



Influence of carbon nanofiber structure on polyethylene nanocomposites

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Nanofiber morphology significantly affects the electrical and mechanical properties of linear, low-density, polyethylene/carbon nanofiber (LLDPE/CNF) composites.

Carbon fibers provide superior electrical conductivity and mechanical properties to polymer matrices.^{1,2} However, in micro-molding or film applications (product thickness $\sim 25\mu\text{m}$), chopped commercial carbon fibers cannot be used since the diameter of such fibers is $\sim 10\mu\text{m}$ (and length is $\sim 100\mu\text{m}$). Instead, nanofibers may be used since the diameter of these nanoreinforcements is only $\approx 0.1\mu\text{m}$. Therefore, recent studies have addressed the electrical and mechanical properties of carbon nanofiber (CNF) reinforced polymer composites.³

Two different types of CNFs were used for this study: one research grade (MJ) and one commercial grade (PR-24-PS). From x-ray diffraction, we could not detect a significantly different graphitic crystallinity between the two types of CNFs.⁴ We performed a morphological study to determine the effect of the surface structure of CNFs on electrical and mechanical properties of LLDPE/CNF composites.

At 15wt% MJ fiber content, the volume resistivity decreased by more than five orders of magnitude, to $9 \times 10^3 \Omega\text{-cm}$. In contrast, for PR-24-PS composites, the volume resistivity decreased significantly only at a higher content of 25wt% (see Figure 1). The intrinsic volume resistivity of nanofibers in bulk was not significantly different: $0.33\Omega\text{-cm}$ and $0.43\Omega\text{-cm}$ for PR-24-PS and MJ fibers, respectively. It seems likely that differing nanofiber microstructures cause the different electrical behavior of the various composites. Twisted/coiled MJ fibers are more likely to contact each other than straight PR-24-PS fibers are, and this higher degree of connectivity of MJ fibers can result in a lower percolation threshold and reduce the volume resistivity of MJ composites (see Figure 2).^{5,6}

The average modulus, tensile strength, and strain-to-failure values for all of the composites are summarized in Figure 3a–c. The increase

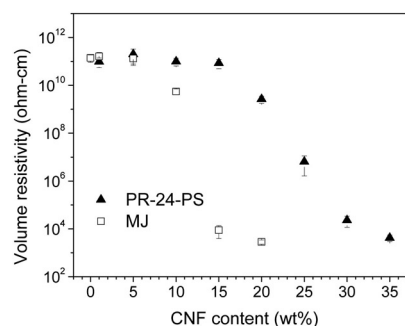


Figure 1. Volume resistivity of pure LLDPE and its composites. Error bars represent 95% confidence intervals. PR-24-PS: Commercial-grade carbon nanofiber (CNF). MJ: Research-grade CNF.

in the modulus was more significant for PR-24-PS fibers (rather than MJ fibers), and is likely a result of their straight shape (see Figure 3a). For all composites, increasing the CNF content reduced the strain-to-failure percentage (see Figure 3c). However, for composites containing MJ fibers, even at 15wt% fiber content, values as high as 460% were measured. In contrast, the 15wt% PR-24-PS composite showed only about 100% strain-to-failure.

It is likely that the interfacial interaction of MJ fibers with the LLDPE matrix is greater than that of PR-24-PS fibers because MJ fibers possess a rougher surface. During deformation, e.g., tensile testing, this interfacial interaction of MJ composites, formed as a mechanical interlock, may prevent fibers from slipping past the LLDPE matrix, leading to elongation in an interphase area. After sufficient elongation of LLDPE in the interphase area, tensile stress transfers to the bulk matrix, which leads to a high strain-to-failure. In contrast, PR-24-PS fibers possessing a smooth surface have a poor interfacial interaction with LLDPE. The interfacial interactions break before LLDPE is fully stretched during tensile testing. This interfacial failure acts as defects,

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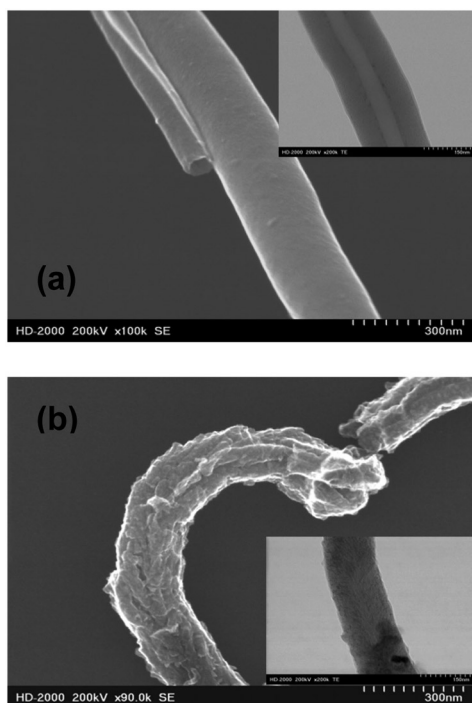


Figure 2. Scanning transmission electron micrographs (TEMs) of: (a) PR-24-PS and (b) MJ carbon nanofibers. The inset images are TEM micrographs.⁵

which propagate so that PR-24-PS composites fail earlier than their MJ counterparts.

In summary, we found that incorporating of CNFs into LLDPE using intensive mixing led to a fairly uniform dispersion of CNFs. A significant drop of volume resistivity occurred in nanocomposites at 15 and 25wt% of MJ and PR-24-PS fiber content, respectively, although intrinsic resistivities of CNFs were not significantly different. It was inferred that twisted/coiled MJ nanofibers led to better inter-fiber connectivity and significantly reduced resistivity, compared to that for fairly straight PR-24-PS nanofibers. Further, this morphological difference of CNFs contributed to a higher tensile modulus of PR-24-PS composites than that of MJ composites. Strain-to-failure decreased with the addition of CNFs. These results establish the role of CNF coiled shape in controlling the electrical and mechanical properties of nanocomposites. Future studies will assess the role of twisted carbon nanotubes on electrical and mechanical properties. We shall also study the effect of high shear- and extension-rates on the orientation of nano reinforcements.

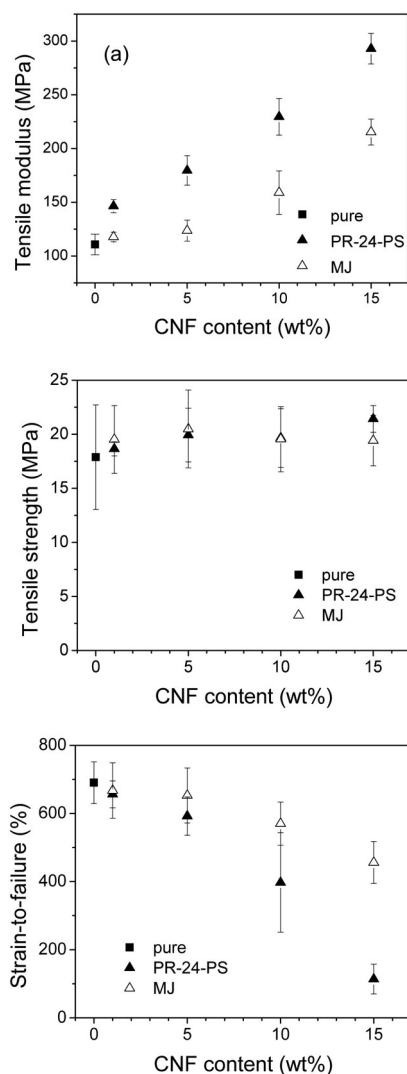


Figure 3. (a) Tensile modulus, (b) yield strength, (c) tensile strength at break, and (d) strain-to-failure of pure LLDPE and its composites. Error bars represent 95% confidence intervals.⁶

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