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Directional electromigration-enhanced interdiffusion in the Cu–Ni system

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The effect of a dc on the interdiffusivity \tilde{D} in the Cu–Ni system was investigated over the temperature range of 650–850 °C and at current densities in the range of 0–1000 A cm⁻². Interdiffusivities were calculated using the Sauer–Freise–den Broeder method and the values calculated in the absence of a current were in agreement with previously published results. The influence of the current on \tilde{D} depended on its direction relative to the two interfaces in the trilayered Cu–Ni–Cu samples. When the electronic flow was from Ni to Cu (cocurrent interface), the interdiffusivity showed a marked increase relative to copper content but was unchanged when the electronic flow was from Cu to Ni (countercurrent interface). The increase of \tilde{D} in the cocurrent interface depended on concentration and temperature. At lower temperatures, the increase becomes significant at higher copper concentrations but for the same value of current density, the increase is apparent at lower concentrations. The effective activation energy of interdiffusivity depended on concentration and decreased with the application of a current. The decrease was largest for higher copper concentrations. The results are interpreted in terms of a proposed vacancy-atom interaction for copper with the implication that the electron wind effect on Cu is counteracted by the effect of vacancies. © 2007 American Institute of Physics. [DOI: 10.1063/1.2809444]

I. INTRODUCTION

Current-induced mass transport (electromigration) has been recognized and investigated for decades.^{1,2} Motivated by fundamental and practical considerations, investigations have been carried out to determine current-induced mass transport enhancement in solid-solid and solid-liquid interactions.^{3–7} The primary practical relevance of electromigration relates to the design of integrated circuits.⁸ Component miniaturization with the concomitant increase in current density leads to the electromigration and the formation of failure-inducing defects. Consequently, a significant portion of research has been prompted by circuit reliability concerns, with the vast majority of work aimed at improving conductor lifetime. Conductor lines are usually composed of pure metals (primarily Al and more recently Cu) or dilute alloys (up to ~2 at. % diluent),⁸ and thus electromigration research has focused primarily on such materials. More recent research on metal-metal interactions under the influence of a current has shown significant enhancement of the growth rate of the intermetallic phases resulting from such interactions.^{3–7} However, the effect of the magnitude of the dc and its direction on interdiffusion could not be unambiguously separated from its influence on the nucleation and growth of the resulting intermetallics. To avoid this complication, we have chosen for this investigation the copper-nickel system in which only solid solutions form.

In the previously reported investigations on the effect of

a current on metal-metal interactions,^{3–5} the results showed no dependence of the kinetics on current direction, in contrast to the anticipated results from the classical electromigration model.^{1,2} In one recent investigation, it was shown that current enhancement of mass transport (in the Ni–Ti system) was the consequence of a decrease in the activation energy of defect mobility.⁹

As indicated above, the nonthermal contributions (i.e., those not related to Joule heating) of a current to mass transport have been investigated in pure metals, dilute alloys, and intermetallics. In this paper, we describe results of an investigation on the Cu–Ni system where the complete mutual solubility of the two metals makes the investigation of interdiffusion over a wide range of concentration without the complication of new phase formation possible.

II. EXPERIMENTAL PROCEDURE

Copper-nickel diffusion layers were assembled by placing one copper foil on each side of a nickel foil thus forming a sandwich ensemble, Cu–Ni–Cu. With this arrangement it is also possible to investigate the influence of dc direction on diffusion. The foil assemblies were square in shape with 5 mm sides. The Cu foils (Alfa Aesar) had a thickness of 250 μm and a reported purity of 99.9999%. The Ni foils were 99.99% pure (ACI Alloys, San Jose, CA) and were also 250 μm thick. In addition, molybdenum foils were used as protection layers between the electrodes and the sample. A schematic of the sample assembly is shown in Fig. 1. Prior to putting the foils together, each was polished using 600 and 1200 grit SiC metallographic papers and cleaned with acetone.

The samples were then annealed in a specially built apparatus in which annealing can be affected under varying

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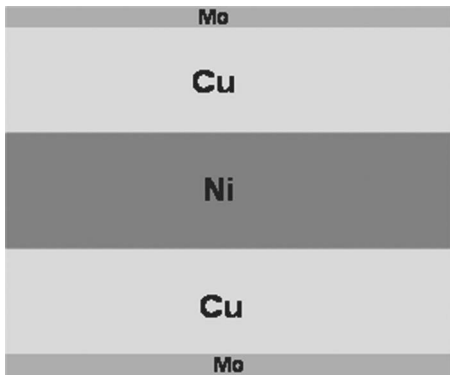


FIG. 1. Schematic of Cu-Ni diffusion couple assembly.

current density at a constant temperature. The apparatus is schematically shown in Fig. 2 and described in more detail in previous publications.³⁻⁵ Experiments were conducted under current densities ranging from 0 to 1000 A cm⁻² and at temperatures ranging from 650 to 850 °C. In all cases, the annealing time was 2 h. Temperatures in this range are considerably higher than values obtained by Joule heating due to the application of a current. At any fixed experimental temperature, a change in current density had no observable effect on the temperature of the sample. The relatively large surface area and small thickness of the foils ensured a uniform temperature in the sample under the conditions described above. At the end of each experiment, the cooled samples were mounted in conductive epoxy resin (Buehler Konduktomet), sectioned perpendicularly to the metal interfaces, and prepared metallographically. The morphology of Cu-Ni diffusion couple was determined using scanning electron microscopy (SEM). Electron probe microanalysis (EPMA) was used to provide concentration profiles at both interfaces. Line scans made across the interfaces shown in Fig. 3 were made in steps of 0.5 μm.

III. RESULTS

The micrograph shown in Fig. 3 is a backscattered electron SEM image of a sample annealed at 650 °C for 2 h without the imposition of a current. Figure 4(a) shows an

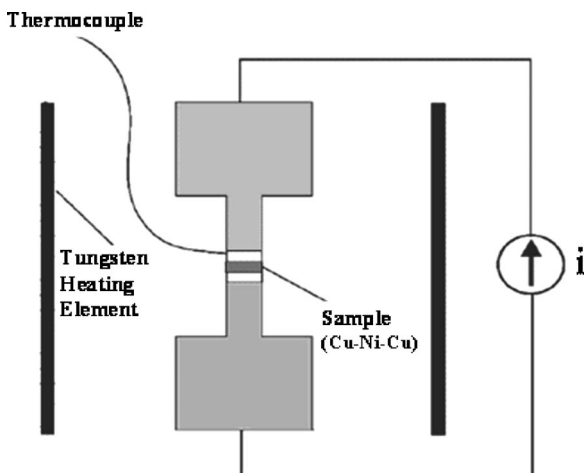


FIG. 2. Schematic of experimental apparatus.

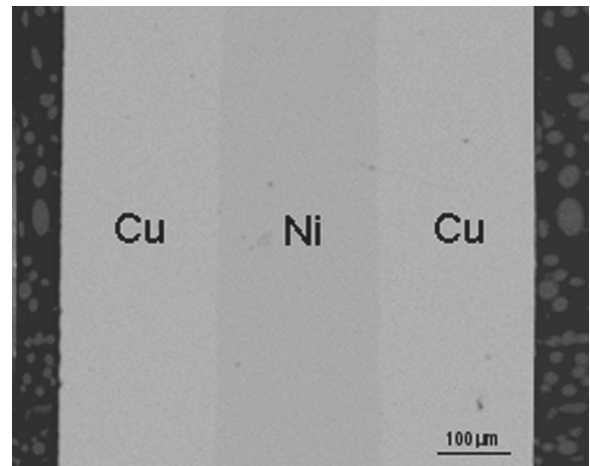


FIG. 3. Backscattered electron SEM image of Cu-Ni-Cu diffusion layers annealed at 650 °C for 2 h with no current.

EPMA line scan (atomic percent of Ni and Cu versus distance) of this sample. The points shown in this figure have a maximum 2σ error bars of ±0.5%, a value that is not readily discernable in this figure and in Fig. 4(b). There are five distinct regions observable in these scans, as indicated at the top of the figure. From left to right, these are (1) the pure Cu layer (100% Cu, 0% Ni), (2) an interdiffusion layer (100-0%

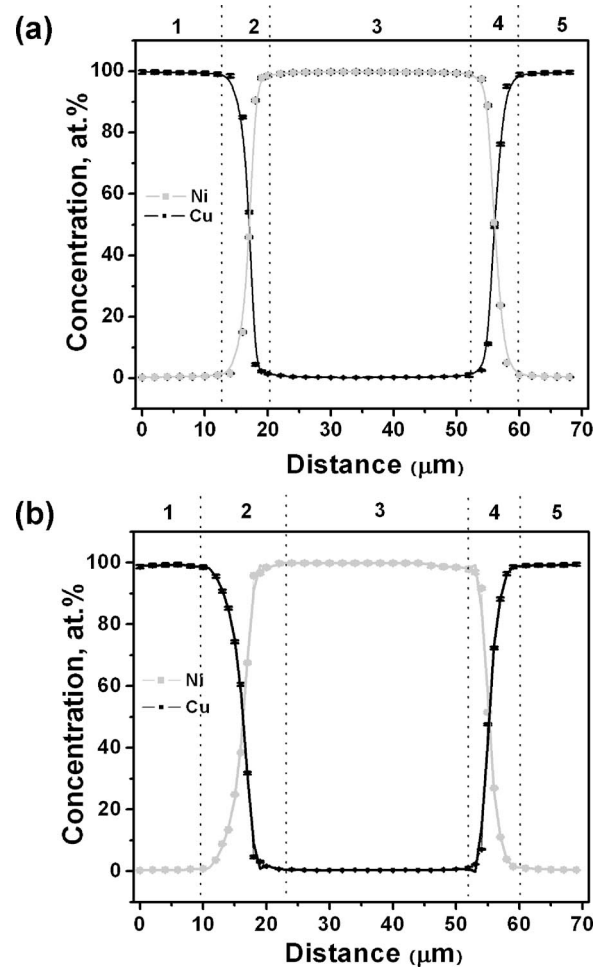


FIG. 4. Concentration profiles in Cu-Ni-Cu samples annealed at 650 °C for 2 h: (a) 0 and (b) 800 A cm⁻².

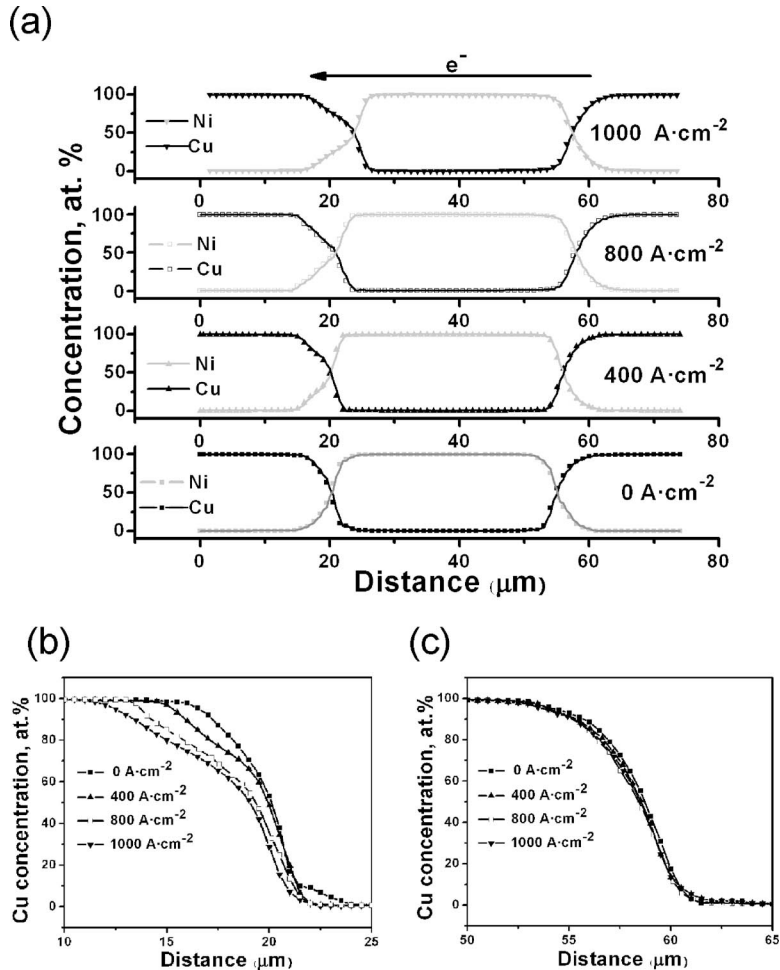


FIG. 5. (a) Concentration profiles of Cu and Ni in samples annealed at 700 °C for 2 h with varying current densities. (b) Concentration profiles of Cu on the cocurrent region. (c) Concentration profiles of Cu on the countercurrent region.

Cu, 0–100% Ni), (3) the pure Ni layer (Ni 0% Cu, 100% Ni), (4) the second interdiffusion layer (0–100% Cu, 100–0% Ni), and finally, (5) the second pure Cu layer (100% Cu, 0% Ni). Careful examination of Fig. 4(a) reveals that the curves are symmetric about a plane halfway in the Ni layer. In other words, the interdiffusion layers [regions 2 and 4 in Fig. 4(a)] are very similar in this case (where no current was applied). In addition, the shape of the curves suggests qualitatively that more Ni diffuses into the Cu layer than Cu diffuses into Ni.

Figure 4(b) shows an EPMA line scan of a sample annealed at the same temperature and for the same time (650 °C for 2 h) but in this case, under an applied current of 800 A cm⁻². The arrow near the top of the figure indicates the direction of the flow of electrons during annealing. Analogous to the sample reacted without a current [Fig. 4(a)], there are five distinct regions observable in this sample. These regions are equivalent to the regions described for the earlier sample in terms of the changes in the concentrations. However, in contrast to the previous case, the concentration curves in Fig. 4(b) are no longer symmetric since the first interdiffusion layer (region 2) is noticeably larger than the second (region 4). It should be noticed that this larger interdiffusion layer corresponds to the case when the Ni atoms were diffusing into the Cu in the same direction as the flow of electrons. We will subsequently call this the cocurrent (Co-*e*⁻) interface. Qualitatively, region 4 in the

present case is nearly identical to region 4 in the previous case [Fig. 4(a)] in which no current was applied. We will refer to region 4 as the countercurrent (Ct-*e*⁻) interface (again, with reference to Ni diffusion relative to the electron flow).

Figure 5(a) shows concentration profiles across Cu–Ni diffusion couples annealed at 700 °C for 2 h with different current densities, ranging from 0 to 1000 A cm⁻². The curves were smoothed using ORIGINLAB software adjacent average smooth function, with ten point setting. Visual observations on the profiles show that as the current density is increased, atomic diffusion is enhanced at the interface and the interdiffusion region is increased in the Co-*e*⁻ case (region 2). The width of the interdiffusion layer increases from 7.5 to 12 μm when the current density is increased from 0 to 1000 A cm⁻², as can be seen from Fig. 5(b). It is to be noted that in all cases in Co-*e*⁻ (region 2), the profile of Ni into Cu is less steep than the profile of Cu into Ni. With increasing current density, this difference in the slope of concentration becomes more pronounced. In the Ct-*e*⁻ case, differences in profiles are not readily observed with a change in current density, as can be seen in Fig. 5(c). These results indicate that the current density has a significant effect on atomic diffusion at the Co-*e*⁻. The difference between profile shapes at the Ct-*e*⁻ and Co-*e*⁻ indicates that the atomic diffusion is dependent on the direction of current.

IV. ANALYSIS AND DISCUSSION

To determine the effect of the current on the diffusion between copper and nickel, we calculated the interdiffusion coefficient \tilde{D} using the Sauer–Freise–den Broeder (SFB) method.^{10,11} Analysis by Kailasam *et al.*¹² has shown that this method provides results in agreement with the commonly used Boltzman–Matano (BM) method^{13,14} but without the need for the laborious process of determining the Matano interface.

In the SFB method, the term relative concentration Ψ is defined as

$$\Psi = \frac{C' - C_R}{C_L - C_R}, \quad (1)$$

where C' is the concentration at a given point and C_L and C_R represent the left- and the right-hand side end concentrations of the same component in the diffusion couple. The interdiffusivity \tilde{D} is calculated from the following:

$$\tilde{D}(C') = \frac{1}{2t \left(\frac{dC}{dx} \right)_{x'}} \left[(1 - \Psi) \int_{x'}^{\infty} (C' - C_R) dx + \Psi \int_{-\infty}^{x'} (C_L - C') dx \right], \quad (2)$$

where C is the concentration, x is the distance coordinate, x' is the location corresponding to C' , and t is the time of annealing. Since in our case, we used pure Cu and Ni foils for the diffusion couples, $C_L=1$ and $C_R=0$, for Cu at the ends of a diffusion couple. Thus, Ψ can be expressed as

$$\Psi = \frac{C' - C_R}{C_L - C_R} = \frac{C' - 0}{1 - 0} = C'. \quad (3)$$

Thus, Eq. (2) is simplified to

$$\tilde{D}(C') = \frac{1}{2t \left(\frac{dC}{dx} \right)_{x'}} \left\{ (1 - C') \int_{x'}^{\infty} C' dx + C' \int_{-\infty}^{x'} (1 - C') dx \right\}. \quad (4)$$

The interdiffusion coefficient can be calculated as a function of Cu concentration from the EPMA profiles.

Because of uncertainties in the determinations of slopes and areas under the concentration profile near the limiting compositions (very low or very high) of each diffusion couple, the interdiffusion coefficients calculated by the SFB and BM methods are subject to large errors near these limits.¹⁵ To avoid these uncertainties, we chose the concentration range of 10–90 at. % to calculate the interdiffusion coefficient \tilde{D} .

In the absence of a current, the calculated \tilde{D} values at both Co- e^- and Ct- e^- interfaces are nearly identical, as shown in Fig. 6. The figure shows the dependence of \tilde{D} on Cu concentration at four different temperatures for both interfaces. At all temperatures, \tilde{D} increases with concentration with the rate of increase being higher at higher temperatures.

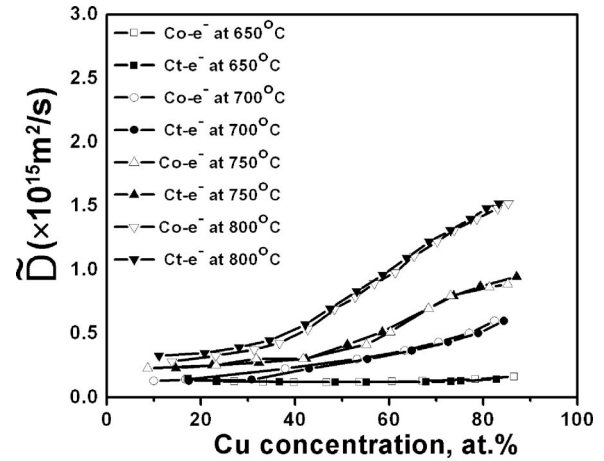


FIG. 6. Interdiffusivity across cocurrent and countercurrent regions of samples annealed at different temperatures with no current.

Investigations on the interdiffusion in the Cu–Ni system have been made previously under conditions where no current was applied.^{16–19} In Fig. 7(a), we compare our results, calculated from the nickel concentration profiles, with those of Kaja¹⁶ and Schwarz *et al.*¹⁷ at a common temperature of investigation, 650 °C. There is a very good agreement between the present results and those of Kaja¹⁶ over the concentration

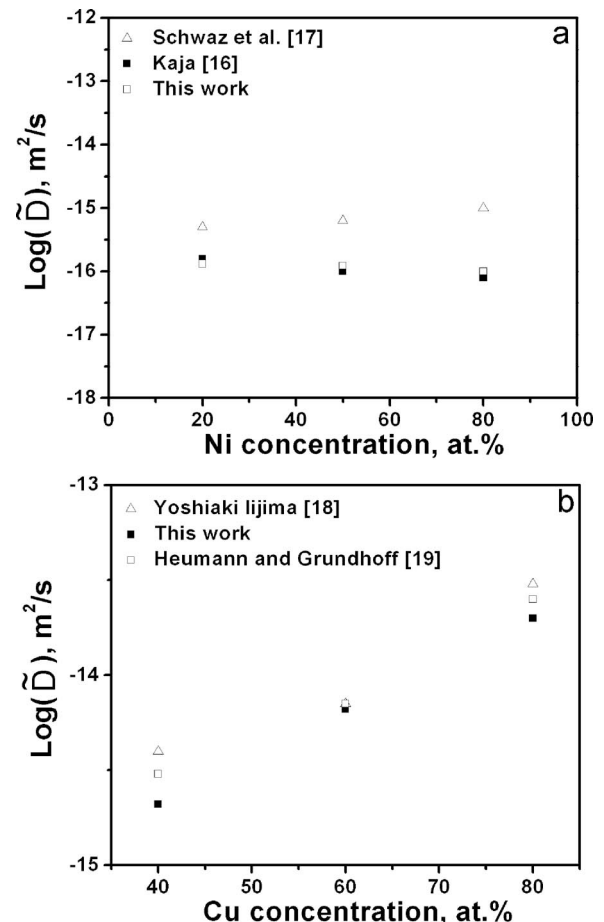


FIG. 7. (a) Comparison of measured and reported interdiffusivities in the Cu–Ni system at 650 °C. (b) Comparison of measured and reported interdiffusivities at 1000 °C in the Cu–Ni system. Results from current study extrapolated from lower temperatures.

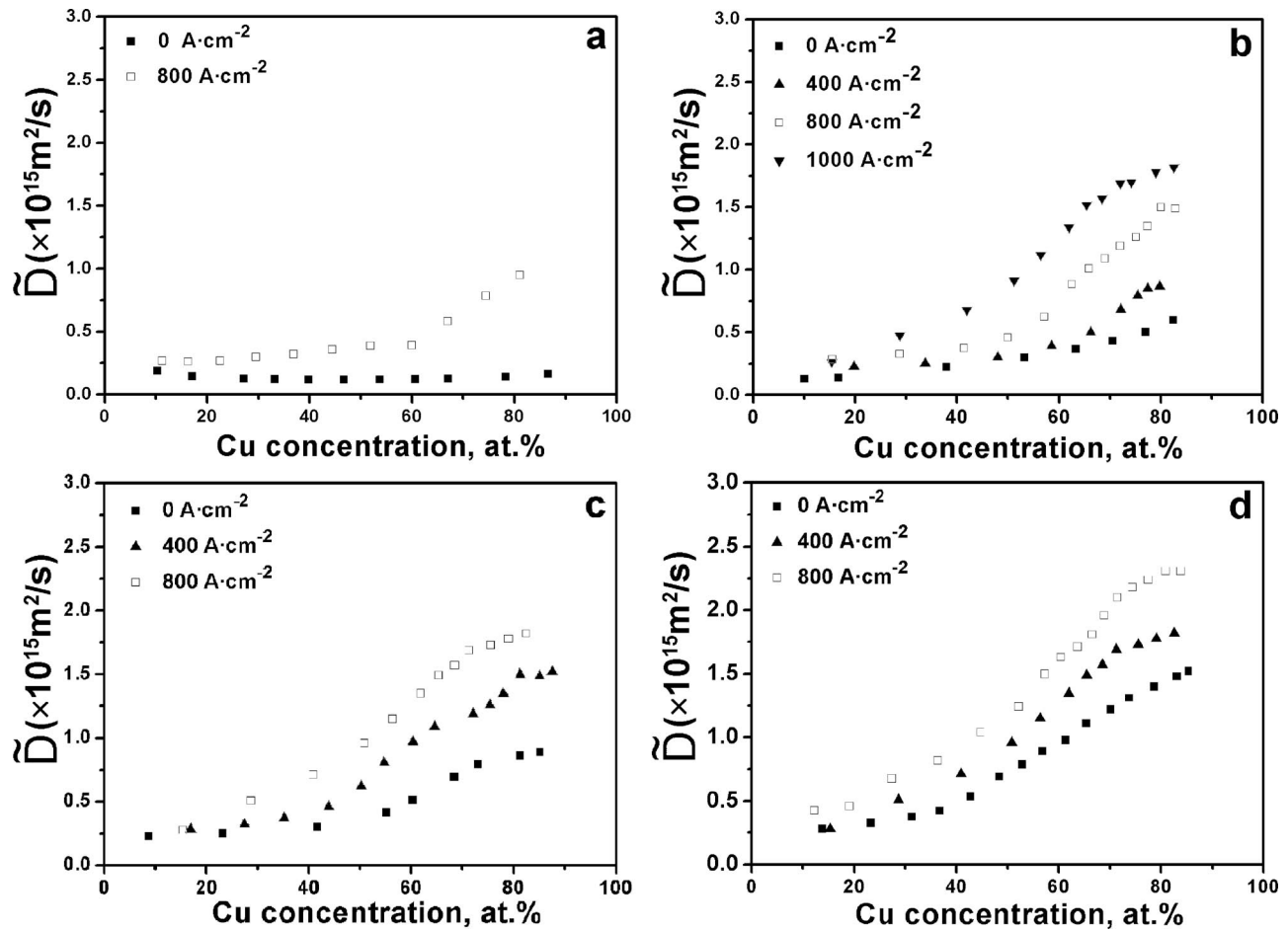


FIG. 8. Interdiffusion coefficient in Cu–Ni system in samples annealed at different temperatures with varying current density: (a) 650, (b) 700, (c) 750, and (d) 800 °C.

range of 20–80 at % Ni. In contrast, the more recent results of Schwarz *et al.*¹⁷ are as much as about an order of magnitude higher than those of Kaja and the present study. In comparing their results to those of Kaja, Schwarz *et al.* attributed the higher values they calculated to grain size effect. The latter authors used materials with a grain size of 10 μm , while Kaja samples had a grain size of 25 μm . The lower activation energy for grain boundary diffusion is given as a reason for the grain size dependence. In this study, the grain size for both Cu and Ni was in the range of 26–30 μm and thus is closer to the value reported by Kaja.¹⁶ Because of the increasing dominance of volume diffusion at higher temperatures, grain size effect becomes less important as the temperature of annealing is increased. Results of interdiffusion studies at a much higher temperature, 1000 °C, were reported by Iijima *et al.* over a wide range of copper concentrations¹⁸ and compared to earlier results reported by Heumann and Grundhoff at the same temperature.¹⁹ In Fig. 7(b), we compare the published results from these studies with extrapolated values (to 1000 °C) from our results for Cu concentrations of 40, 60, and 80 at. %. The agreement between the published values and the extrapolated present results is very good.

The influence of a current on \tilde{D} over a wide range of Cu concentrations for samples annealed at 650, 700, 750, and 800 °C is shown in Figs. 8(a)–8(d), respectively. These re-

sults, which were calculated from profiles represented by the Co- e^- interface (region 2 in Figs. 4 and 5), show a marked increase in interdiffusivity when a current is applied. The magnitude of the increase depends on composition and temperature, as seen in the figures. A comparison of the effect of a current of density of 800 A cm^{-2} on \tilde{D} for the Co- e^- and Ct- e^- interfaces at various temperatures is shown in Fig. 9(a). In all cases, \tilde{D} values obtained at the Co- e^- interface are significantly higher than corresponding values for the Ct- e^- interface. Figure 9(b) shows, in fact, that the same current (800 A cm^{-2}) has no observed effect on interdiffusion at the Ct- e^- interface at the various temperatures reported, within the limit of uncertainties in these measurements. These observations show that the effect of the dc is directional. When the electrons flow from Cu to Ni (at the Ct- e^- interface), there is no effect on interdiffusion, but when the electrons flow from Ni to Cu (at the Co- e^- interface), the current causes a marked increase in the interdiffusion.

According to the electromigration theory, a high density of traveling electrons should cause a force (electron wind force) that causes atoms to diffuse preferentially from the anode to the cathode or vice versa depending on the sign of the effective charge z^* of the diffusing atoms. Values of z^* are unavailable for Ni diffusing in Cu or Cu diffusing in Ni. The z^* values for Ni and Cu self-diffusion are both negative.¹

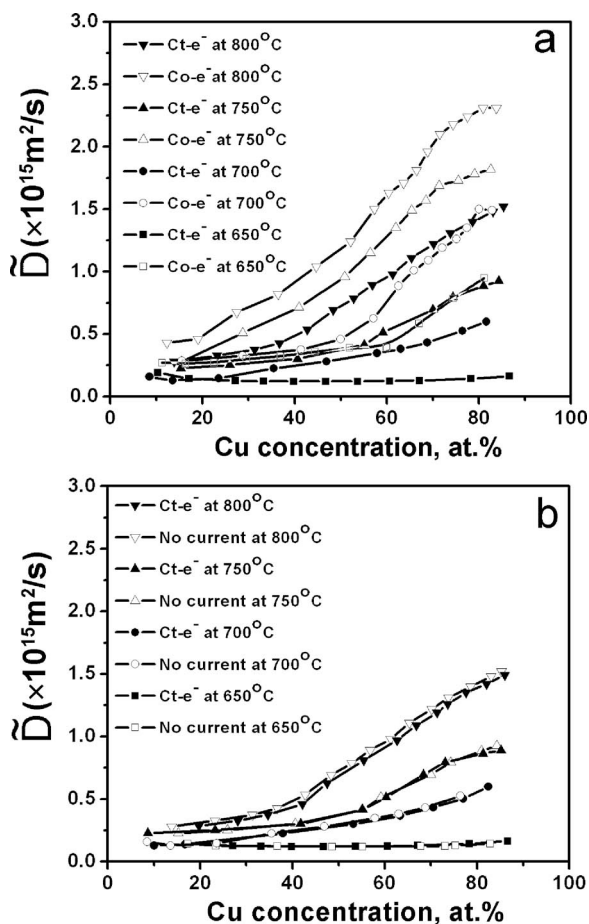


FIG. 9. (a) The effect of current on interdiffusivity at both interfaces of samples annealed at different temperatures. (b) The effect of a current on the interdiffusivity for the countercurrent interface at different temperatures.

Given the similarities between Cu and Ni, it is likely that the substitutional (vacancy-atom exchange) diffusion mechanism is similar in the solid solution as in self-diffusion. It can be assumed that an electron wind in the Cu–Ni solid solution would also cause the atoms to move preferentially in the direction of the electron travel.

As seen above, the current increases the interdiffusivity \tilde{D} over a wide range of concentrations at all temperatures in the cocurrent interface but has no influence on \tilde{D} in the countercurrent direction. The relationship between \tilde{D} and the intrinsic diffusivities of copper D_{Cu} and nickel D_{Ni} is

$$\tilde{D} = D_{\text{Ni}}X_{\text{Cu}} + D_{\text{Cu}}X_{\text{Ni}}, \quad (5)$$

where X_{Cu} and X_{Ni} are the atomic fractions of copper and nickel, respectively. The observations discussed above would suggest that D_{Cu} and D_{Ni} have been enhanced in region 2, while remaining relatively unchanged in region 4. Invoking only the electron wind effect, the increase in diffusivity of one or both elements in one direction (by the current) should be accompanied by an equal decrease in the opposite direction. By this argument the interdiffusivities should be increased on one interface and decreased on the other. As mentioned previously, this was not the case in this study. Such an apparent discrepancy might be reasoned by taking into ac-

count effects of vacancy motion and current-induced vacancy motion.

Previous work on interdiffusivities and intrinsic diffusivity^{18–20} shows that the intrinsic diffusivity of Cu D_{Cu} is higher than D_{Ni} at all concentrations. Such a situation leads to preferential diffusion of one element and to the well-known effects of Kirkendall shifts and Kirkendall porosity. Vacancies play an important role in electromigration since the flux of atoms due to the electron wind requires an equal and opposite flux of vacancies. In a binary system with complete solid solubility, such as the Cu–Ni system, if the binding energy between a vacancy and an atom is the same for both components, then the flux of each will be proportional to its composition. However, it is more likely that in real system, the binding energies are not the same and thus the flux of atoms will not be proportional to composition, potentially leading to retardation of electromigration. There are no data on the interactions of a vacancy with Cu or Ni in a Cu–Ni solid solution. The slow diffusion of Cu in Ni (relative to that of Ni in Cu)^{21,22} would lead us to speculate that the interaction energy between a copper atom and a vacancy is negative (repulsive). In the presence of an electric field, the increased concentration of vacancies due to the migration of Ni would lead to a further decrease in the diffusion of copper. This implies that the electron wind effect on Cu is counteracted by the effect of vacancies and would be consistent with the present observation in region 4.

Finally, the effect of the current on interdiffusion is now examined in the terms of its influence on the activation energy. Figures 10(a)–10(c) are Arrhenius plots of interdiffusion coefficient for Cu concentrations of 40, 60, and 80 at.%, respectively. In each case, plots for 0 and 800 A cm^{−2} current densities are shown. The calculated activation energy values and their uncertainties (standard deviations) are shown in Table I. For lower Cu concentrations (≤ 40 at %), the imposition of a current has no apparent effect on the activation energy. However, at higher concentrations, the presence of a current decreases the activation energy significantly; at 80 at. % Cu, the imposition of a current density of 800 A cm^{−2} results in a 61% decrease in the activation energy. In the absence of a current, higher interdiffusivities at high copper concentration in the Cu–Ni system have been reported and attributed to void formation resulting from high vacancy concentration.¹⁸ In previous investigations, it was shown that the imposition increases the concentration of vacancies in a Cu–Al alloy²³ and decreases the activation energy for defect mobility in a Ti–Ni intermetallic.⁹ In view of these observations, the lack of a significant effect by the current on the activation energy at low copper concentrations in this study would suggest that the current effect on mobility is likely to be the cause of the decrease in the activation energy in high concentration samples. At lower copper concentrations, the low vacancy concentration appears to dominate the effect such that there is no apparent change in the activation energy.

V. CONCLUSIONS

The effect of the imposition of a dc on interdiffusion in the Cu–Ni system was investigated over the temperature

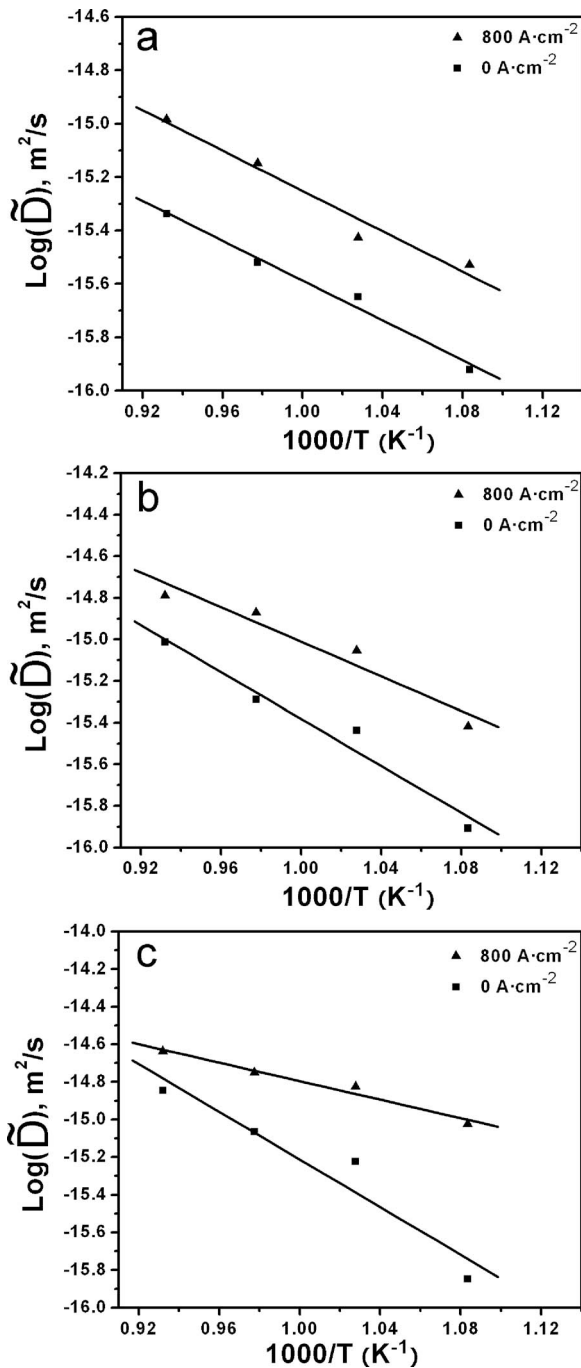


FIG. 10. Arrhenius plots of interdiffusion coefficient at different Cu concentrations: (a) 40, (b) 60, and (c) 80 at. % Cu.

range of 650–850 °C. Using trilayered Cu–Ni–Cu diffusion samples, the influence of the direction of the current at densities ranging from 0 to 1000 A cm⁻² was studied at the two interfaces provided by this sample geometry: Cu–Ni and Ni–Cu. Samples were annealed at fixed temperatures and current densities for 2 h in all cases and then analyzed by EPMA to provide concentration profiles at both interfaces. From these concentration profiles, interdiffusivities \tilde{D} were calculated using the SFB method. The results for cases in the absence of a current were in good agreement with previously published results. The influence of the current on \tilde{D} depended on its direction relative to the two interfaces. At the interface where

TABLE I. Effect of current density on activation energy for interdiffusion coefficient in the Cu–Ni system.

Cu concentration (at. %)	Current density (A cm ⁻²)	Activation energy (kJ mol ⁻¹)
40	0	31.02±4.28
	800	31.40±2.68
60	0	46.82±6.32
	800	35.56±6.32
80	0	52.68±10.81
	800	20.45±2.41

the electronic flow was from Ni to Cu (cocurrent interface), the interdiffusivity showed a marked increase when a current was applied. It was, however, unchanged when the electronic flow was from Cu to Ni (countercurrent interface). The increase of \tilde{D} in the cocurrent interface depended on concentration and temperature. The effective activation energy of interdiffusivity depended on concentration and decreased with the application of a current. The decrease was largest for higher copper concentrations. The results are interpreted in terms of a proposed vacancy-atom interaction for copper with the implication that the electron wind effect on Cu is counteracted by the effect of vacancies.

ACKNOWLEDGMENTS

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