Effect of Filler Type and Concentration on Some Mechanical and Electrical Properties of Poly(methyl methacrylate)

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ABSTRACT: Polymer composites of poly(methyl methacrylate) (PMMA) and cuprous oxide (Cu_2O), magnesium oxide (MgO), copper biphthalocyanine, iron (II) chloride ($FeCl_2$), and iron metal were prepared by different filler-to-polymer percentages of 10, 20, 30, 40, and 50%. With increasing filler concentration, brinell hardness increases. Copper biphthalocyanine resulted in the highest hardness, and iron metal resulted in the lowest one. The electrical conductivity was found to increase with increasing filler concentration. The iron metal composite gave the highest effect, while the magnesium oxide composite resulted in the lowest one. The 50% concentration samples were subjected to electrical conductivity measurements at temperatures ranging from 25 to 100°C. The electrical conductivity was found to increase with increasing the temperature. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci **67:** 637–641, 1998

Key words: PMMA; composite; electric conductivity; filler; hardness

INTRODUCTION

Particulates (fillers) are added to the polymer matrix for a number of reasons. Some have a synergistic effect; and particulates may reduce costs, lower the coefficient of linear expansion, reduce shrinkage, reduce molding cycles, increase thermal conductivity, and lower resistivity.[1] Mechanical properties of filled polymer are affected by size, shape, and orientation of fillers, in addition to the adhesive strength between polymer and filler.[2] Shifts in glass transition temperature T_g to a higher temperature as a function of filler concentration have been reported for composites involving a wide variety of polymers and fillers.[3–7] The purpose of this article is to investigate the effect of filler type and concentration on the mechanical hardness and the electrical conductivity of the filled poly(methyl methacrylate) (PMMA).

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EXPERIMENTAL

The polymer composites of PMMA and Cu_2O, MgO, copper biphthalocyanine, iron (II) chloride, and iron metal were prepared by different filler to polymer percentages of 10, 20, 30, 40, and 50%. The polymer PMMA and the filler were ground and sieved to 250 microns separately; then they were mixed in a small mortar, where they were subjected to further grinding and mixing until becoming a uniform solid mixture, then molded at 160°C and compressed at 38 N/mm^2 for 10 min. A rectangular disc was obtained of thickness about 0.3 cm, a length of 2 cm, and a width of 1 cm.

The PMMA used in the preparation of composites was prepared by the emulsion techniques. It has a viscosity average molecular weight of 3×10^5 g mol. Methylmethacrylate monomer, a product of Merck Schuchardt (yield 99%) stabilized with 14 ppm hydroquinone [specific gravity at 20/4°C (0.942–0.944)] was purified by washing with a small amount of sodium hydroxide solution (3%), then with distilled water until free from alkali. The MMA was separated with a separatory

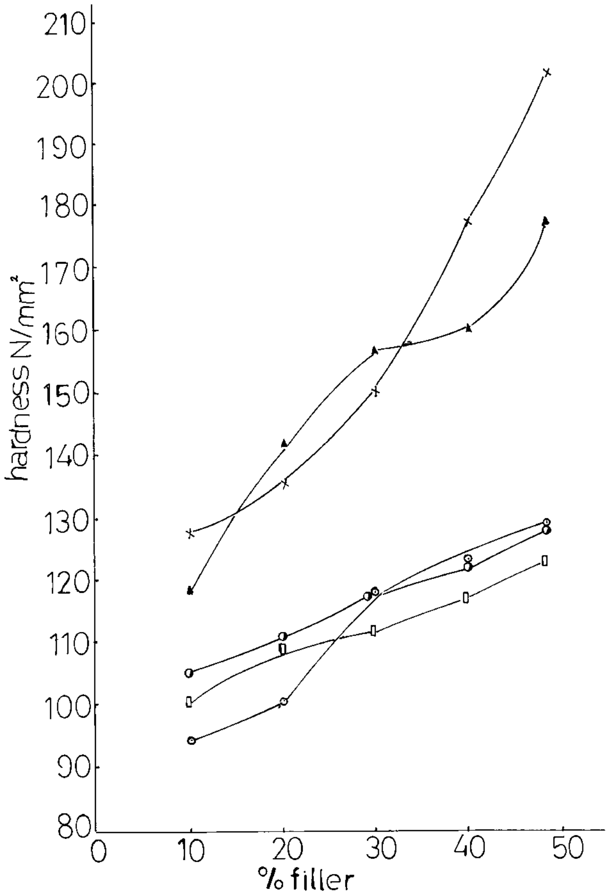

Figure 1 Hardness of PMMA composites: (⊙) Cu_2O, (▲) MgO, (×) copper biphthalocyanine, (◑) $FeCl_2$, and (□) iron metal.

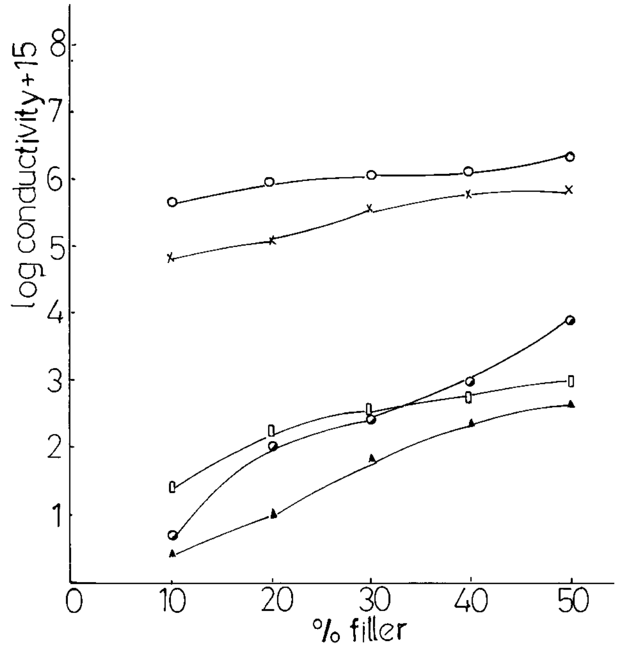

Figure 2 Electrical conductivity of PMMA composites: (◑) Cu_2O, (△) MgO, (□) copper biphthalocyanine, (×) $FeCl_2$, and (○) iron metal.

funnel, then dried over anhydrous sodium sulphate. Sodium bisulphite, sodium sulphate, iron metal powder, iron (II) chloride, and cuprous oxide are from El Nasr Pharmaceutical Company, a product of Riedel de Haen (A.G. D 3016 sleeze 1).

Sodium hydroxide and hydroquinone are products of Merck Schuchardt. Copper biphthalocyanine is prepared according to the method described in a paint technology manual by the urea fusion technique.[8]

RESULTS AND DISCUSSION

Figure 1 shows the effect of filler type and concentration percentage on the mechanical hardness of PMMA composites with (Cu_2O, MgO, $FeCl_2$, iron metal, and copper biphthalocyanine). The data given in Table I show that the mechanical hardness is directly proportional to the filler concentration and the degree of propertionality depends on the filler type. The highest increase was obtained

Table I Brinell Hardness of Different PMMA Composite Materials of Different Filler Percentages

Concentration (%)	Brinell Hardness (N mm^2)				
	Cu_2O	MgO	Copper Biphthalocyanine	$FeCl_2$	Iron
10	94.5	118	127.5	105	101
20	101.3	142	135	111.5	109
30	118.2	157	150	118	112
40	123	160	177.5	122	117
50	129	177	202	128	123

Table II Conductivity Measurement of PMMA Composites of Different Filler Percentages

Concentration (%)	Conductivity Ω^{-1} cm^{-1} $\times 10^{13}$				
	Cu_2O	MgO	Copper Biphthalocyanine	$FeCl_2$	Iron
10	0.05	0.03	0.26	670	4950
20	1	0.1	1.8	1250	9650
30	2.6	0.75	3	4140	11600
40	9.6	2	5.95	5200	15400
50	80	4	6.5	6000	19650

in the case of copper biphthalocyanine, as when its concentration increased from 10 to 50%; the hardness increased from 127.5 to 202 N mm^2 (blank PMMA gave a hardness of 102 N mm^2).

The least effect was in the case of iron metal, as the hardness increased from 101 to 123 N mm^2 when its concentration increased from 10 to 50%.

Cuprous oxide and iron chloride (II) composites

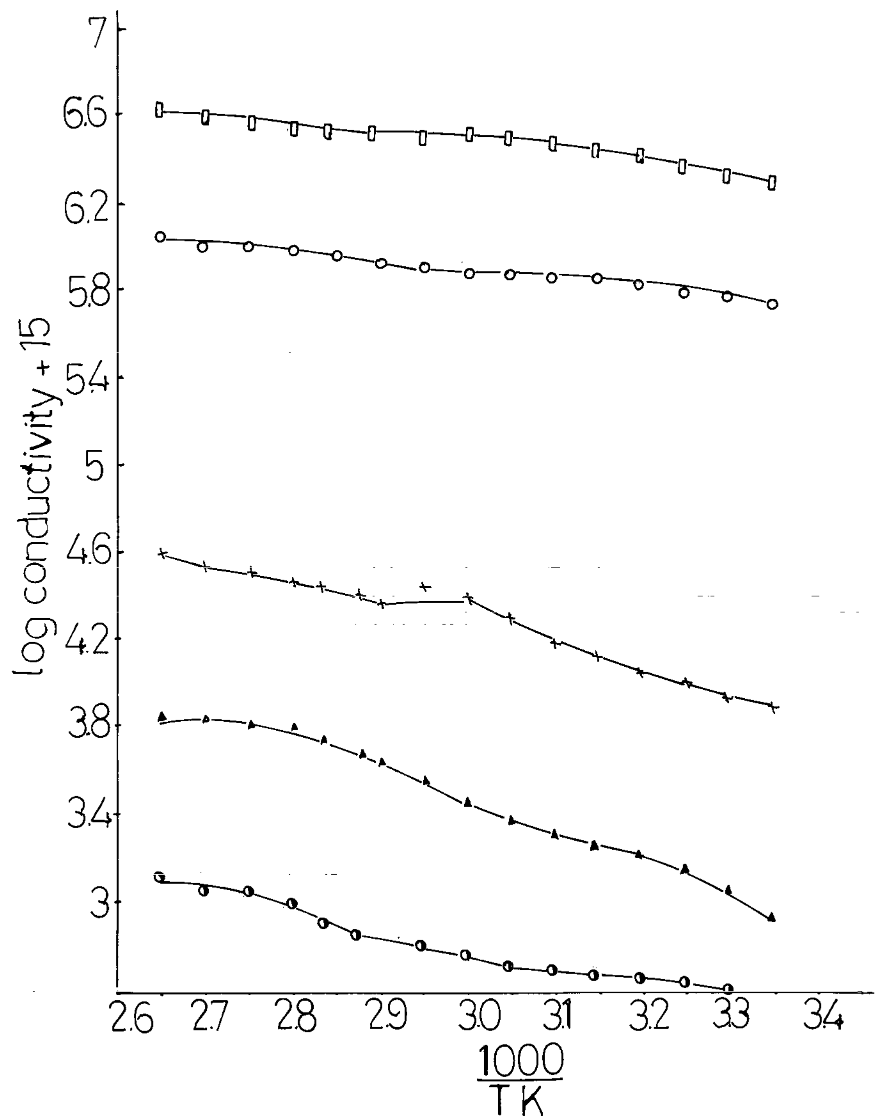

Figure 3 Temperature dependence conductivity of PMMA composites: (×) Cu_2O, (◑) MgO, (▲) copper biphthalocyanine, (○) $FeCl_2$, and (□) iron metal.

Table III Conductivity Measurements for Different PMMA Composites Containing 50% of the Filler at Different Temperatures

Temp (°C)	Conductivity of 50% Filler PMMA Composite $\times 10^{13}\ \Omega^{-1}$ Cm^{-1}				
	Cu_2O	MgO	Copper Biphthalocyanine	$FeCl_2$	Iron
25	80	4	8.5	6000	19650
30	90	4	11.3	6200	22560
35	103	4.3	15	6450	25000
40	111	4.6	17	7100	27500
45	140	4.7	18.7	7200	27900
50	150	4.9	21.3	7430	31000
55	197	5.2	24	7900	32000
60	255	5.9	30	8300	35600
65	290	6.3	36.5	8600	36000
70	234	6.5	43.5	9100	36500
75	265	7.2	48.5	9150	37100
80	295	7.8	55	9500	37200
85	302	9.2	64.5	9810	38600
90	340	11.3	66	10000	39500
95	371	12	70	10150	40000
100	412	13.4	72.5	11000	42000

gave an approximately equal increase in mechanical hardness as the concentration of filler increased from 10 to 50%. Magnesium oxide gave a higher hardness increment than in the case of cuprous oxide, which indicates that there is some sort of interaction between the polymer and magnesium oxide filler.

The highest hardness of PMMA–copper biphthalocyanine composite is attributed to the lipophilic part of copper biphthalocyanine, which led to the complete compatibility with PMMA; consequently, the polymer composite molecules are very compact, and this resulted in the higher hardness observed. In case of the iron–PMMA composite, the compatibility is the least one; this resulted in the least hardness value.

Figure 2 shows the effect of filler type and concentration on the electrical conductivity of PMMA composites with Cu_2O, MgO, $FeCl_2$, iron metal, and copper biphthalocyanine. The data given in Table II show that these fillers increase the conductivity of pure PMMA. The highest effect was in the case of iron metal powder, which showed a greater increase in the electrical conductivity than in case of PMMA alone; it increased from 10^{-15} to 19.65×10^{-10} ohm^{-1} cm^{-1} when the iron metal concentration increased from 0 to 50%.

MgO (50%) gave the least effect as it increased the electrical conductivity to 0.03×10^{-13} ohm^{-1} cm^{-1}. The order of increasing the electrical conductivity of the composite was as follows: Fe $> FeCl_2 > Cu_2O >$ copper biphthalocyanine $>$ MgO. These results are attributed to the high flux of electrons transfer of the iron metal powder compared to that of the rest of the other fillers.

Figure 3 shows the temperature dependence of the electrical conductivity. The conductivity increased with a rise in the temperature (Table III). The iron metal–poly(methyl methacrylate) composite has the highest effect as the conductivity increased from 19.65×10^{-10} to 42×10^{-10} ohm^{-1} cm^{-1} when the temperature increased from 25 to 100°C. The effect of temperature on the electrical conductivity of the PMMA composites is explained in terms of the general exponential law of polymer conductivity, as follows:

$$X = A \exp(-\Delta u/RT)$$

where X is the electrical conductivity, A is the coeffecient weakly dependent on the temperature ($A \approx 1/T$), and R is the universal gas constant. ΔU is the energy difference between the active and the initial state.

CONCLUSION

Poly(methyl methacrylate) composites were prepared using different types of fillers, Cu_2O, MgO, copper biphthalocyanine, iron II chloride, and iron metal, at different filler-to-polymer concentrations of 10, 20, 30, 40, and 50%. Brinell hardness was found to increase with increasing filler concentration.

The electrical conductivity was found to increase with increasing filler concentration. In addition, the electrical conductivity of the composites of the filler concentration (50%) was found to increase with increasing temperature from 25 to 100°C.

REFERENCES

1. T. Richardson, *Composites: A Design Guide,* Industrial Press, New York, 1987.
2. K. Hasaka and K. Shibayama, *J. Appl. Polym. Sci.,* **22,** 132 (1978).
3. R. F. Landel, Trans. Soc. Rheol., **2,** 53 (1958).
4. R. F. Landel and T. L. Smith, *Rubber Chem. Technol.,* **35,** 291 (1962).
5. L. Galperin, *J. Appl. Polym. Sci.,* **11,** 1475 (1967).
6. A. Yim and L. E. St. Pierre, *Polym. Lett.,* **B7,** 237 (1969).
7. Y. S. Lipatov and F. Y. Fabulyak, *J. Appl. Polym. Sci.,* **10,** 2131 (1972).
8. J. Occa, *Paint Technology Manual,* Part VI, Wax Chandlers' Hall, London, 1966.