Relation Between Electrical and Mechanical Properties of Conducting Polymer Composites

IVAN CHODÁK,[1] MÁRIA OMASTOVÁ,[1] JÜRGEN PIONTECK[2]

[1] Polymer Institute, Slovak Academy of Sciences, Dúbravská cesta 9, 842 36 Bratislava, Slovak Republic

[2] Institute of Polymer Research Dresden e. V., Hohe Strasse 6, D-01069 Dresden, Germany

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ABSTRACT: The influence of the carbon black content on the mechanical and electrical properties of polypropylene/carbon black composites prepared by different processing procedures was investigated. The formation of a continuous conducting network in the insulating matrix and, consequently, the percolation threshold depend strongly on the processing route and influence both the mechanical and electrical properties of the prepared composites. An interesting coincidence of the dependencies of conductivity and elongation at break on the filler content was found. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 82: 1903–1906, 2001

Key words: composites; fillers; mechanical properties; polypropylene; processing

INTRODUCTION

Electrically conductive composites with a thermoplastic matrix are considered to be an important group of relatively inexpensive materials for special applications. Conductivity in a highly insulating matrix is achieved by the admixtures of conductive additives, for example, fine metal powders or conductive polymers.[1–4] Carbon-based fillers (carbon black, graphite, or carbon fibers) are the most frequently used conductive fillers because of their high conductivity, relatively low price, and good ultimate and processing properties of the composites.[5–7]

A proper balance among the electrical conductivity, mechanical properties, and processing characteristics is an important requirement for the design of electroconductive thermoplastic composites. It is well known that, at low loading of the conductive filler, a modest conductivity increase is observed with an increasing filler content. A sudden conductivity increase occurs in a relatively narrow concentration range around the so-called percolation threshold. At this concentration, a conductive network is formed within the insulating phase and a dramatic increase in conductivity by several orders of magnitude is observed. This phenomenon was broadly studied and well described in numerous articles and several reviews.[8–10] In spite of this, after consideration of various statistical, geometric, and thermodynamic percolation and conductivity models, no theory exists which can explain all the experimentally observed results,[10] among them also the effect of processing procedures on the percolation phenomenon.

The presence of a filler in a thermoplastic matrix has a significant effect on the behavior of the material, especially regarding mechanical properties and processing characteristics. The presence of a filler leads usually to higher stiffness of the material, demonstrated by an increase in the Young's modulus and a decrease in deformability and toughness. The extent of these changes depends on the concentration and on the parame-

Correspondence to: I. Chodak (upolchiv@savba.sk).

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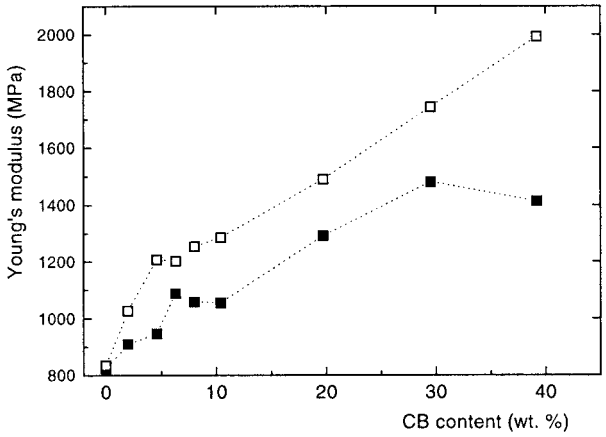

Figure 1 Comparison of the Young's modulus of PP/CB composites prepared by (□) injection molding and (■) by compression molding at different filler concentrations.

ters of the filler, especially on the filler surface area, which is decisive for the filler-reinforcing ability. An addition of the filler results always in a more difficult processing because of the increase of the melt viscosity. The above-mentioned effects are rather pronounced if carbon black is added as a conductive additive because of the highly reinforcing nature of the filler.

In this article, the electrical and mechanical properties of polypropylene/carbon black (PP/CB) composites were investigated. A comparison of the properties of materials prepared by injection molding and by compression molding was done.

EXPERIMENTAL

Materials

PP (Tatren TF-411, MFI = 10 g/10 min, $M_w = 2.13 \times 10^5$ g/mol, M_n = 25 100 g/mol, Slovnaft, Bratislava, Slovak Republic) and CB (VULCAN® XC-72R, Cabot Corp., Billerica, MA) were used as received.

Composite Preparation

PP/CB composites were prepared by the mixing of PP with CB in a 50-mL mixing chamber at 75 rpm for 10 min at 200°C using a Plasti-Corder kneading machine PLE 330 (Brabender, Germany).

Analytical Methods

The exact weight percentage of CB in the composites was determined using thermogravimetry with a TG-7 (Perkin–Elmer, USA). For the electrical conductivity measurements, the prepared composites were compression-molded at 200°C for 2 min under 22.5 kN/cm^2. The thickness of the compression-molded samples was about 0.2 mm. The electrical conductivity in the polymer composites was measured by a standard four-probe method. Samples were cut into strips of about 5 $\times$ 25 mm^2. Silver paste was used to achieve good electrical contacts of the sample surface with the electrodes of the conductivity tester. All measuring instruments (for current and voltage) were connected to a PC to collect and calculate the data.

The mechanical properties of the samples were determined using a testing instrument Instron 4301 (U.K.) for specimens obtained by injection molding using a low mass injection unit ES 200 H/80 V/50 HL-2F (Engel, Germany) or by compression molding at 200°C for 10 min under 22.5 kN/cm^2. The shape of the injection-molded dumbbell test pieces was 85 $\times$ 15 $\times$ 1 mm with a gauge section of 26 $\times$ 6 $\times$ 1 mm. The compression-molded dumbbell specimens with a gauge section of 30 $\times$ 3.5 $\times$ 1 mm were cut from a plate.

RESULTS AND DISCUSSION

The dependencies of the tensile mechanical properties on the filler content for the PP filled with CB are shown in Figures 1–3. It is seen that the

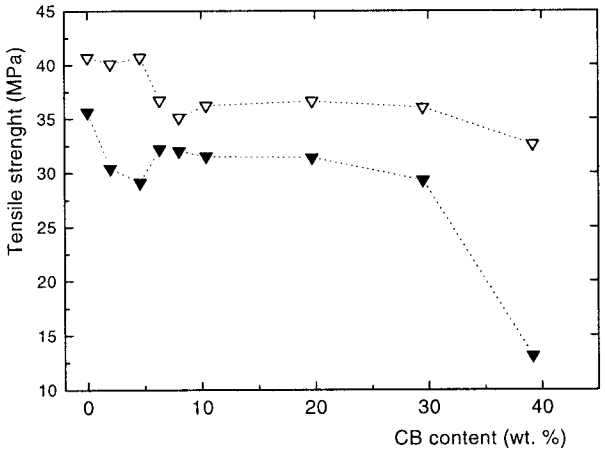

Figure 2 Comparison of the tensile strength of PP/CB composites prepared (▽) by injection molding and (▼) by compression molding at different filler concentrations.

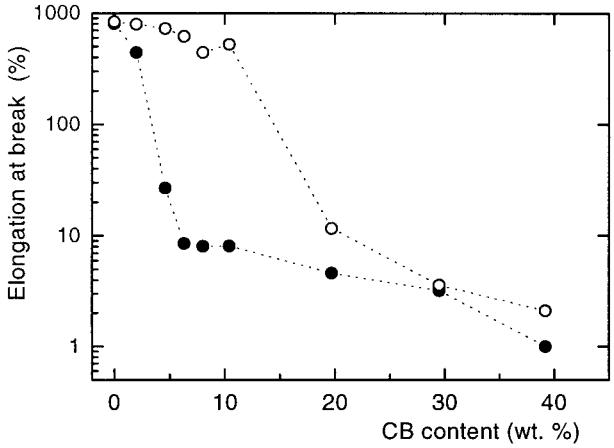

Figure 3 Comparison of the elongation at break of PP/CB composites prepared (○) by injection molding and (●) by compression molding at different filler concentrations.

properties of injection-molded materials differ from those of the compression-molded composites.

An increase in the Young's modulus values is observed with increasing filler content (Fig. 1). The increase is more pronounced for injection-molded composites compared to compression-molded materials, although a negligible difference was found for virgin PP. The effect can be attributed to better distribution of the filler in the PP matrix due to additional compounding under shear in the injection-molding equipment. An orientation due to elongational flow during injection molding may also play a role, while compression-molded samples are considered to be isotropic. A decrease in the Young's modulus for compression-molded samples at the highest filler content can be interpreted in terms of less homogeneous dispersion in the compression-molded samples.

A decrease of the tensile strength with increasing filler content was found for both injection-molded and compression-molded samples (Fig. 2). The trend can be explained by a diminishing of and later vanishing of the orientational strengthening due to lower deformability of the material with increasing filler content. The composites with higher CB content break before they are able to achieve a significant degree of orientational reinforcing due to drawing during the tensile tests. Again, the tensile strength of the compression-molded samples is lower than that of the injection-molded materials, indicating the presence of larger or more frequent defects (such as voids, cracks, or stress-concentration spots) due to presumably less perfect CB dispersion. Larger defects or their more frequent occurrence can lead to a formation of critical cracks at lower deformation. As can be seen in Figure 2, the decrease in tensile strength is observed at a rather low filler content, when the relative increase in the defect size and number is high. Further increase in the filler content is less detrimental; some of the strength is even gained back as a result of the reinforcing effect of the filler.

The above-mentioned trends are most clearly demonstrated by the dependencies of the elongation at break on the filler content (Fig. 3). A substantial decrease in the deformability of compression-molded samples occurs at a filler content less than 10 wt %. The decrease is less pronounced for injection-molded material, although, in this case, the drop is also rather steep. The changes can be interpreted in terms of the defect formation as a result of the addition of small amounts of the additive. The shape of the deformation versus the filler content curves is in accordance with the tensile strength variation, considering the prevailing effect of the defect formation at a lower filler content and the increased importance of the reinforcing effect of the filler at higher concentrations. The differences in compression- versus injection-molded samples can be explained in the same way.

An apparent coincidence of the dependencies of elongation at break and conductivity on the filler content is of particular interest. It was mentioned recently[11] that the steep drop in the elongation at break occurs within the concentration range around the conductivity percolation threshold. Figures 4 and 5 demonstrate this effect in both types of PP/CB composites. Although this obser-

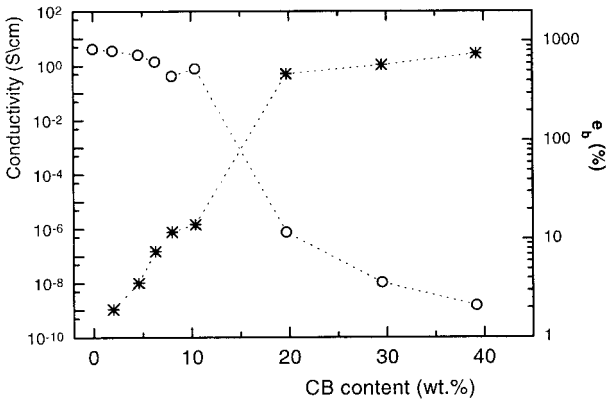

Figure 4 (*) Conductivity and (○) elongation at break of PP/CB composites prepared by injection molding as a function of the CB content.

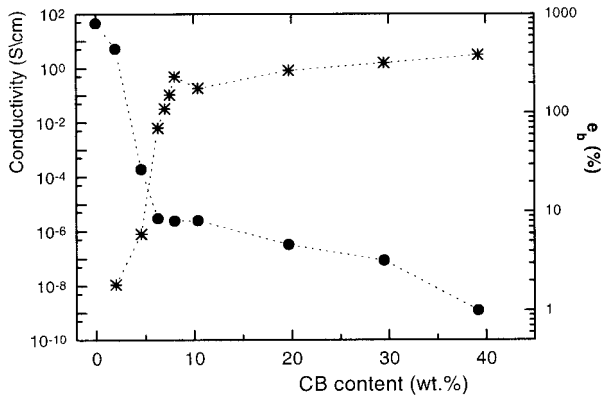

Figure 5 (*) Conductivity and (●) elongation at break of PP/CB composites prepared by compression molding as a function of the CB content.

vation is phenomenological at present, it is quite acceptable that the formation of the conductive network, leading to a substantial increase in conductivity, has a pronounced effect on the mechanical properties via a formation of a more or less continuous filler phase. The critical crack formation and unstable crack growth in such a system is expected to be much easier and faster than in the virgin polymer matrix or in the material where the filler is present but the continuous network has not been formed yet. These considerations are supported also by differences between injection- and compression-molded materials regarding both conductivity and deformation dependencies on the filler content. In the compression-molded samples, the CB is less perfectly distributed in the PP matrix. This leads to the formation of conducting CB "channels" already at lower concentrations, which leads to higher conductivity, on the one hand, but also to the formation of more frequent failure sites, on the other hand.

CONCLUSIONS

An evident difference was observed in the mechanical properties and electrical conductivity of PP/CB composites when comparing the materials prepared by injection molding and compression molding. For injection-molded materials, better mechanical properties but lower electrical conductivity were found compared to compression-molded composites with the same filler content.

A coincidence of a sudden increase in the electrical conductivity (percolation threshold) and a dramatic decrease in the deformation at break was observed for the dependencies on the filler content. The effect is explained in terms of the formation of a continuous network consisting of filler particles, which has a positive effect on the electrical but a negative effect on the deformational behavior of the material.

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