JOURNAL OF APPLIED POLYMER SCIENCE VOL. 20 (1976)

Effect of Metal Concentration on the Electrical Conductivity and Some Mechanical Properties of Poly(methyl Methacrylate)–Copper Composites

Metal–polymer composites have emerged as a new group of engineering materials lately. Possible applications include (a) discharging static electricity, (b) heat conduction, (c) electrical shielding, (d) electrical heating, (e) converting mechanical to electrical signals, (f) absorbing electromagnetic waves, and (g) friction–antifriction materials. The materials are likely to have the following advantages in comparison with metals: high corrosion resistance, lower specific weight, ease of workability when manufacturing products of complex shapes are desired, cheapness, and great accessibility.

Most of the early works on metal–polymer composites are patented, and little systematic investigation has been carried out in this field, specially on the particulate composite systems. Gurland[1] was the first to report that compaction of a mixture of powders comprising a polymeric insulator (Bakelite) and a metallic conductor (silver) gave a product with an electrical resistivity much lower than the insulating polymer alone for compositions of metal in excess of a critical volume of 35% by volume. Microscopic examination revealed that uniform spheres of silver were distributed more or less randomly in the polymer matrix.

Subsequently, it was demonstrated by Turner and co-workers[2–5] that the critical composition of metal could be reduced considerably by segregation of the metallic particles. Segregation was effected by admixture of large particles of polymer with smaller particles of metal followed by compaction. They made preliminary investigations on PVC/Ni, polyethylene/Ni, and polyurethane/Ni systems. In the present communication, we report results of our studies on poly(methyl methacrylate)–copper composites.

Poly(methyl methacrylate), hereafter referred to as PMMA, was obtained from Fisher Scientific Co. and had the following characteristics: (a) density, 1.202 g/cc; (b) molecular weight, 3.1×10^5 (viscosity method); (c) particle size distribution, 150 to 75 μ, 62%; 75 to 50 μ, 33%, and <50 μ, 5%. Electrolytic copper powder was obtained from Assam Carbon Products Pvt. Ltd. and had the following characteristics: (a) density, 8.82 g/cc; (b) electrical resistivity, 1.7×10^{-6} ohm-cm; (c) particle size distribution, 150 to 75 μ, 10%; 75 to 50 μ, 26%, and <50 μ, 64%.

Weighed amounts of PMMA and copper powders were mixed by tumbling until a homogeneous product was obtained. The mixture was compacted in a steel cylindrical die of height 5.0 cm and diameter 2.5 cm under a pressure of 100 kg/cm^2 for 10 min at 145°C. The die was then cooled to 70°C by a high-speed fan. The compacted sample was ejected from the die and allowed to cool to room temperature. These cylindrical samples were cut into small pieces of specified dimensions for use in measurements of electrical and mechanical properties and for metallographic studies. The composites can be easily machined with a lathe.

Prior to measurements of electrical resistivity, the samples were kept under vacuum for at least 48 hr to minimize any effect due to moisture. Measurements of high resistivity were made using a vibrating-read electrometer (Type No. 616B). Measurements of low resistivity were made using a Simpson 260 volt-ohm-multimeter and a Universal d.c. meter. The compressive strength and the transverse rupture strength were measured with a Universal Testing Machine (Trevel-Werk, Duesselderf, Model UPM 4). The load was applied at a rate of 2.5 cm/min. The microhardness on the polymer surface of the composite was measured by a Soviet-made Microhardness Tester (Model PMT-3). Section of the composites were polished and examined by Soviet-made MEEM-8M Horizontal Metallographic Microscope.

Figure 1 shows the plot of log (resistivity) against metal loading. At very low loadings, the composite resistivity is essentially of the insulating polymer (about 10^{13} ohm-cm). As the metal loading reaches 12% by volume, a sharp fall in resistivity occurs. At much higher loading (20% by volume and above), the resistivity decreases rather slowly. Optical photomicrographs of different composites are shown in Figure 2. At 5 vol-%, the metal particles are randomly distributed; at 10 vol-%, the metal particles tend to form segregated networks; and at 20 vol-%, the formation of metallic networks is complete. The sudden drop of resistivity around 12 vol-% is possibly due to formation of continuous conducting paths.[2–4]

Figure 3 shows the plot of microhardness on the polymer surface against metal loading in the

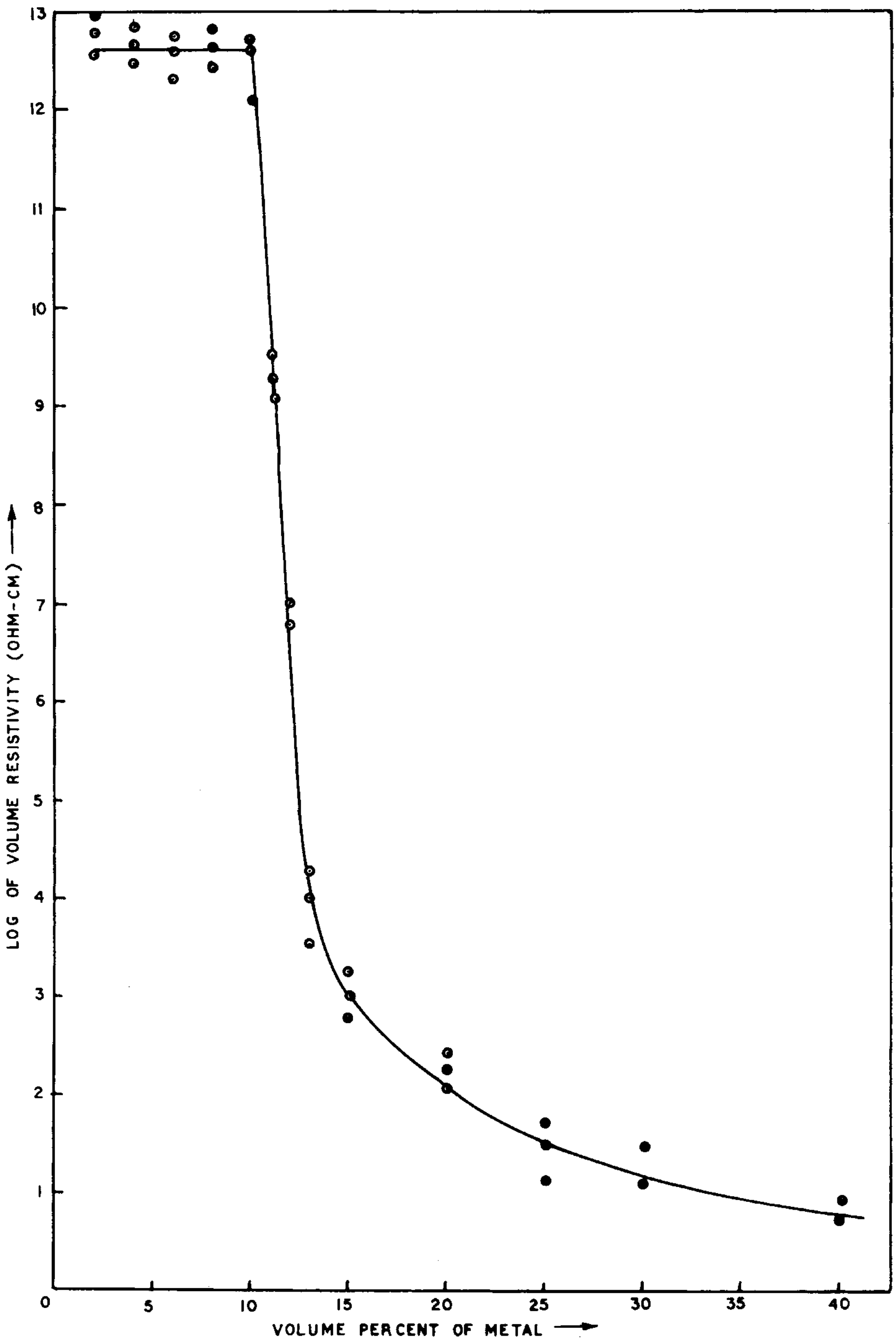
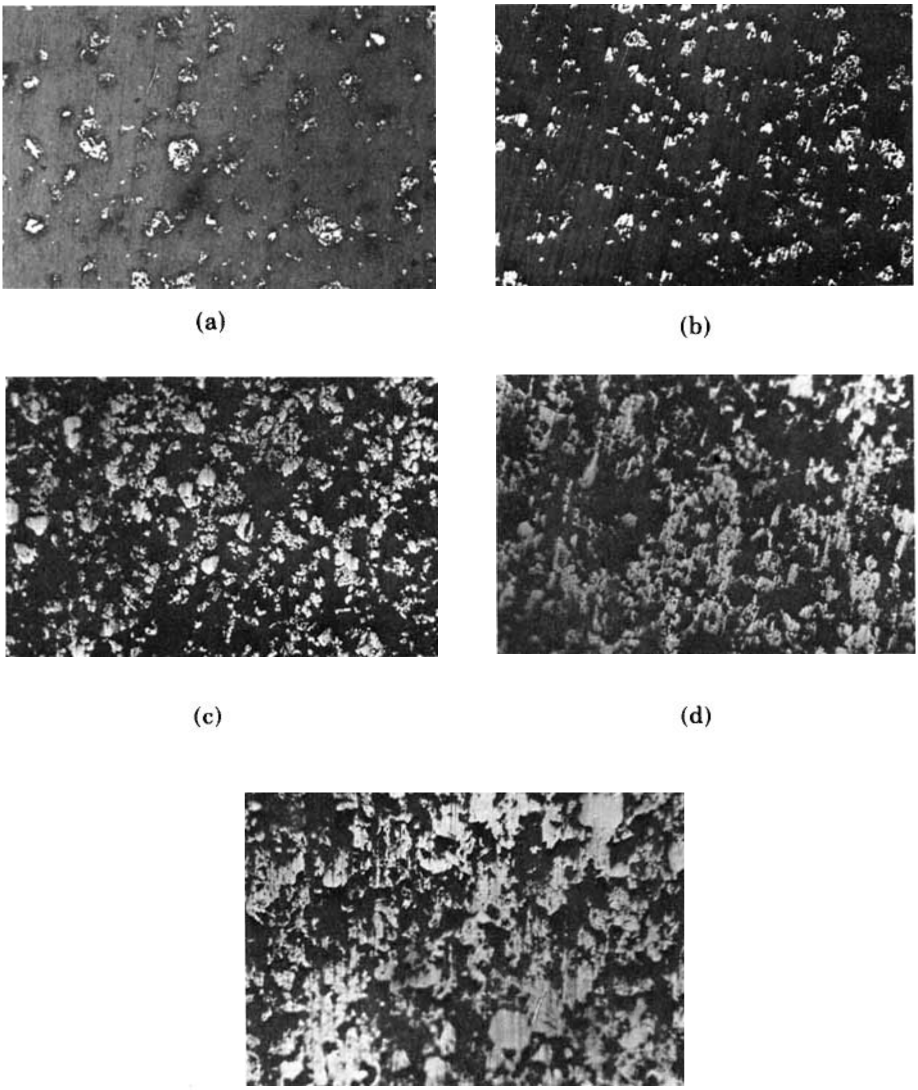


Fig. 1. Resistivity as a function of metal loading.



(a) (b) (c) (d) (e)

Fig. 2. Photomicrographs of PMMA/Cu composites (60×): (a) 5% Cu; (b) 10% Cu; (c) 20% Cu; (d) 30% Cu; (e) 40% Cu.

composite. Microhardness increases with increase in metal concentration, but beyond 20% by volume, it becomes almost constant. Under the compaction conditions, the polymeric particles in the composite composition do not coalesce completely, and metal particles activate the coalescence phenomenon by improved heat transfer.[4] As the coalescence increases, the polymer matrix loses porosity and the hardness increases. At 20 vol-% of metal and above, when segregated networks of the metallic particles are well established, the coalescence reaches the saturation point with no further increase in microhardness.

Variation of compressive strength with metal concentration is shown in Figure 4. Compressive strength decreases sharply with increase in metal loading up to 20% by volume, above which compressive strength becomes almost constant. At high metal loading, fracture under compressive stress occurs possibly through the metal–metal contact points, which requires less energy

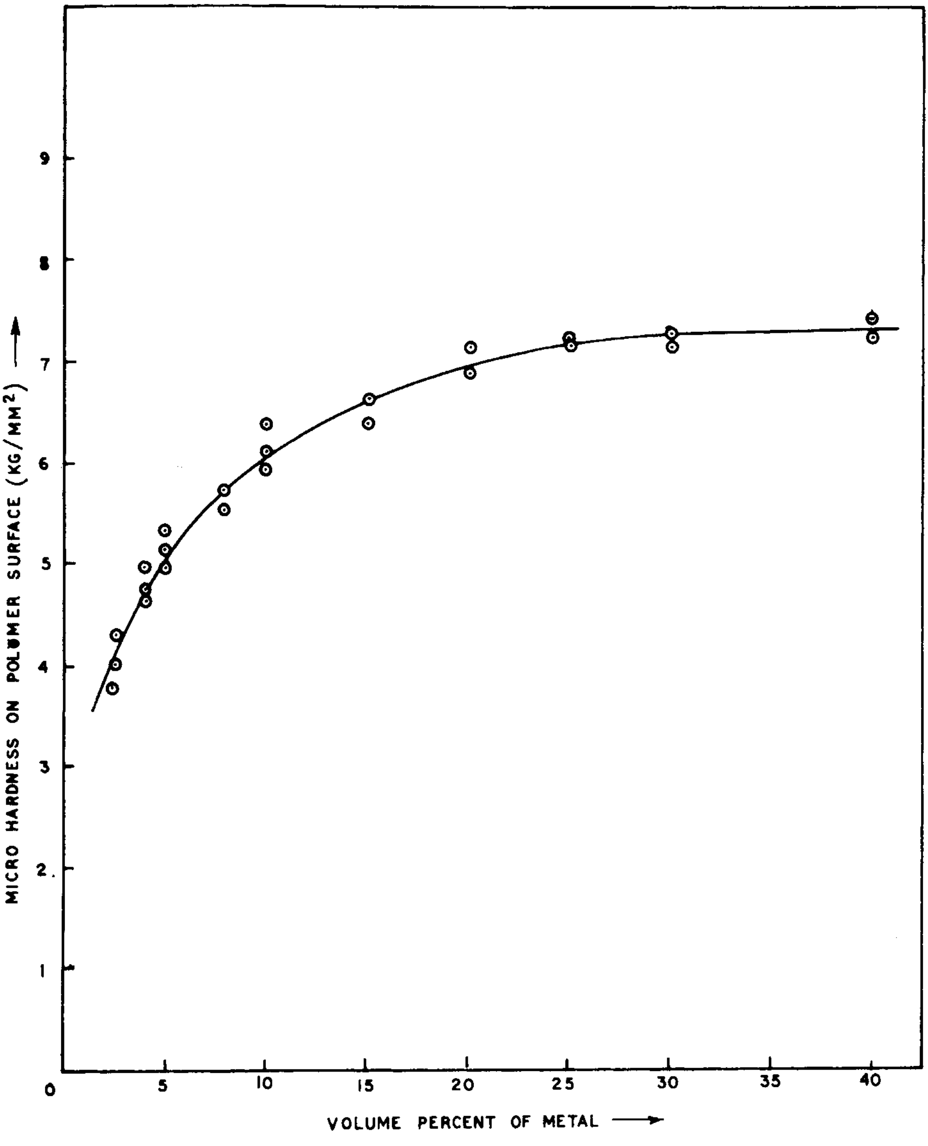


Fig. 3. Microhardness on polymer surface as a function of metal loading.

than for a fracture to occur through metal–polymer surface at low metal loading. Above 20 vol-%, the formation of segregated networks is complete and the compressive strength attains almost a constant value. It is to be noted that the compressive behavior of the composite follows the same pattern as the electrical resistivity since both, as explained, depend upon metallic contacts.

A plot of transverse rupture strength against metal loading in the composite is shown in Figure 5. Up to 10 vol-% of metal, coalescence of the polymeric particles is not yet complete and a section of the polymer matrix is occupied by the metal particles, resulting in decrease of transverse rupture strength with increase in metal loading. Above 10 vol-%, matrix hardening, as revealed by microhardness studies, overtakes the section effect and the strength increases. As the metal loading reaches 20 vol-%, metal–metal contacts are established; and at very high metal loading, the strength decreases rather sharply, approaching the green strength of the metal powder compacts.

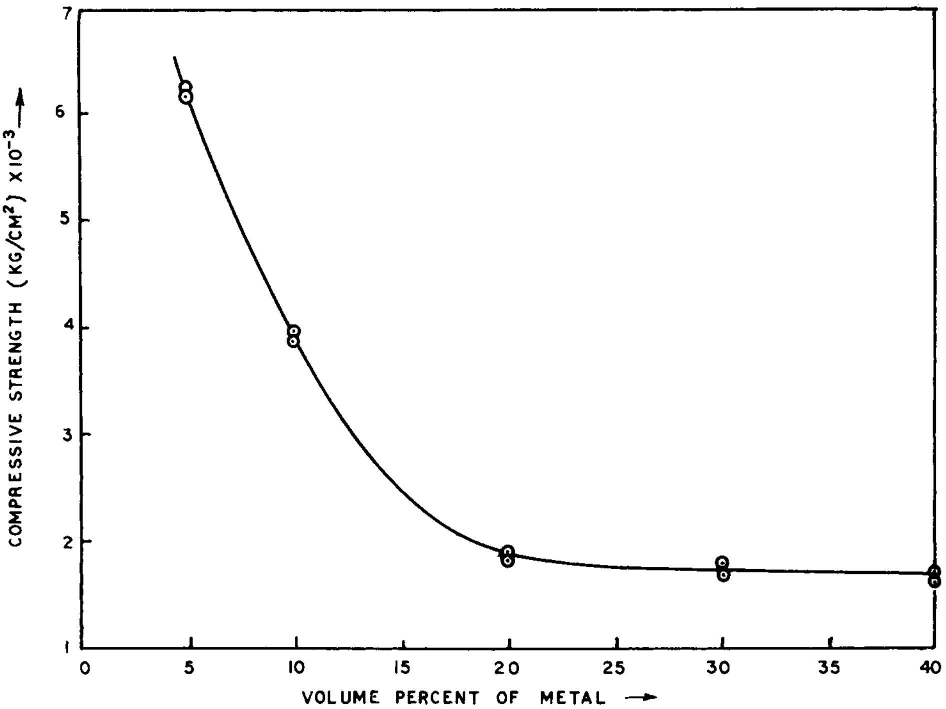


Fig. 4. Variation of compressive strength with metal concentration in the composite.

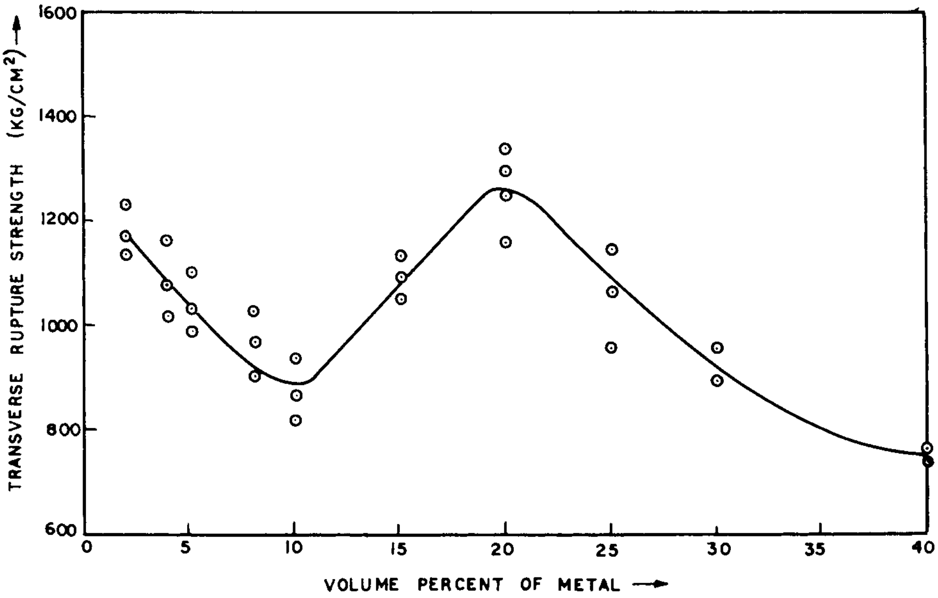


Fig. 5. Variation of transverse rupture strength with metal concentration in the composite.

References

1. J. Gurland, *Trans. Met. Soc. AIME,* **236,** 642 (1966).
2. A. Malliaris and D. T. Turner, *J. Appl. Phys.,* **42,** 614 (1971).
3. R. P. Kusy and D. T. Turner, *J. Appl. Polym. Sci.,* **17,** 1631 (1973).
4. R. P. Kusy and D. T. Turner, *SPE J.,* **29,** 56 (1973).
5. J. E. Scheer and D. T. Turner, *Advan. Chem.* **99,** 572 (1971).

RABINDRA MUKHOPADHYAY
SADHAN K. DE
SANJAY BASU

Department of Chemistry and
Materials Science Centre
Indian Institute of Technology
Kharagpur-2, India

Received October 9, 1975
Revised December 4, 1975