Preparation, electrical contact performance and arc-erosion behavior of Cu/La$_2$NiO$_4$ composites

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Abstract. La$_2$NiO$_4$ conductive particles were incorporated into Cu matrix for enhanced electrical contact performance, while their effect on the arc erosion behavior of contacts was investigated. The results indicate that the particles in the hot-pressed Cu/La$_2$NiO$_4$ composites accelerate the separation of oxide scales from contact surface during arc erosion cycling, suggested by increased mass loss of roughly 3 to 9 times larger than pure Cu. Thus, the growth of oxide scales on contact surface was blocked by a self-cleaning-like function. As a result, the contact resistance declines to 21.6 mΩ after 5000 cycles, in contrast with 29.5 mΩ for Cu, and correspondingly the temperature rise degrades about 30.7%.

Introduction

In long-term service, oxide scales involving CuO and Cu$_2$O are generated on Cu contact surface due to the arc-erosion-derived effects during turning on and off contact pairs in air; and therefore, the growth of the low-conductive scale layers results in the development of high contact resistance, potentially leading to performance deterioration or failure of devices [1]. To strengthen Cu for electrical contact applications, alloying is an effective approach, e.g., Cr, W for Cu contacts in vacuum interrupters [2,3]. Anidow et al. [4] alloyed Cu with 9 at.% La to reduce the contact resistance through doping cuprous oxide. But introducing alloying elements remarkably weakens the bulk conductivity of Cu base [5]. The reinforcements like Al$_2$O$_3$, ZnO, and Y$_2$O$_3$ powders are also incorporated into Cu to enhance arc erosion resistance; nevertheless, the maximum bulk conductivity only approximating to 80% IACS are obtained by adding 4 wt.% ZnO particles [6].

In this work, we prepared novel Cu contact materials for low-voltage switches by incorporating 5 wt.% La$_2$NiO$_4$ ceramic particles with electrical conductivity close to metals [7], not only to preserve high bulk conductivity of Cu base, but also to improve the electrical contact performance via a self-cleaning-like surface that can induce accelerated separation of scale from contact surface during circuit opening and closing.

Experimental

Preparation of Samples. La$_2$NiO$_4$ powders were synthesized through a sol-gel strategy that both Ni(NO$_3$)$_2$ and La(NO$_3$)$_3$ were used as precursors, as shown in detail in Ref. [8]. The synthesized sol was heated at 130 °C to transform to dry gel, and sintered at 850 °C to form crystalline La$_2$NiO$_4$ particles. Next, La$_2$NiO$_4$ powders with a content of 5 wt.% were homogeneously mixed with commercially elemental Cu powders (99.5%, <45 μm). The blended powders were cold-pressed at 600 MPa, and then sintered at 980 °C in vacuum furnace, accompanied with a densification process that was hot-pressed at 750 °C and 50 MPa for 20 min, so as to obtain densely bulk Cu/La$_2$NiO$_4$ composites. For comparison, pure Cu ones were fabricated in the same way.

Characterization of Samples. The phases of materials were determined by D8-ADVANCE X-ray diffractometer (XRD). A JSM-7000F field-emission scanning electron microscope (FESEM) was applied to observe the microstructures. The electrical conductivity was measured by Sigma2008A
eddy current conductivity meter. The arc erosion test was performed on a device like that shown in Ref. [6,9], using $\Phi$ 6 mm $\times$ 12 mm sized-samples for both upper (fixed) and lower (movable) contacts. The test was operated by continually switching on and off 1000, 2000, 3000, 4000 and 5000 cycles at parameters of AC current 10 A, contact gap 4 mm and contact force 3 N. For per 1000 cycles, the mass of both upper and lower contacts were measured to obtain the total mass loss; and the contact resistance was measured via voltage-ampere method at DC current of 1 A, while the temperature rise was determined in the conditions of AC current 10 A, contact force 3 N and contact time 30 min.

Results and Discussion

Phase and Morphology of La$_2$NiO$_4$ Particles. Fig. 1 shows the XRD pattern of the synthesized La$_2$NiO$_4$ powders, suggesting crystalline La$_2$NiO$_4$ as main component for its high electrical conductivity, and a little residual La(NO$_3$)$_3$ precursor. The irregularly shaped particles demonstrate sizes less than 10 $\mu$m (the inset).

![Fig. 1](image1.png)

Fig. 1 XRD pattern and SEM image (the inset) of sol-gel-derived La$_2$NiO$_4$ particles.

Phase and Microstructure of Cu/La$_2$NiO$_4$ Composites. Fig. 2 presents the XRD pattern of the hot-pressed Cu/La$_2$NiO$_4$ composites. The very strong peaks are attributed to Cu matrix, and the small content of La$_2$NiO$_4$ results in its weak peaks, while slight oxygen contamination may present during sample preparation for the possible presence of Cu$_2$O. The results indicate that the addition of 5 wt.% La$_2$NiO$_4$ particles just weakly affects the composition of Cu base. The inset is the SEM image of the composites, demonstrating dispersed La$_2$NiO$_4$ particles in continuous Cu matrix.

![Fig. 2](image2.png)

Fig. 2 XRD pattern and SEM image (the inset) of Cu/La$_2$NiO$_4$ composites.

Electrical Contact Performance of Cu/La$_2$NiO$_4$ Composites. Fig. 3(a) shows the electrical conductivity of Cu/La$_2$NiO$_4$ composites and pure Cu, revealing the conductivity of 85% and 98% IACS, respectively. The value for adding La$_2$NiO$_4$ into Cu is higher than that for other incorporations, reported in Ref. [1,6,10,11], whose values are in an approximate range of 11.8—81.9% IACS, though different fabrication conditions such as the variety (e.g., TiB$_2$, Y$_2$O$_3$, ZrC, ZnO, Al$_2$O$_3$) and content (e.g., 0.5—6.0 wt.% or 1—2 vol.%) of reinforcements. The continuously distributed Cu matrix acts as a high-performance pathway for electrical conduction, and primarily determines the overall conductivity of the composites; meanwhile, La$_2$NiO$_4$ conductive ceramic reduces the electrical resistance originated from the second phase in conventional cases.
Fig. 3 Electrical conductivity (a) of Cu/La$_2$NiO$_4$ and Cu contacts as well as contact resistance (b), temperature rise (c) and mass loss (d) of both during arc erosion test.

Figs. 3(b)—(d) present the electrical contact performance of Cu/La$_2$NiO$_4$ composites during arc erosion test, and compared with that of pure Cu. As shown in Fig. 3(b), both materials display similar contact resistance at the first 2000 cycles; but, subsequently, Cu/La$_2$NiO$_4$ composites display rather smaller resistances than Cu contacts. The resistance decreases about 19.5%—35.8% from 3000 to 5000 cycles. The temperature rise of Cu/La$_2$NiO$_4$ is also smaller than that of pure Cu (Fig. 3(c)), which declines roughly 30.7%—69.5%; and moreover, the remarkable decline appears around 2000 cycles, approximating to the threshold of contact resistance decrease, shown in Fig. 3(b). More importantly, the mass loss for Cu/La$_2$NiO$_4$ is significantly larger than that for pure Cu, which differs from the common results demonstrated in Cu contacts strengthened with particle reinforcements to prevent mass loss. As reported by Güler and E. Evin [6], an introduction of ZnO, Al$_2$O$_3$ and Y$_2$O$_3$ particles into Cu causes significant decreases of mass loss compared with pure one by arc erosion testing 3000, 6000 and 9000 counts. In detail, the mass loss of Cu/La$_2$NiO$_4$ composites is about 0.55, 0.63, 1.54, 1.92, 2.29 mg larger than that of pure Cu after 1000, 2000, 3000, 4000 and 5000 counts, implying accelerated removal of oxide scales from contact surface in service.

**Arc-Erosion Behavior of Cu/La$_2$NiO$_4$ Composites.** Figs. 4(a) and (b) show the SEM image of the surface morphology of Cu/La$_2$NiO$_4$ composites after 5000 cycles, exhibiting solidified droplets, cracks and pores derived from speedy melting and solidification due to arc erosion. Fig. 4(c) presents the XRD pattern of the sample, indicating Cu$_2$O as the product after thousands of cycles of test. Fig. 4(d) shows the corresponding cross-sectional SEM image, apparently displaying two different areas in oxide layer, pointed by arrow 1 and 2, while their enlarged views are shown in Figs. 4(e) and (f). For area 1, a thinner scale is well bonded with substrate; however, a thicker scale is isolated from substrate by a crack passing through the scale/substrate interface in area 2. Besides, it is noticeable that the significant difference between both areas is whether La$_2$NiO$_4$ particles present at scale/substrate interface, schematically illustrated by the insets in Fig. 4(e) and (f).

It is a well-established fact that the generation of oxide is consequent on arc erosion during switching on and off Cu based contacts in air, while stress applies on scales due to the contact force, conducing to the fracture of the brittle scale and mainly causing mass loss [6]. Therefore, the growth of scale layers is a dynamic process of oxide generation and separation; nevertheless, the generation rate is usually larger than the separation rate, so that the layer continuously thickens to an unacceptable contact resistance. In conventional cases, the reinforcements significantly impede the isolation of
scales, indicated by decreased mass loss for ZnO, Al₂O₃ and Y₂O₃ reinforced Cu contacts [6]. It therefore forms a thick scale layer to prevent or weaken further arc erosion of the inner part of contact materials; but the layer, producing high contact resistance and large temperature rise, deteriorates the performance stability of switches as well. For Cu/La₂NiO₄ composites, on the other hand, La₂NiO₄ particle at scale/substrate interface plays a role in initiating crack to enhance the partial removal of scale layers, assumed by a comparison between Figs. 4(e) and (f), thus achieving self-cleaning-like function during cycling without any accessional aid, and also the small contact resistance and temperature rise. Finally, practical uses for the function can be also realized owing to the similar conditions between the arc erosion test and the service as well as the low cost of Cu compared with Ag base.

Fig. 4  (a), (b) SEM images of the surface morphology of Cu/La₂NiO₄ composites after 5000 cycles; (c) The corresponding XRD pattern; (d) The corresponding cross-sectional SEM image; (e), (f) Enlarged views of area 1 and 2 pointed by arrows in Fig. 4(d), whose insets demonstrate schematic illustrations for the origin of self-cleaning-like functions.

Conclusions

The hot-pressed Cu/La₂NiO₄ composites present a high bulk conductivity, 85% IACS, while their contact resistance decreases 19.5%—35.8% during 5000 cycles in contrast with pure Cu, and correspondingly the temperature rise degrades about 30.7%—69.5%. The particles in the composites accelerate partial separation of oxide scales from contact surface during arc erosion cycling, like the self-cleaning functions, indicated by increased mass loss of roughly 3 to 9 times larger than Cu, so that the contact resistance and temperature rise reduce for an enhanced performance in service.

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References