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Current effects on neck growth in the sintering of copper spheres to copper plates by the pulsed electric current method

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The effect of a pulsed dc on the sintering of copper spheres to copper plates was investigated. It was shown that the current had a marked effect on neck growth between the spheres and the plates. The enhancement of sintering under the effect of the current was attributed to electromigration. Microstructural observations on fracture surfaces of necks formed under high currents showed considerable void formation. It was also observed that the current resulted in increased evaporation and the formation of bunched evaporation steps. Formation of these steps and their location relative to the neck were consistent with current density distributions. The results of this investigation provide direct evidence for the role of the current in the sintering in the pulse electric current sintering method. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2743885]

I. INTRODUCTION

The use of a current to activate sintering processes is more than seven decades old, but the recent availability of commercially built devices and the demonstrated decided advantages of their utilization have generated renewed widespread interest in field activated sintering.[1] The use of the pulsed electric current sintering (PECS) [or spark plasma sintering (SPS)] method has provided considerable evidence of advantages over conventional methods such as pressureless sintering and hot pressing. The advantages include lower sintering temperature, shorter holding time, and significant improvements in properties relative to those consolidated by conventional methods.[1] Among the many advantages of lower temperature and shorter sintering time, the feasibility of consolidating nanostructured powders to full density with minimal grain growth has provided a notable example of success.[2] The ability to prepare dense materials and maintain the nanostructure has provided the opportunity to determine heretofore unrecognized nanoscale effects in materials.[3,4]

Of the more than 1000 papers on sintering by PECS published in the last decade, few have attempted to provide fundamental understanding of the process[5] and thus the reasons for its superiority over conventional means remain largely not well understood. A common explanation provided in many publications is the role of plasma, proposed to exist between powder particles during sintering. It is suggested that the plasma removes surface impurity layers and possibly activates sintering. However, the occurrence of plasma has not been unambiguously demonstrated. Pulsing of the dc is advocated to be part of the generation of the plasma. But in a recent series of investigations on the fundamentals of the PECS process, it was shown that pulse patterns had no effect on mass transport.[6–8] While the debate on the existence of plasma remains unsettled, two considerations that distinguish the PECS method (aside from pressure which is also used in hot pressing) are commonly accepted as contributing to its advantage. These are the high heating rate and the enhanced mass transport by electromigration.[9] Heating rates of 1000 °C min^{-1} and higher are achievable, thus making the bypassing of nondensifying sintering mechanisms occurring at low temperatures possible.[10] Recently, Olevsky and Froyen provided a modeling approach to the PECS process and concluded that electromigration contributes significantly to the observed enhanced diffusion.[11]

However, while the existence of electromigration (or electrotransport) is recognized, its role in enhancing mass transport during PECS has not been adequately considered. In recent investigations the current has been shown to increase reactivity in multilayer systems in diffusion couple[12–14] and in PECS experiments.[8] Recently, Wang *et al.*[15] conducted a study on diffusion under PECS conditions and compared it to results obtained under hot pressing. In that work the effect of the current on sintering of spheres was evaluated from shrinkage measurements, with information on neck growth between spheres obtained indirectly from porosity measurements. While this work represents an important step in providing a better understanding of the PECS process, it did not provide a direct evaluation of the effect of the current, since the work compares behaviors obtained independently by two different methods, PECS and hot pressing.

The difficulty in conducting a study to directly evaluate the effect of the current is the requisite of separating the Joule effects from the intrinsic effects of the current. Under PECS conditions temperature and current are dependent parameters. As will be described in more detail below, in this work we have devised an experimental method in which we can vary these parameters independently. In doing so, we have investigated the effect of the current on the neck growth between copper spheres and copper plates in a PECS apparatus.

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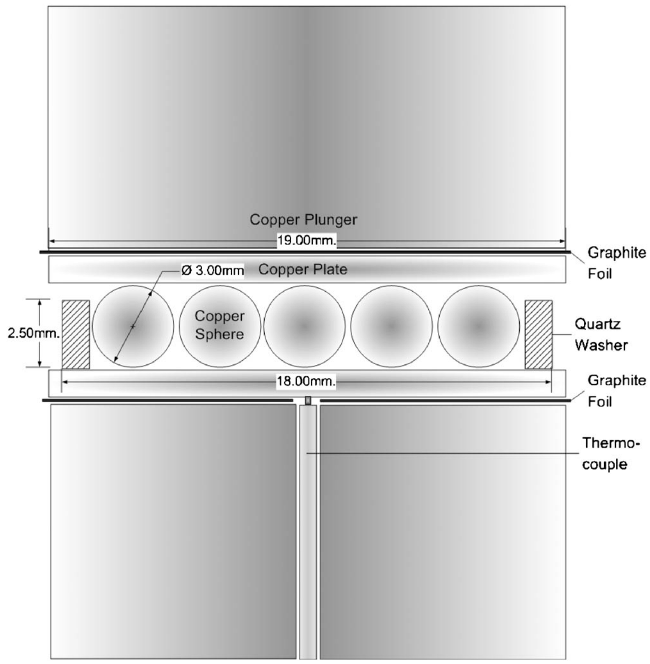

FIG. 1. Schematic of sample of sphere to plate sintering geometry.

II. EXPERIMENTAL MATERIALS AND METHODS

Neck growth between copper spheres and copper plates was investigated by the use of the PECS method. The copper spheres, obtained from Goodfellow (Devon, PA), were 99.9% pure with a diameter of 3 ± 0.01 mm. The copper plates, in the form of disks, were also 99.9% pure, obtained from ESPI Metals (Ashland, OR). The plates were 1 mm thick and had a diameter of 18.25 mm. The plates and spheres were polished using 1 μm diamond paste and cleansed with 100:1 (vol) nitric acid solution. Polishing and cleaning assure that the surfaces are free from irregularities and surface contaminants. The spheres were placed between the plates and contained within a quartz circular ring that is 2.5 mm thick with an outer diameter of 18 mm and an inner diameter of 16 mm. The height of the ring was less than the diameter of the spheres so that the upper plate only touches the spheres. The placement of the spheres inside the quartz ring helped ensure a reproducible location between the plates while situated in the PECS apparatus. Quartz was chosen because it is an electrical insulator that has a high melting point with low isotropic thermal expansion. With this assembly, 21 touching spheres can be placed between the plates in any given experiment.

A copper plunger is placed both above and below the plate-sphere setup. In between the copper plate and the copper plunger is a thin layer of graphite foil. Since graphite is quite insoluble in copper, this thin foil separates the plate from the plunger to prevent possible adhesion between the two during the PECS process. A small hole is drilled in the center of the bottom copper plunger to allow for placement of a K-type thermocouple which was insulated with alumina. The thermocouple wires are 250 μm in diameter and are in contact with the bottom copper plate. The setup is shown schematically in Fig. 1.

The lowest force exerted by the uniaxial press in the

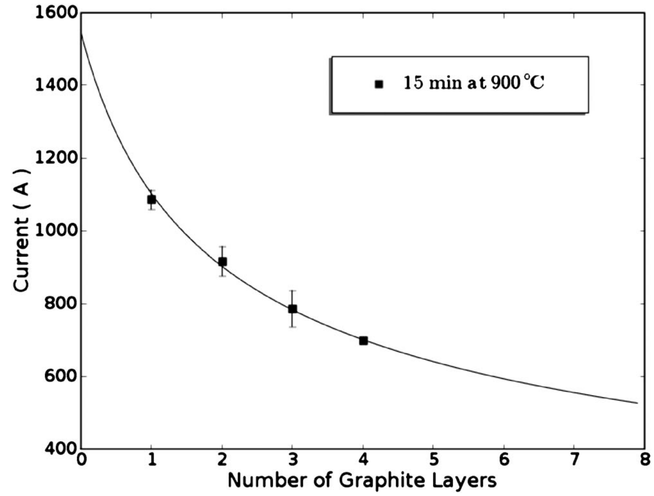

FIG. 2. Dependence of total current on the number of graphite foil layers at 900 °C.

PECS apparatus is sufficiently high that plastic deformation of the copper spheres and plates at the high sintering temperatures is a concern. Thin disks of graphite felt were added above the top and below the bottom copper plungers of Fig. 1. Graphite felt is used because under relatively small loads it returns to its original shape after being compressed, even when subjected to high temperatures.

As indicated above, the temperature and current are dependent parameters in the PECS method. To be able to study the influence of the current independent of temperature, modifications were made on the sample setup. This was done by making the copper plungers multilayered on both sides, consisting of a layer of copper followed by a thin less conductive graphite foil layer. The purpose of adding layers is to increase the total resistance which decreases the current density for the same amount of power generated. This is due to the added resistances from the graphite foil and contact interfaces. Since the only source of heat in the SPS is from Joule heating it can be shown that the power generated to maintain a steady-state temperature is constant. Thus the generated power can be written as

$$P = I^2[2\mathrm{R}_{\mathrm{cp}} + 2(x-1)R_{\mathrm{cu}} + 4xR_{\mathrm{co}} + 2xR_{\mathrm{gf}} + R_{\mathrm{sa}}], \quad (1)$$

where R represents the resistance of the item indicated in the subscript (cp=copper plunger, cu=copper disk, co=contact, gf=graphite foil, and sa=sample), I is the current, and x the number of graphite layers in the upper plunger. R_{co} is the contact resistance between the copper plate and the graphite foil. Since R_{co} and R_{gf} are much greater than the other resistances, Eq. (1) can be simplified to

$$I = \frac{1}{x^{1/2}}\left(\frac{P}{4R_{\mathrm{co}} + 2R_{\mathrm{gf}}}\right)^{1/2}. \quad (2)$$

Since for any power P (and thus temperature), R_{co} and R_{gf} are constants, the current is proportional to the inverse of the square root of the number of layers in the upper plunger. It should be noted that Eq. (2) is only valid when R_{co} and R_{gf} are much greater than R_{cp}, R_{cu}, and R_{sa}. Thus, the equation cannot be used to accurately predict the current for setups with one or less graphite foil layer, $x \leqslant 1$. Figure 2 shows experimentally determined current values obtained at a measured constant temperature of 900 °C as a function of number of graphite foil layers. The figure also shows the best-fit curve from the relation of Eq. (2).

To determine neck formation in the absence of a current, the plate-sphere ensemble was placed inside a graphite die that was bored out on both ends to 2.54 cm (1 in.) in diameter and a depth of 1.27 cm (0.5 in.). This limits the plunger depth and creates a cruciblelike environment for the copper plate-sphere setup. Instead of copper plungers as previously mentioned, 2.54 cm graphite plungers were now used. Above the copper plate graphite felt was placed to act as a spring that approximated the force applied to the copper spheres in the current case. The plate-sphere ensemble was surrounded by a thin layer of alumina felt that is thin enough to allow heat conduction to the copper while electrically insulating it from the graphite. To further electrically insulate the copper, all of the graphite walls that form the space where the copper sample sits were coated with a boron nitride spray.

To assess the effect of a current on sintering under PECS conditions, the initial focus is on the reliability of the results obtained under current-free conditions. One of the difficulties of accurately measuring the neck growth rate is to ensure that there is sufficient pressure on the copper plates and spheres to provide a good electrical contact while at the same time minimizing the effects of pressure on sphere deformation. Two sets of tests were conducted using the zero current setup to verify that the maximum applied force of 3 N used in the experiment did not significantly affect the neck growth measurements. The first set consisted of samples sintered at 900 °C using the setup described above. The graphite felt placed between the alumina felt and the upper graphite plunger transmitted a force of not more than 3 N. In the second set the graphite felt was removed from between the alumina felt and graphite plunger, thus effectively eliminated all external forces on the copper plate-sphere ensemble. The measured necks under both conditions were in agreement within a maximum of 4%, indicating the absence of an effect by the 3 N force applied during sintering.

The PECS apparatus used is a SPS device, model 1050 made by Sumitomo Company (Tokyo, Japan). The heating rate up to the experimental temperature of 900 °C was approximately 90 °C min^{-1}. The dc pattern used was 12 pulses on and 2 off (12:2); the duration of each pulse is 3 ms. Since it took about 10 min to reach the experimental temperature, neck measurements were made on samples heated to 900 °C and cooled with no hold time. This neck diameter is considered the zero time value presented in the results.

After each sintering experiment the copper plates and spheres were encased in epoxy resin and cured for a minimum of 24 h. This ensures that the spheres, necks, and plates remain in a fixed location. Using a sharp razor to break the lower plate away from the epoxied spheres revealed two measurable neck locations. One is at the epoxy-neck interface where the spheres are encased in epoxy and the other is on the previously sinter-bonded regions from the removed plates. The lower plate was used simply because it is in direct contact with the thermocouple and hence the measured temperature represents that of measured necks.

Of course, the neck measurement accuracy is based on the assumption that the location where the copper plate separates from the spheres is at the neck. This assumption is reasonable since the neck constitutes the cross section of the smallest diameter which makes it the most probable fracture location. Additionally, literature supports this method as a valid and accurate way to measure the neck size diameters during the initial stages of sintering, i.e., $x/R \leqslant 0.3$, where x is the neck radius and R is the sphere diameter.[16] Neck sizes were measured from scanning electron microscope (SEM) images obtained with Philips XL30s equipped with a field emission gun and operated at 5 kV and 200 nA.

III. RESULTS

Figure 3 shows SEM images of typical neck geometries on the copper plate (with the same magnification) resulting from sintering at 900 °C for 60 min under various currents. Specifically, parts (a), (b), (c), and (d) show necks formed with currents of 0, 750, 850, and 1040 A, respectively. The effect of the current on neck growth is clearly seen from these images. As will be shown later, fracture surfaces of necks formed at higher currents and longer times include evidence of void formation. Figure 4 shows the increase of the neck size with sintering time at 900 °C under different currents, including the zero current case. The figure shows the ratio x/R plotted versus time, with the error bars representing standard deviations in the measurements of the neck size. As indicated above, the ratio at $t=0$ represents the neck formed during the ramp up to temperature with no hold time. The neck size after 60 min of sintering in the presence of the highest current, 1040 A, is nearly five times larger than that resulting from sintering in the absence of a current under the same conditions.

During the initial sintering stage for a simplified model for spheres under isothermal conditions the time dependence of the neck ratio, x/R, is expressed as[17]

$$\left(\frac{x}{R}\right)^n = \frac{Bt}{R^m}, \tag{3}$$

where B (containing the diffusion coefficient) is a constant and n and m are mechanism dependent constants.[10] A plot of x/R vs t of the result obtained in this study is shown in Fig. 5. The slope, which represents the reciprocal of the constant n, decreases markedly as the magnitude of the applied current is increased. Table I gives the calculated values for n for the different currents and for current per copper sphere.

IV. DISCUSSION

The calculated value of 4.13 for the exponent n in Table I for current-free sintering suggests a volume diffusion mechanism, with possible contributions from a nondensifying mechanism, possibly evaporation condensation. Moon *et al.* concluded that volume diffusion was the operative mechanism in a study on the sintering of copper spheres of about 1 mm diameter.[18] In an earlier study on the sintering of 0.127 mm copper spheres at 1020 °C, it was shown that the mechanism constant n is different than the anticipated value of 5 for volume diffusion.[19] A comparison with simulation

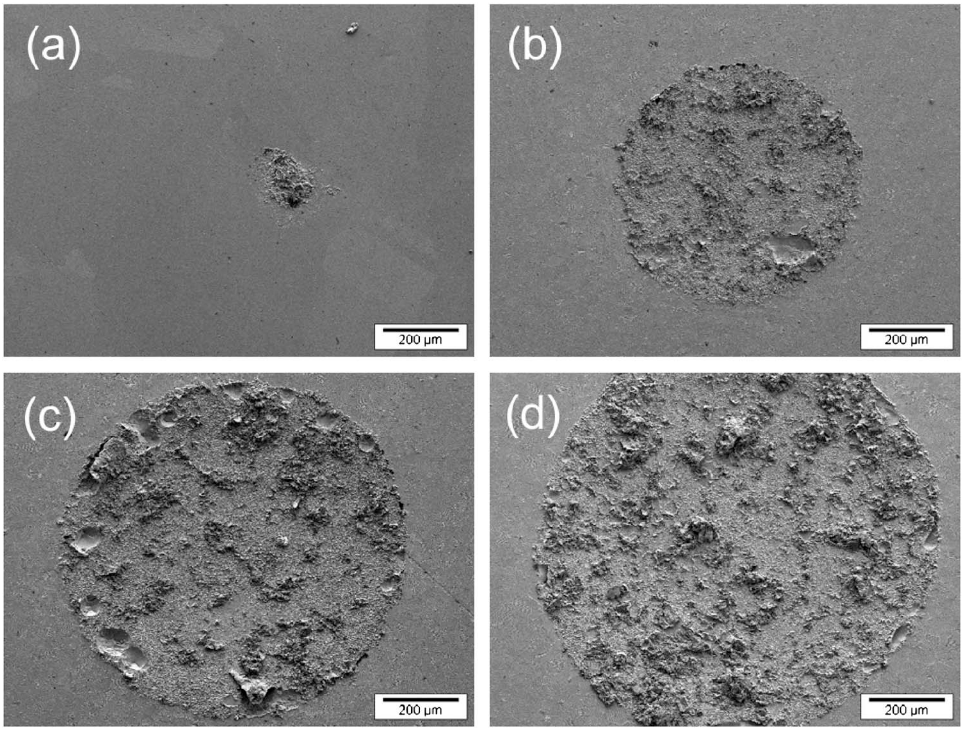

FIG. 3. SEM images showing the effect of current on the neck formation between copper spheres and copper plates sintered at 900 °C for 60 min: (a) zero current, (b) 700 A, (c) 850 A, and (d) 1040 A.

prediction gave a best fit for a combined volume and surface diffusion mechanisms. In the present study, evaporation-condensation is a possible contributor; evidence for evaporation was noticed especially when a current was applied, as will be discussed below.

As seen in Table I, the application of a current results in effective exponent constants that have no basis in the physical sense in terms of the classic sintering mechanisms. Enhanced mass transport by electromigration contributes significantly to the marked increase in neck growth when sintering is carried out under the influence of a current. Because of geometric considerations, the density of the current is highest in the neck area, especially near the perimeter of the neck, as can be seen from the simulation results depicted in Fig. 6. The simulation was intended to approximate the conditions for one sphere between two plates with an imposed current of 30 A. Figure 6(a) shows a pictorial two-dimensional (2D) distribution of the current density for the chosen geometry. The current density profile along a line running from the center of the 300 μm radius neck to the plate is shown in Fig. 6(b) and a corresponding temperature profile is shown in Fig. 6(c). The simulation results show that the temperature inside the neck is constant but decreases very slightly beyond the perimeter of the neck. In contrast, the current density distribution is uneven, showing a peak at the edge of the neck and decreasing to a zero value beyond the perimeter. The existence of high current density outside the perimeter of the neck appears to have an effect on evaporation and the development of corresponding surface morphology, as will be discussed later.

The current density is a function of neck size and hence

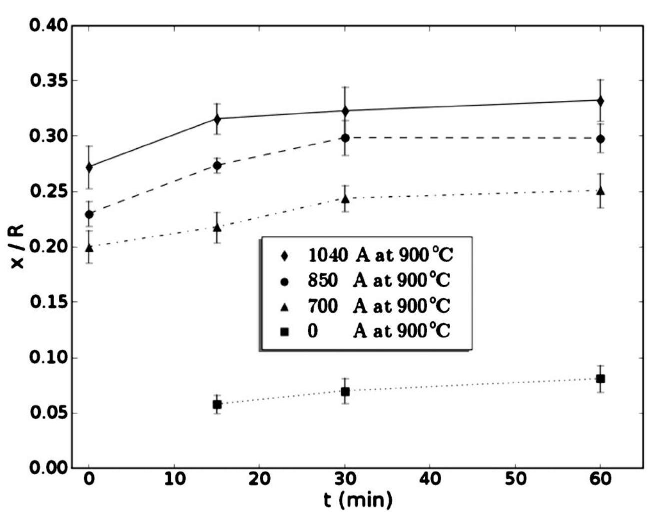

FIG. 4. Time dependence of neck growth between copper spheres and copper plates at 900 °C under different currents. The neck size at zero time refers to the value obtained during ramp up to temperature.

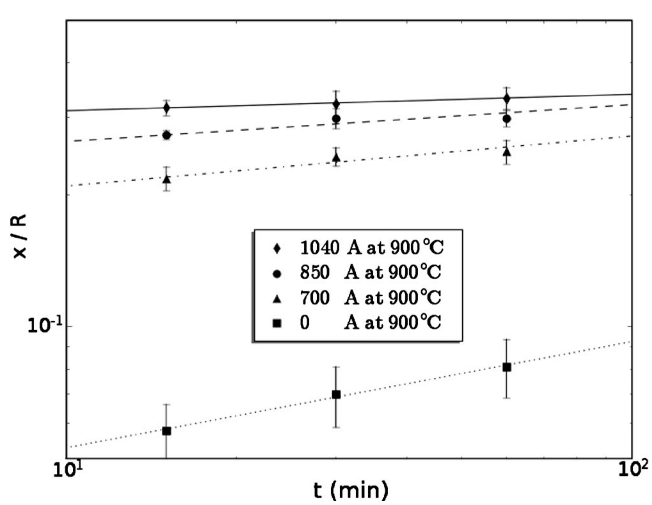

FIG. 5. Log-log plot of neck ratio x/R vs time for sintering of copper spheres to copper plates at 900 °C.

TABLE I. Calculated values for the mechanism dependent constant n for sintering of copper spheres to plates at 900 °C.

Total steady-state current (A)	Steady-state current per sphere (A)	Sintering exponent (n)
0	0	4.13
700	33.33	8.76
850	40.47	11.89
1040	49.52	20.64

changes with sintering time. Using the total measured neck areas for all 21 spheres at various times and the total current, we calculate the change in current density with sintering time for the three total current levels. The results are shown in Fig. 7. The results for the total currents of 850 and 1040 A are significantly lower than those for the 700 A case. For example, at $t=30$ min, the current density for the 700 A case is nearly 2 kA higher than the value for the other total currents. This discrepancy is attributed to void formation in the neck regions for the high current cases, as stated earlier. Formation of voids can be clearly seen from Fig. 8 which represents SEM images of necks formed at different times with a total current of 1040 A. At 15 min, the microstructure appears free of voids, but voids become prominent near the periphery with increased sintering time.

The presence of voids, resulting from the coalescence of vacancies, as a consequence of electromigration has been observed in numerous studies and has been heavily investigated on metallic interconnects in integrated circuit (IC) devices.[20,21] It was shown that void formation depended on current density and that the presence of voids did not influence the local temperature until they constituted more than 95% of the total cross section.[22] An increase in vacancy concentration due to the application of a current has been reported by Asoka-Kumar *et al.* on aluminum-copper alloys.[23] The coalescence of these vacancies and their association with impurities leads to the formation of the observed voids.[21] Closer examination of Fig. 8 shows that void formation occurs preferentially near the edge (outer surface) of the neck and that sintering time is a factor in the formation of the voids. The location of the voids corresponds to the highest current density, which, as Fig. 6(b) shows, is located at the perimeter of the neck.

An interesting microstructural observation in this study is the presence of halolike features surrounding the necks, as can be seen in the optical micrograph of Fig. 9. A closer inspection by SEM reveals that the halos are, in fact, ledge patterns and etched grain boundaries, as Fig. 10 shows. The ledge patterns suggest evaporation in this region.[24,25] Evidence of evaporation of copper was provided by the observation of thin copper-colored deposits at nearby cooler regions in the apparatus. In the present study, however, the occurrence of the ledges was not uniform but depended on the distance from the edge of the neck. Ledges became less abundant as the distance from the neck increased. Moreover, their occurrence was seen to relate to the magnitude of the current, as can be seen in Fig. 11. In the absence of a current, these features are virtually absent (within the resolution of the images) and when a current is imposed, their apparent density decreases with radial distance from the neck.

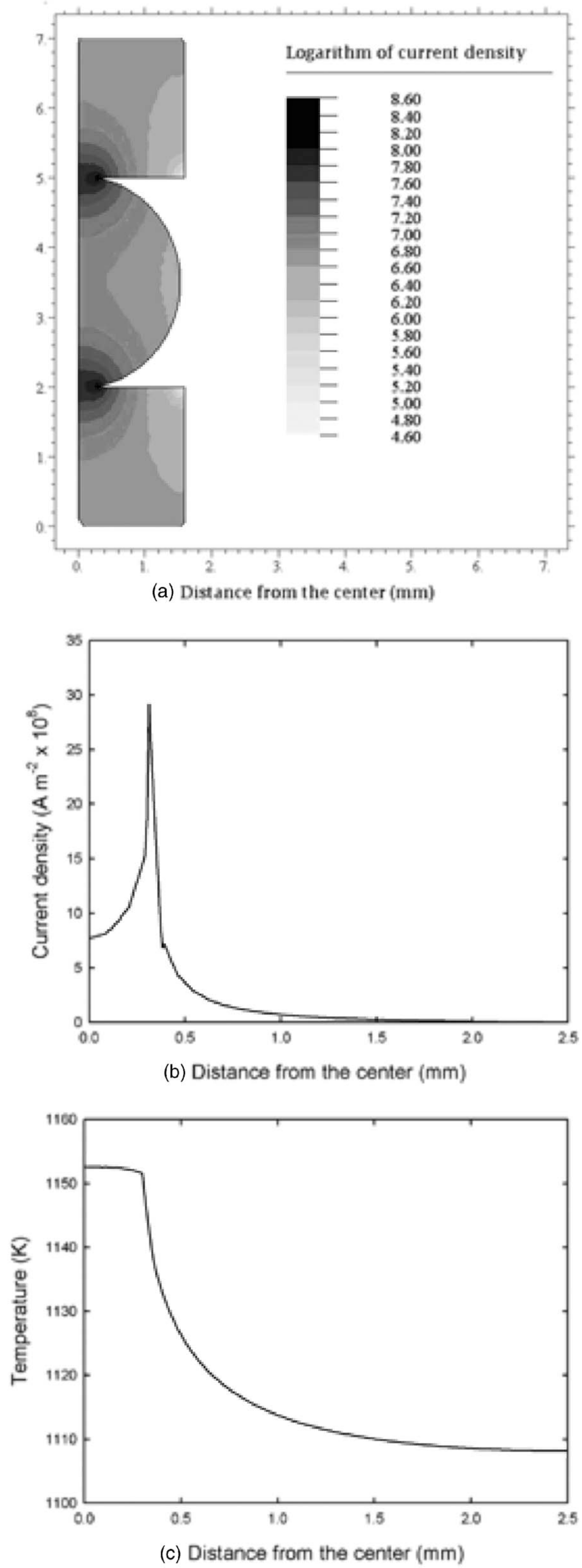

FIG. 6. Simulation of current distribution in a copper sphere between two copper plates: (a) Current density distribution (scale is in base-ten logarithm of current density), (b) current density profile, and (c) temperature profile.

At 900 °C the vapor pressure of copper is about 10^{-4} Pa ($\sim 10^{-6}$ torr),[26] a relatively low value that should not contribute significantly to mass transport. Thus the occurrence of

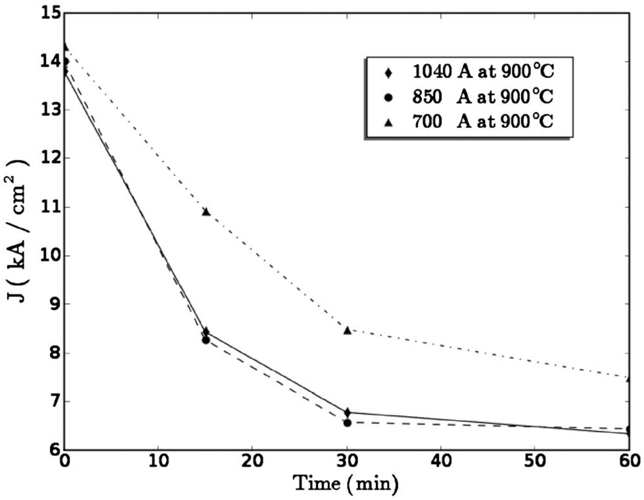

FIG. 7. Variation of calculated current density with time during sintering of copper spheres to copper plates at different applied total currents.

the evaporation ledges in the presence of a current and their decrease with distance from the neck strongly suggest a dependence on current density. There are no literature accounts in which a direct effect of a current on the evaporation rates of metals is demonstrated (indirect observations have been reported recently[27]). However, evidence for evaporation in electromigration experiments has been provided for aluminum and copper.[28,29] Also, recent investigations have shown that the application of a dc during evaporation of Si results in marked changes in the morphology as a consequence of bunching of steps on vicinal surfaces.[30–33] The origin of step bunching is related to surface electromigration[28] with adatoms influenced by the external force resulting from the current. The force F acting on adatoms is expressed as

$$F = Z^* eE, \quad (4)$$

where Z^* is the effective charge of the adatom, e is the electronic charge, and E is the electric field on the surface. The parameter Z^* represents contributions from an electrostatic force and electron wind force. The surface diffusion field under the influence of a current can induce an attractive force between surface steps and that this force counteracts the normal step-step repulsive interactions.[34]

In addition to influencing surface diffusion, we may assume that the electromigration force also influences detachment of adatoms from ledges[35] or their desorption from terraces, and thus anticipate an effect of the current on evaporation. This would explain the occurrence of the ledges and the presence of considerable copper deposit when sintering was carried out under a current. As stated earlier, copper deposits were virtually absent when a current was not utilized and ledges were not observed. With this proposed explanation we can anticipate a spatial dependence of the effect of the current, dictated by density distribution, as shown by our simulation results, Fig. 6(b). Higher current densities in the vicinity of necks give rise to higher step bunching and higher evaporation rate. The bunching of the steps facilitates their observation by the SEM.

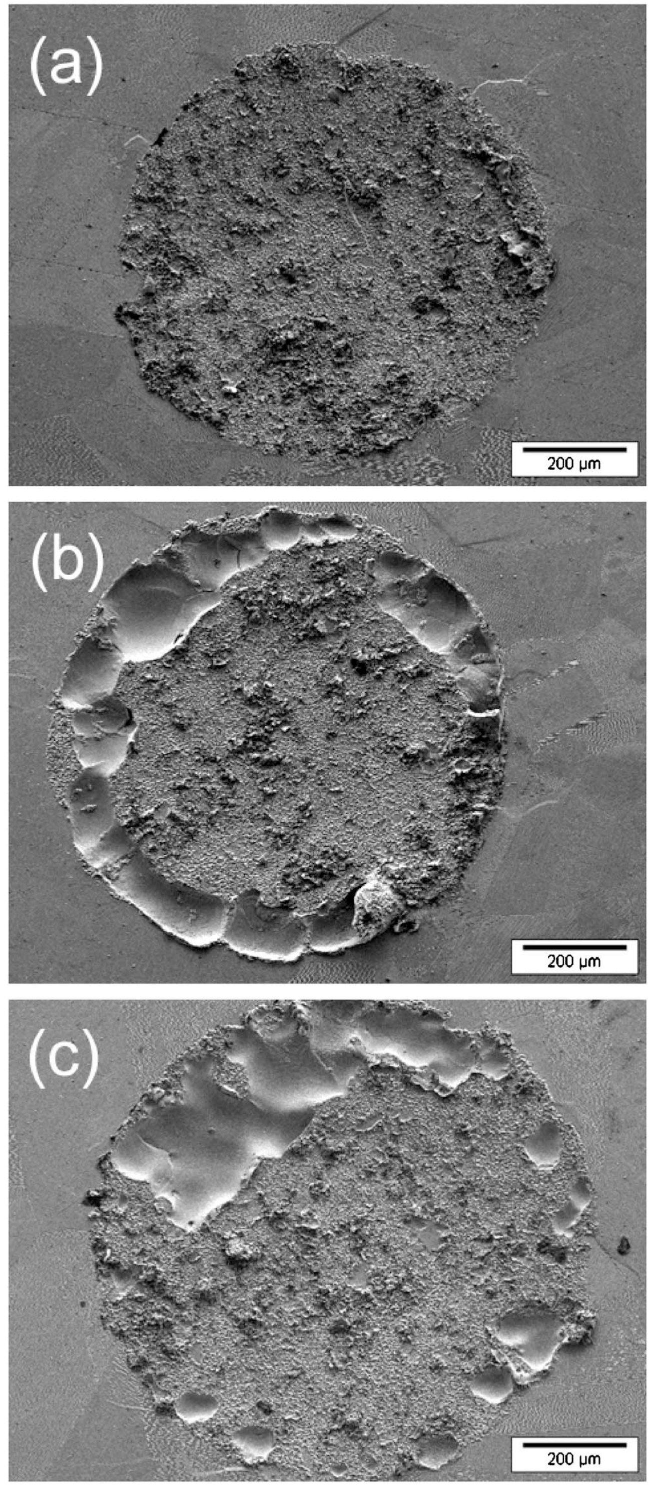

FIG. 8. SEM micrographs showing void formation during sintering at 900 °C with a total current of 1040 A for (a) 15 min, (b) 30 min, and (c) 60 min.

V. CONCLUSIONS

The sintering of copper spheres to copper plates was investigated under the influence of current in a pulse electric current sintering (PECS) conditions. A special experimental design was utilized such that the current can be varied while maintaining a constant sintering temperature. The current was shown to have a marked effect on neck growth between

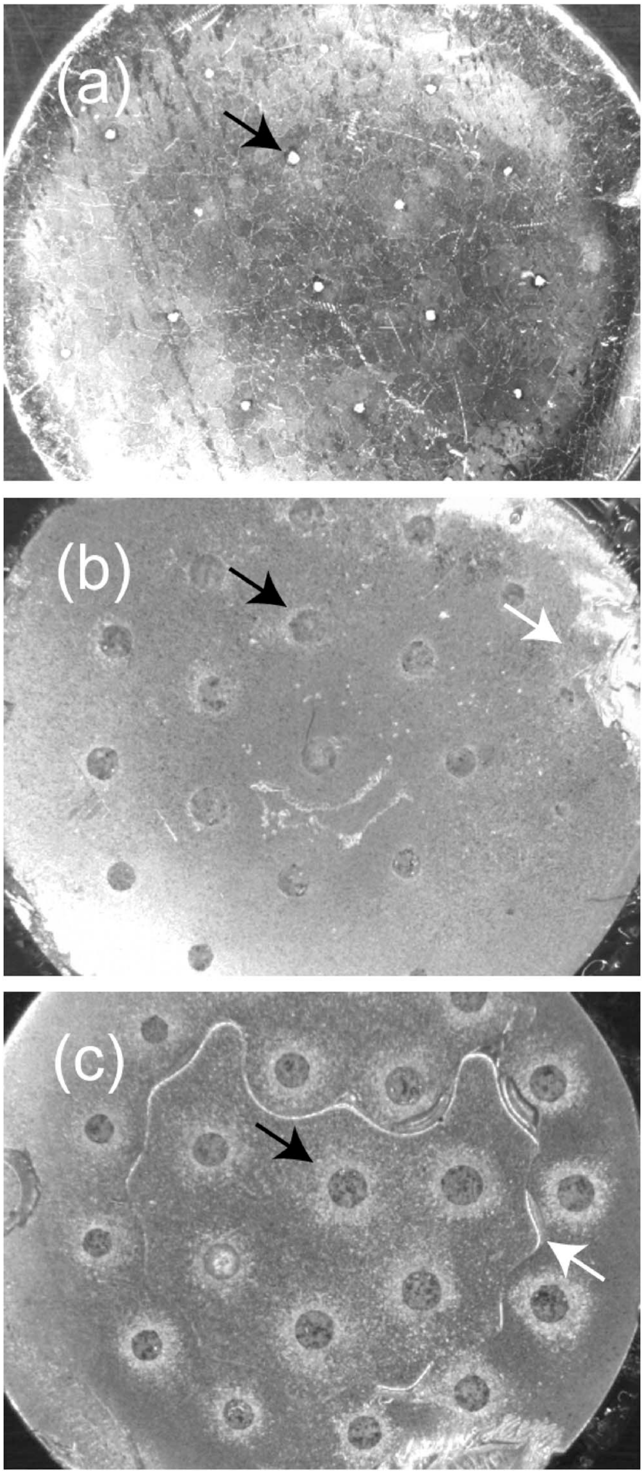

FIG. 9. Optical micrographs of neck images on copper plate showing the "halo" formation around perimeters of necks: (a) zero current, (b) 700 A, and (c) 1040 A.

the spheres and the plates. In the absence of a current, sintering was consistent with a volume diffusion mechanism with a contribution from evaporation-condensation. The enhanced neck growth under the effect of the current was attributed to electromigration.

Microstructural observations on fracture surfaces of necks formed under high currents showed considerable void formation, resulting from coalescence of vacancy whose for-

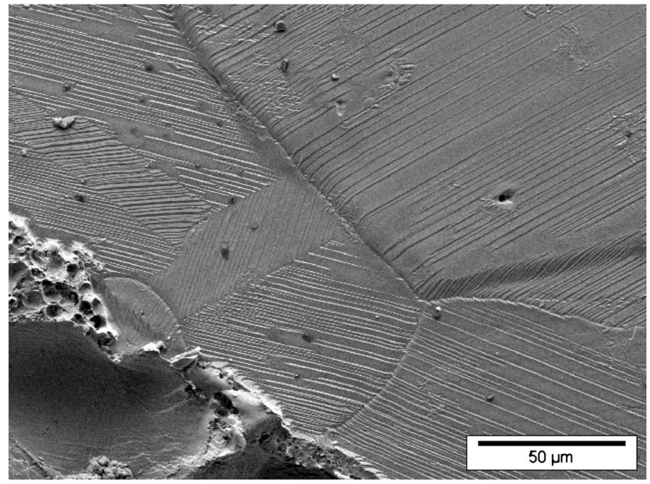

FIG. 10. SEM image near the edge of a neck showing the formation of ledges.

mation is enhanced by the current. These observations are consistent with prior accounts on interconnects in IC devices. Additionally, it was observed that the current resulted in increased evaporation with the consequence of step bunching of evaporation steps, as has been observed on Si surfaces and

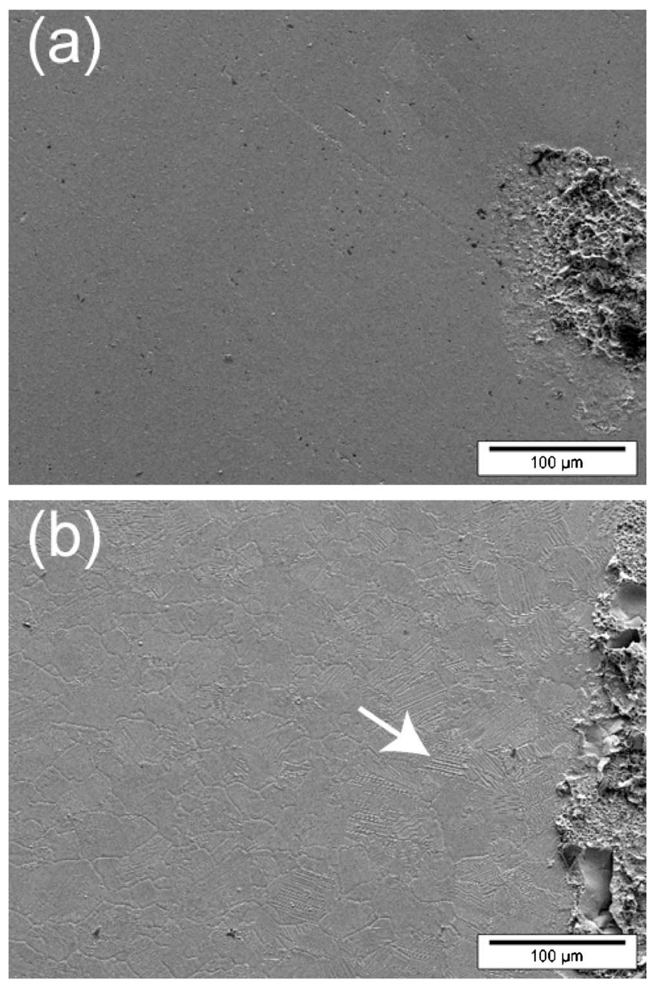

FIG. 11. SEM images illustrating the dependence of the abundance of ledges on current and distance from the neck during sintering for 15 min: (a) zero current and (b) 1040 A.

attributed to electromigration. Formation of these steps and their location relative to the neck were consistent with current density distributions.

The results of this investigation provide direct evidence for the role of the current in the sintering in the PECS method.

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