Plastics Research Online
10.2417/spepro.006969

Adding carbon nanotubes to elastomer blends for improved mechanical performance

Seyed Mohamad Reza Paran, Ghasem Naderi, and Aghil Babakhani

The addition of carbon nanotubes into a natural rubber/ethylene propylene diene monomer blend enhances the cure characteristics and physical and mechanical properties of the resultant nanocomposites.

The effective life of a manufactured part can be significantly extended by enhancing the mechanical properties of its constituent components. For this reason, determining the mechanical behavior of rubber composites has become a crucial component of their development. In recent years, researchers have introduced different types of nanoscale reinforcement into natural rubber and ethylene propylene diene monomer (NR/EPDM) blends with the aim of improving the mechanical properties of these rubber composites[1] and thus extending their area of application.

Of the nanoscale reinforcements that are used to strengthen rubber compounds, carbon nanotubes (CNTs) are a promising new class. CNTs are popular for reinforcing applications because they exhibit a high aspect ratio and have a number of unique properties (e.g., a high modulus and good electrical conductivity) that arise as a result of the rigid chemical structure of the carbon network.[2] There are difficulties associated with mixing CNTs into rubber matrices, however. These issues stem from the tendency of CNTs to form bundles (or aggregates) within the matrix.[3] To achieve enhanced mechanical properties in rubber compounds, CNTs must be well dispersed.[4] Furthermore, it is desirable for the procedure that achieves this level of dispersion to be feasible in view of industrial applications.[5]

With this in mind, we have investigated the effect of CNTs on the cure characteristics of elastomer nanocomposite samples by varying the CNT content in NR/EPDM/CNT samples. We also studied the effect on tensile parameters (e.g., the tensile strength and modulus) at different CNT loadings. To prepare the samples, we used a two-roll mixer (with 1mm distance between the rolls) and mixed for 20min. First, the NR was masticated for 4min to achieve a pulp. We then added the CNTs and mixed for a further 4min. When the NR and CNTs were combined, we added the EPDM and other curing agents (i.e., zinc oxide, stearic acid, n-tert-butyl-2-benzothiazyl sulfenamide, 2-mercaptobenzothiazoledisulfide, and sulfur) and continued mixing for a further 12min until a homogeneous medium was achieved. We then prepared dumbbell-shaped specimens for tensile tests using a Zwick hot press (under 160°C for 5min).

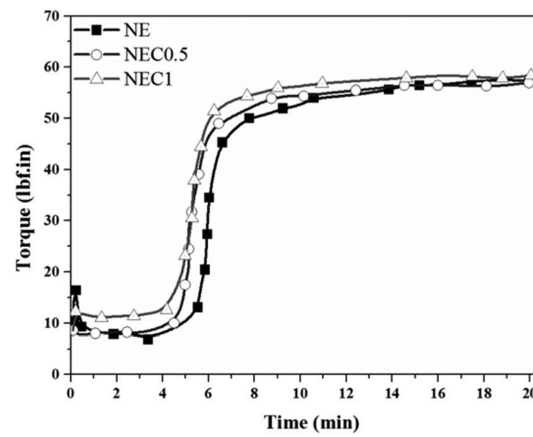

Figure 1. *Cure curves for elastomer-blend nanocomposites with different carbon nanotube (CNT) loadings. NE: Natural rubber/ethylene propylene diene monomer (NR/EPDM). NEC0.5: NR/EPDM with 0.5wt% CNTs. NEC1: NR/EPDM with 1wt% CNTs. lbf.in: Pound-force inch.*

The cure characteristics of the prepared NR/EPDM/CNT nanocomposites are shown in Figure 1. Results for the minimum torque values, which are associated with the viscosity decrease of the rubber matrix,[6] show a slight increase at higher CNT loadings. We attribute this increase to the nanocomposites' higher viscosity, which arises due to the stiffness of the CNTs. This correlation—between the maximum torque value and the CNT concentration of the nanocomposites—may also be related to the effect of CNTs on both the crosslinking behavior and the reinforcement of the rubber compounds.[7] The results indicate that the

Continued on next page

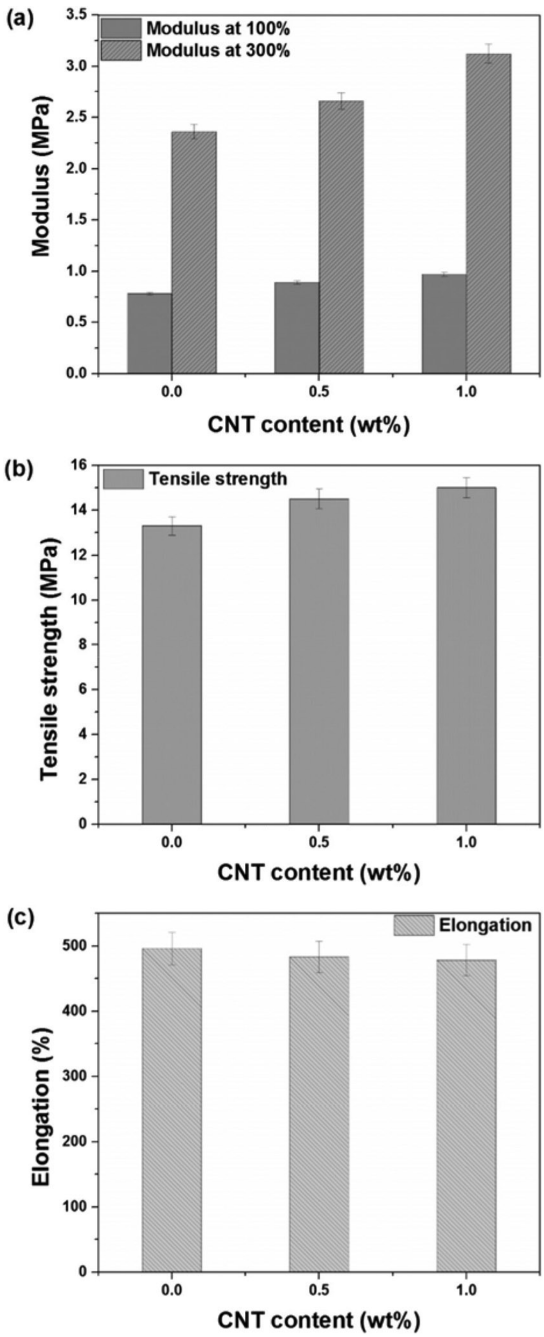

Figure 2. *The effect of CNT loading on the mechanical properties of NR/EPDM/CNT nanocomposites. (a) The modulus at elongation values of 100 and 300%, (b) tensile strength, and (c) elongation at break for nanocomposites with CNT loadings of 0, 0.5, and 1wt%.*

highest maximum torque value and torque difference occurred at CNT concentrations of 1 and 0.5wt%, respectively. Additionally, the minimum scorch and cure times were measured at a CNT concentration of 1wt%. Furthermore, the calculated cure-rate index[8] achieved a maximum value at this concentration. These results suggest that CNTs can act as an accelerator during the curing process, and can increase the crosslink density of NR/EPDM blends.[5]

The results of our investigation into the effect CNT loading has on the mechanical properties of the nanocomposites are shown in Figure 2. We find that the moduli of the nanocomposites when subject to an elongation of 100 and 300%—see Figure 2(a)—increases with CNT content. However, the rate at which the modulus increases at any CNT loading is not constant. Indeed, it decreases in the nanocomposites with 1wt% CNTs. We attribute the increase in the tensile strength at higher CNT content—see Figure 2(b)—to the toughening effect that occurs at higher nanotube concentrations. Finally, our experiments on the elongation at break of the composites—see Figure 2(c)—indicate that the addition of CNTs to the rubber matrix leads to a marginal decrease of the elongation at break. These results suggest that the incorporation of CNTs into the NR/EPDM matrix leads to an enhancement of the mechanical properties of the composite. It has been suggested that this relates to the dispersion state of CNTs,[9] which is highly affected by mechanical interactions between the matrix and nanotubes. Indeed, we believe that the interactions between the CNTs and the NR/EPDM matrix during the vulcanization process give rise to the improvements that we observe in the cure characteristics and the mechanical behavior of the final samples.

In summary, we prepared elastomer (NR/EPDM/CNT) nanocomposites and carried out tensile tests to determine the effect of CNT loading on the mechanical performance of the samples. Our results indicate that higher CNT loadings reduce the cure time and increase torque values in the resultant composites. Furthermore, our tensile test measurements suggest a rise in both the modulus and the tensile strength of NR/EPDM/CNT blends, which we attribute to the reinforcing effect of carbon nanotubes. In the next stage of our research, we plan to investigate the influence of nanotubes on the plastic deformation of a nanocomposite by considering interfacial interactions. In particular, we will evaluate the effect of adding silane-modified halloysite nanotubes (HNTs) on the mechanical properties of polyamide-6 (PA6) by using models that consider the interfacial interactions between the modified HNTs and the PA6 matrix.

Continued on next page

Author Information

Seyed Mohamad Reza Paran and Ghasem Naderi
Department of Polymer Processing
Iran Polymer and Petrochemical Institute
Tehran, Iran

Aghil Babakhani
Department of Polymer Engineering
Islamic Azad University
Tehran, Iran

References

1. I. Kang, Md. A. Khaleque, Y. Yoo, P. J. Yoon, S.-Y. Kim, and K. T. Lim, *Preparation and properties of ethylene propylene diene rubber/multi walled carbon nanotube composites for strain sensitive materials*, **Compos. Part A: Appl. Sci. Manuf. 42**, pp. 623–630, 2011.
2. F. K. Molavi, S. Soltani, G. Naderi, and R. Bagheri, *Effect of multi-walled carbon nanotube on mechanical and rheological properties of silane modified EPDM rubber*, **POJ 3**, pp. 69–77, 2016.
3. P.-C. Ma, N. A. Siddiqui, G. Marom, and J.-K. Kim, *Dispersion and functionalization of carbon nanotubes for polymer-based nanocomposites: a review*, **Compos. Part A: Appl. Sci. Manuf. 41**, pp. 1345–1367, 2010.
4. A. Das, K. W. Stöckelhuber, R. Jurk, M. Saphiannikova, J. Fritzsche, H. Lorenz, M. Klüppel, and G. Heinrich, *Modified and unmodified multiwalled carbon nanotubes in high performance solution-styrene–butadiene and butadiene rubber blends*, **Polymer 49**, pp. 5276–5283, 2008.
5. S. M. R. Paran, G. Naderi, and A. Babakhani, *An experimental study of the effect of CNTs on the mechanical properties of CNTs/NR/EPDM nanocomposite*, **Polym. Compos.**, 2017. doi:10.1002/pc.24467
6. R. N. Mahaling, G. K. Jana, C. K. Das, H. Jeong, and C. S. Ha, *Carboxylated nitrile elastomer/filler nanocomposite: effect of silica nanofiller in thermal, dynamic mechanical behavior, and interfacial adhesion*, **Macromol. Res. 13**, pp. 306–313, 2005.
7. S. M. R. Paran, G. Naderi, and M. H. R. Ghoreishy, *Mechanical properties development of high-ACN nitrile-butadiene rubber/organoclay nanocomposites*, **Plastics Rubber Compos. 45**, pp. 389–397, 2016.
8. W. Arayapranee and G. L. Rempel, *A comparative study of the cure characteristics, processability, mechanical properties, ageing, and morphology of rice husk ash, silica, and carbon black filled 75:25 NR/EPDM blends*, **J. Appl. Polym. Sci. 109**, pp. 932–941, 2008.
9. M. Pramanik, S. K. Srivastava, B. K. Samantaray, and A. K. Bhowmick, *Rubber–clay nanocomposite by solution blending*, **J. Appl. Polym. Sci. 87**, pp. 2216–2220, 2003.