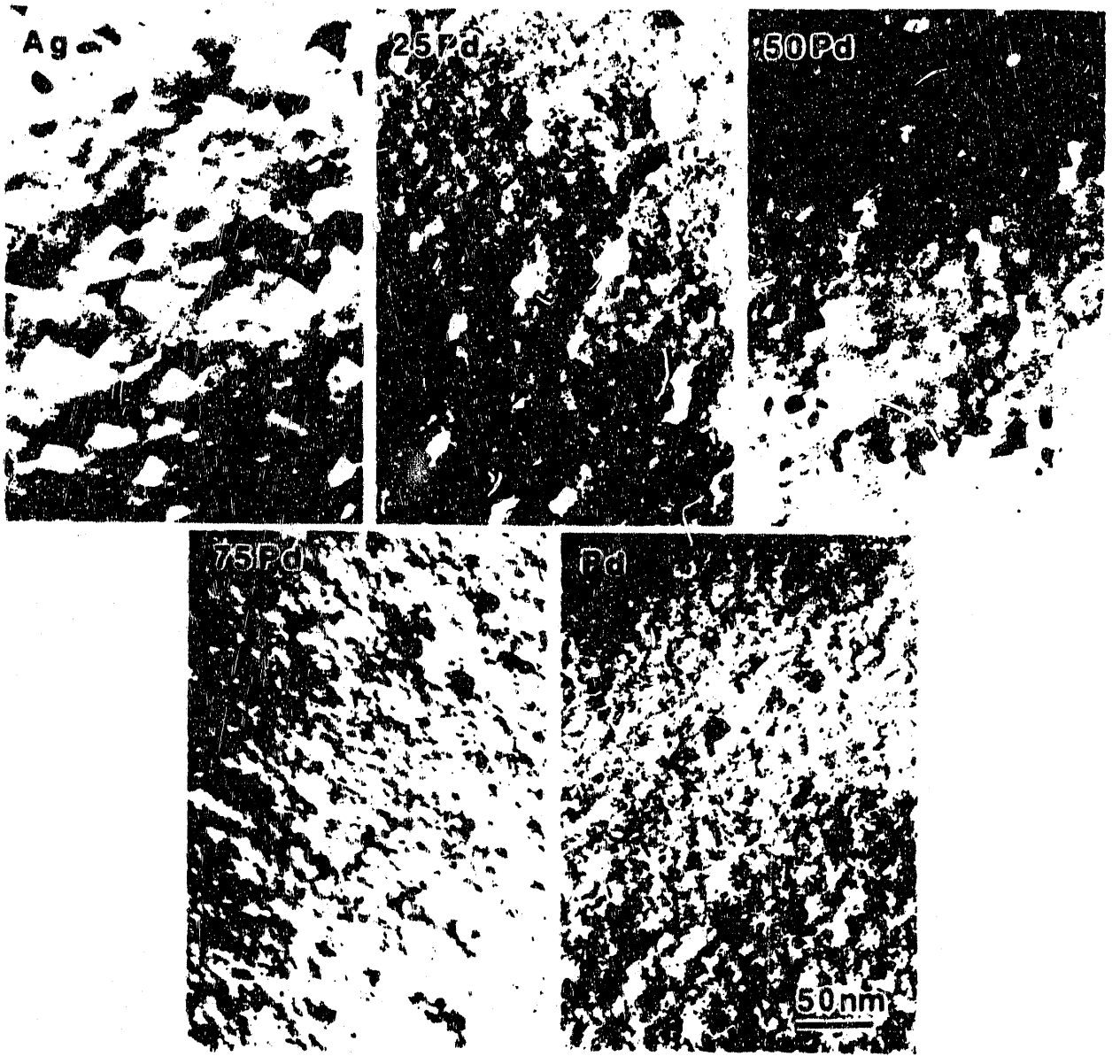

DEFECT YIELD
1.0
0.8
0.6
0.4
0.2
0.0
0
10
20
30
40
50
60
70
80
90
100
wt % Ni
1800
1700
1600
1500
1400
1300
1200
1100
1000
MELTING TEMPERATURE (K)

Fig 2


Ag
25Pd
50Pd
75Pd
Pd
50nm

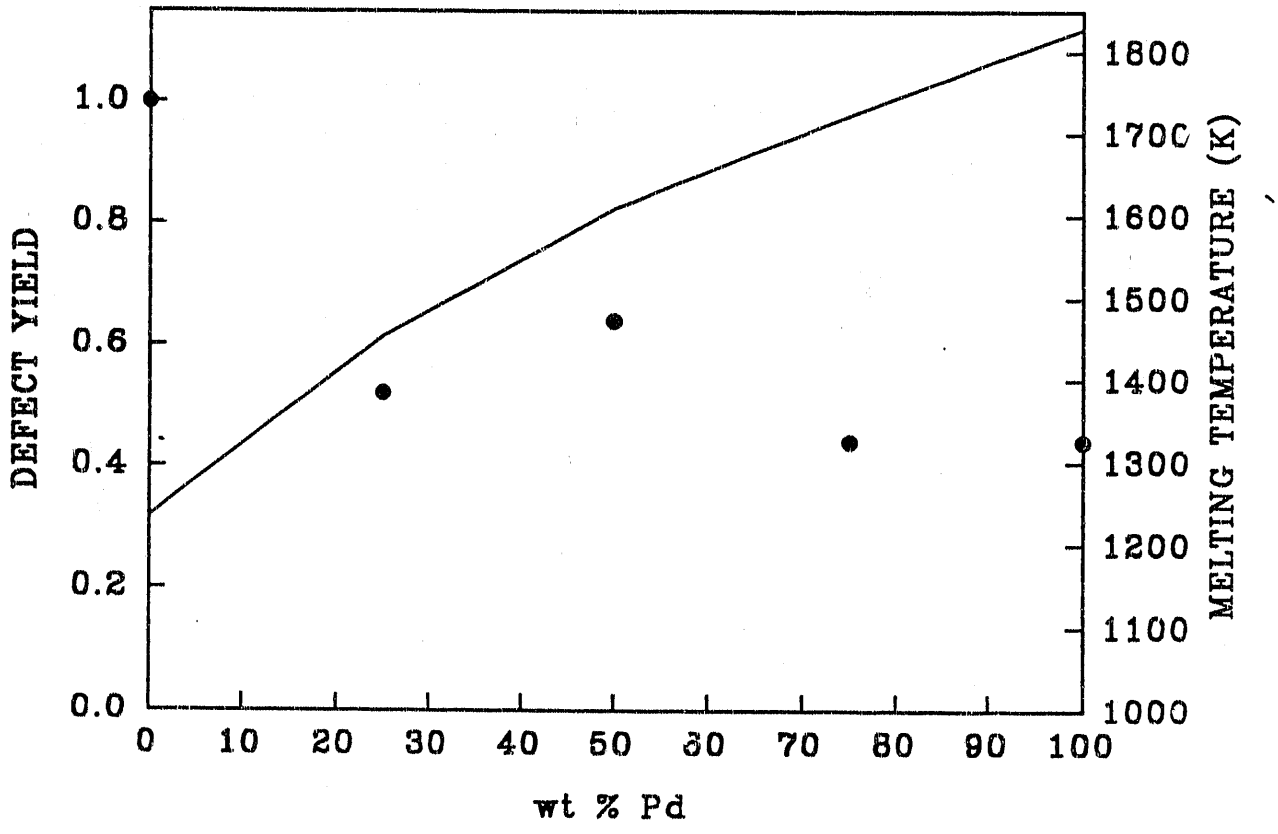
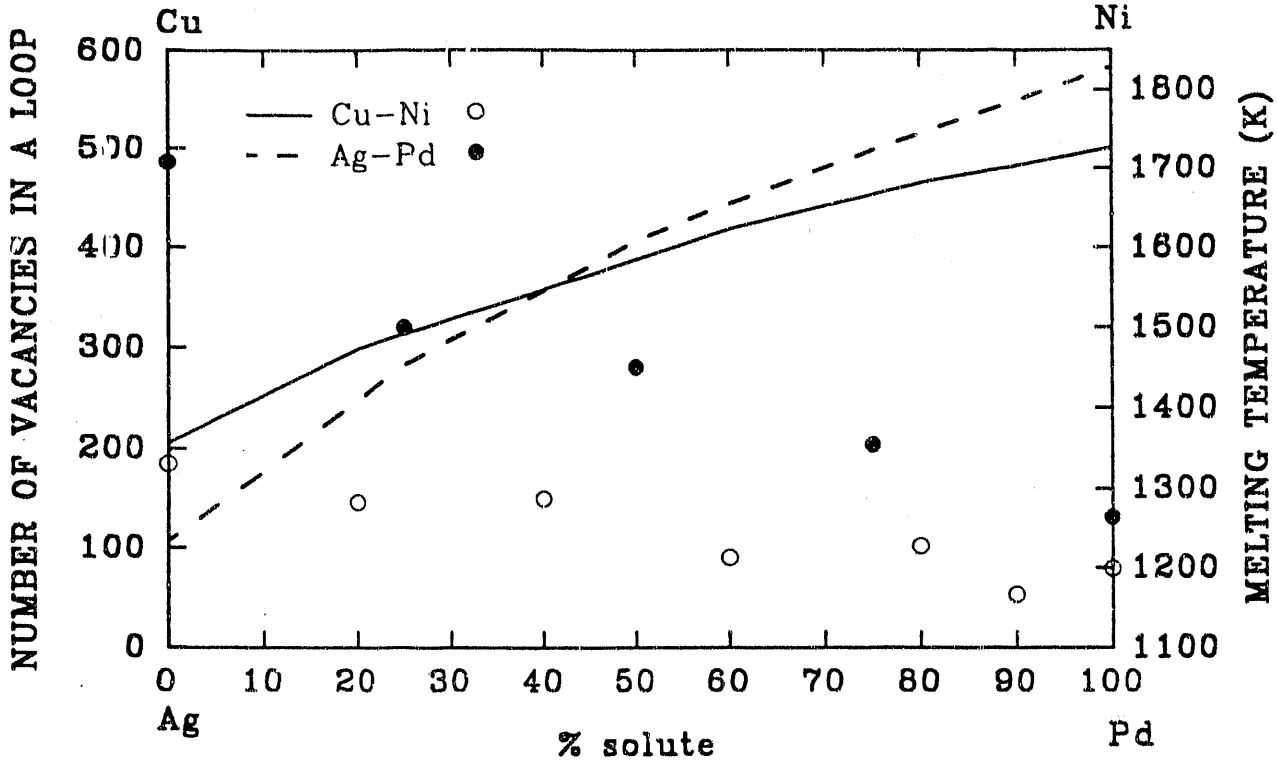

DEFECT YIELD
1.0
0.8
0.6
0.4
0.2
0.0
0
10
20
30
40
50
60
70
80
90
100
wt % Pd
1800
1700
1600
1500
1400
1300
1200
1100
1000
MELTING TEMPERATURE (K)

Figure 4.



SMALINSKAS et al Figure 1

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