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Electrical conductivity of a bulk metallic glass composite

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The authors report the electrical conductivity of a bulk metallic glass (BMG) based composite fabricated by warm extrusion of a mixture of gas-atomized glassy powders and ductile α-brass powders. The conductivity of the BMG composite can be well modeled by the percolation theory and the critical percolation threshold volume of the high-conductive brass phase was estimated to be about 10%. It was found that the short irregular brass fibers can dramatically reduce the resistivity of the BMG, leading to an improved material with both high strength and good conductivity for functional applications. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2795800]

Bulk metallic glasses (BMGs) with ultrahigh strength and low elastic modulus have the excellent capabilities to support large Hookean elastic strain and high stress for the applications in pressure sensors and microelectromechanical systems (MEMSs).[1–3] However, the electrical conductivity of metallic glasses is generally two orders of magnitude lower than that of their crystalline counterparts, which limits their applications where both excellent mechanical properties and good electrical conductivity are required. The very low conductivity of metallic glasses originates from their intrinsic disordered structures.[4–6] Composition modification and the reduction of defects, the two effective ways to improve the conductivity of crystalline materials, have been proved to be unsuccessful in metallic glasses.[6–8] Therefore, developing BMG composites that contain high-conductive crystalline phases may be a viable approach to improve the conductivity of BMGs. Recently, a variety of BMG composites with crystalline components have been developed with the focus on enhancing the plasticity of BMGs.[9,10] The effect of crystalline phases on the electrical properties of BMGs has not been reported so far. In this letter, we present the results showing that the electrical conductivity of a Ni-based BMG can be dramatically improved by adding α-brass fibers and the conductivity of the composite can be quantitatively modeled by a percolation theory.

A $Ni_{59}Zr_{20}Ti_{16}Si_2Sn_3$ (at %) BMG composite containing 40 vol % α brass was used in the present study. The material was fabricated by warm extrusion of a mixture of gas-atomized fully $Ni_{59}Zr_{20}Ti_{16}Si_2Sn_3$ amorphous powders and ductile α-brass ($Cu_{80}Zn_{20}$, at %) powders and details can be found in Ref. 11. Mechanical tests have demonstrated that the composite possesses high strength and improved ductility.[11] For comparison, the monolithic $Ni_{59}Zr_{20}Ti_{16}Si_2Sn_3$ BMG sample prepared by copper-mold casting was used as the reference. The resistivity measurements were performed by using the standard four-probe technique at a temperature ranging from 2 to 300 K. The applied current directions were either parallel or perpendicular to the extrusion direction in order to evaluate the longitudinal and transverse resistivities, respectively.

Figures 1(a) and 1(b) show typical optical micrographs of the BMG based composite on the longitudinal and transverse sections, respectively. The brass phase appears to uni-

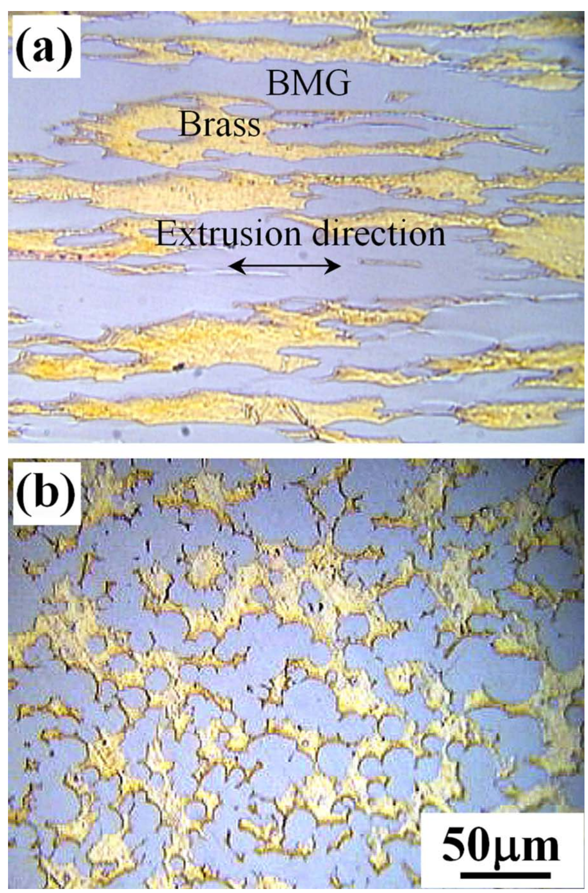

FIG. 1. (Color online) Optical micrographs of the BMG composite on (a) the longitudinal section and (b) transverse section.

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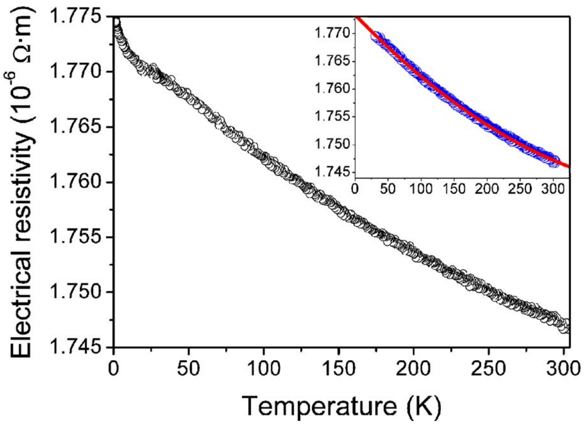

FIG. 2. (Color online) Temperature dependence of electrical resistivity of the monolithic Ni-based BMG in a temperature range from 2 to 300 K.

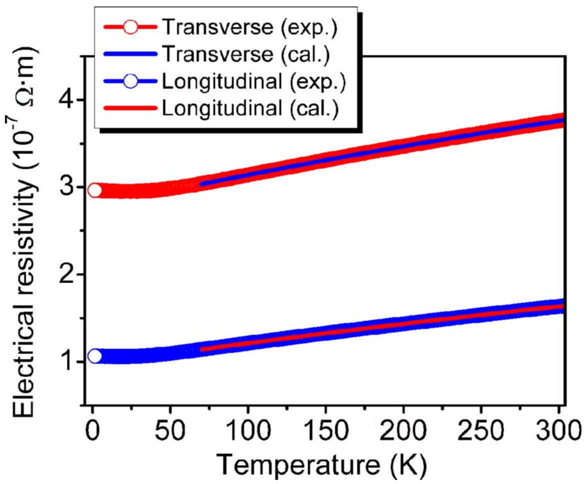

FIG. 3. (Color online) Electrical resistivity of the BMG composite in the longitudinal and transverse directions at temperatures ranging from 2 to 300 K. The solid lines represent the calculated results based on the GEM equation.

formly distribute in the BMG matrix in both sections, and visible voids at the BMG/brass interfaces have not been found. Because the flow stress of brass was relatively lower than that of the glassy matrix, the ductile brass phase with an irregular shape was elongated along the extrusion direction and the overall morphology of the brass particles can be described as short fibers embedding in the glassy matrix. Separate transmission electron microscope characterization demonstrates that the BMG particles keep fully amorphous after the extrusion and the interfaces between the BMG matrix and the brass phase are well bonded with a 100-nm-thick interdiffusion zone.

For comparison, monolithic $Ni_{59}Zr_{20}Ti_{16}Si_2Sn_3$ BMG sample was used as the reference and its electrical resistivity is shown in Fig. 2. The room-temperature resistivity of the BMG was measured to be $(1.75\pm0.02)\times10^{-6}$ Ω m. A weak temperature dependence of the electrical resistivity (ρ) with a negative temperature coefficient was observed, which is consistent with a large number of observations and in accordance with the Mooij criterion.[6] The ρ exhibits a linear temperature dependence at high temperatures then varies as T^2 in the intermediate temperature region, which follows Ziman theory,[12,13] and the experimental data can be well fitted by a polynomial relation,[14]

$$\rho(T) = a + bT + cT^2, \qquad (1)$$

with parameters $a=1.77$, $b=-1.23\times10^{-4}$, and $c=1.19\times10^{-7}$. Below ~30 K, a Kondo-effect temperature dependence of ρ was observed, arising from the dominant scattering at ordering magnetic clusters.[14,15]

Intrinsically different from the monolithic BMG, the composite exhibits very good electrical conductivity and metal-like positive temperature dependence. Figure 3 shows the measured electrical resistivity of the BMG composite at different temperatures along two sample directions (transverse and longitudinal). In both directions, the electrical resistivity decreases linearly with the temperature down to about 70 K. The temperature coefficient of resistivity is estimated to be $8.07\pm0.32\times10^{-4}$ and $1.32\pm0.08\times10^{-3}$ K^{-1} for the transverse and longitudinal directions in the temperature range from 200 to 298 K, which is much larger than that of monolithic Ni-based BMG $(-3.675\pm0.007\times10^{-5}$ K$^{-1})$ and very close to that of brass $(1.54\pm0.14\times10^{-3}$ K$^{-1})$, in particular, along the longitudinal direction. The resistivities of the two directions are about one order of magnitude lower than that of the monolithic BMG in the entire temperature range. For example, at room temperature $(\rho_{298\text{ K}})$ the longitudinal resistivity $(1.63\pm0.02\times10^{-7}$ Ω m) is much lower than that $(1.75\pm0.02\times10^{-6}$ Ω m) of monolithic BMG and is only 2.5 times higher than that $(6.20\times10^{-8}$ Ω m) of high-conductive brass $(Cu_{80}Zn_{20})$. In the composite, the resistivity along the longitudinal direction is about three times smaller than that along the transverse direction. The difference suggests that the conductivity of the BMG based composite is anisotropic and sensitive to the morphology of the high-conductive phase.

The conductivity of binary composites with a randomly distributed high-conductive phase has been found to follow the percolation theory. A generalized effective media (GEM) equation, which combines both percolation and effective media theories, has been applied to a large number of percolation composites.[16] The GEM equation can be expressed as

$$\frac{(1-\phi)(\sigma_l^t-\sigma_m^t)}{\sigma_l^t+[(1-\phi_c)/\phi_c]\sigma_m^t}+\frac{\phi(\sigma_h^t-\sigma_m^t)}{\sigma_h^t+[(1-\phi_c)/\phi_c]\sigma_m^t}=0, \qquad (2)$$

where ϕ is the volume fraction of the high-conductive phase, σ_l the conductivity of the low-conductive phase, σ_h the conductivity of the high-conductive phase, and σ_m the conductivity of the composite. This equation contains two important parameters, i.e., the critical percolation threshold volume ϕ_c of the high-conductive phase at which the conductivity of composite undergoes an abrupt transition from the low-conductive behavior to the high-conductive behavior, and an exponent t, which depends on ϕ_c and a characteristic demagnetization coefficient for the dispersions. Both parameters depend on the shape and orientation of the reinforcement particles. For the present composite, the low conductive phase is BMG matrix and the high conductive phase is the reinforcing brass fibers. There is about two orders of magnitude difference in the conductivity between the two phases. Because ϕ_c and t are temperature independent, they can be determined based on the measured resistivities of the composite, brass, and BMG at different temperatures. By using the resistivity data at room temperature and 70 K, the calculated values of ϕ_c and t are 0.07 and 0.11 along the longitudinal direction and 0.11 and 0.19 along the transverse direction, which are in fair agreement with the observations in a wide range of composites.[16–18] The calculations suggest that the BMG composite has a pretty low percolation threshold (about 10%) and certain anisotropy in conductivity. The validity of Eq. (2) and the calculated ϕ_c and t is verified by modeling the two experimental curves (Fig. 3). According to the known temperature dependence of resistivity for brass $[Cu_{80}Zn_{20}, \rho_l(T)=1/\sigma_h(T)]$ given by[19]

$$\rho_l(T) = 3.64 + 8.75 \times 10^{-3}T \quad (\times 10^{-8}\ \Omega\ \text{m, for } T > 70\ \text{K}), \tag{3}$$

and the temperature dependence of the BMG matrix $[\rho_h(T) = 1/\sigma_l(T)]$ by fitting the experimental data (Fig. 2),

$$\rho_h(T) = 1.77 - 1.23 \times 10^{-4}T + 1.19 \times 10^{-7}T^2 \quad (\times 10^{-8}\ \Omega\ \text{m, for } T > 70\ \text{K}), \tag{4}$$

the temperature dependence of the composite can be theoretically calculated by substituting Eqs. (3) and (4) into Eq. (2) along with the determined ϕ_c and t. The calculated curves are plotted in Fig. 3 and are remarkably consistent with the experimental data along both longitudinal and transverse directions, suggesting that the conductivity of the BMG composite obeys the percolation theory and can be quantitatively described by the GEM equation.

In summary, the conductivity of the Ni-based BMG can be dramatically improved by introducing a high conductive component. Quantitative calculations demonstrate that the composite effect on electrical conductivity obeys the percolation theory and the BMG composite has a low percolation threshold volume. This study has implications for developing improved BMG based materials with both high strength and good conductivity for the applications in pressure sensors and MEMSs.

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