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Ultrasonic twin-screw extrusion for polypropylene/carbon nanotube nanocomposites

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An innovative ultrasonic twin-screw extruder with different screw designs is applied to improve the dispersion efficiency of carbon nanotubes in polypropylene.

Carbon nanotubes (CNTs) are widely used to manufacture polymer nanocomposites with antistatic, electromagnetic shielding, transparent, and conductive functions.[1] These functionalities derive from the intrinsic properties of CNTs, including high aspect ratio (length-to-diameter ratio of about 1000), as well as excellent mechanical,[2] thermal,[3] and electrical[4] characteristics. However, widespread use of CNTs in polymers is limited owing to their inhomogeneous dispersion in the polymer matrix and weak interaction between the CNTs and the polymer.[5]

In recent decades, our group has thus been working on an innovative ultrasonic-aided extrusion technology to improve the dispersion of nanofillers in polymers during melt mixing. Figure 1 shows a schematic of our ultrasonic twin-screw extruder. A detailed description of the device is available elsewhere.[6]

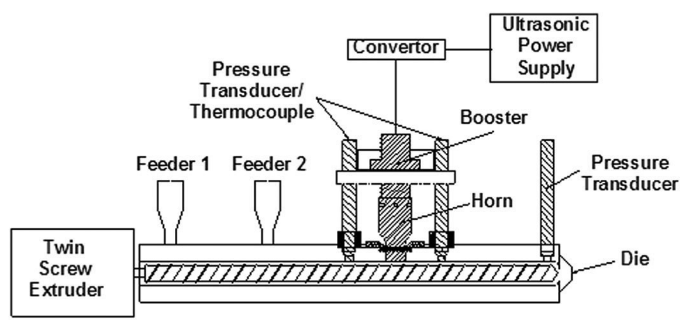

Figure 1. *Schematic diagram of the ultrasonic twin-screw extruder.*

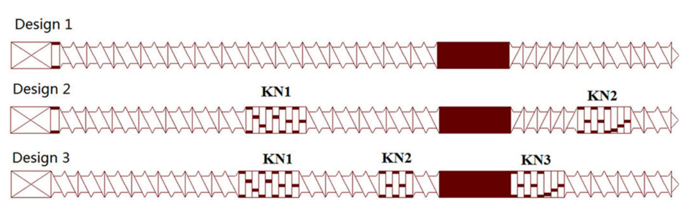

Figure 2. *Schematic illustration showing three different screw configurations. KN1: 5 × 60° RH + 4 × 90° RH. KN2: 5 × 90° RH. KN3: 4 × 90° RH + 4 × 30° LH. RH: Right-handed elements. LH: Left-handed elements.*

In the present study, we prepared polypropylene/carbon nanotube (PP/CNT) composites using our ultrasonic twin-screw extruder with three different screw configurations (see Figure 2).[7] Based on pressure transducer readings, we find that the pressure in the ultrasonic treatment zone is highest in Design 1 and lowest in Design 3. In addition, the residence time is longest in Design 2 and shortest in Design 1. Generally, the polymer degrades during ultrasonic extrusion. However, as seen in Figure 3, for Designs 1 and 2, the degradation only appears at an amplitude of 13μm, and not at lower amplitudes. For screw Design 3, degradation of PP starts to occur at an amplitude of 7.5μm with further degradation at higher amplitudes. The latter is related to the pressure in the ultrasonic treatment zone. In particular, the pressure in the ultrasonic treatment zone of Designs 1 and 2 is higher than that of Design 3. This shows that the higher pressure could suppress the chain scission (i.e., bond cleavage) effect on PP.

Information relating to the dispersion of CNTs in PP can be obtained by measuring dynamic properties at small amplitude oscillatory shear (see Figure 4). The composites prepared using Design 3 show the highest storage modulus (G$'$) and complex viscosity ($|\eta*|$) both with and without ultrasonic treatment, followed by Design 2. Design 1 shows the lowest G$'$ and $|\eta*|$. This suggests that the ranking of the mixing effect is Design 3 > Design 2 > Design 1. As far as the effect of ultrasonic treatment is concerned, improved dispersion is seen in all the designs from the higher value of G$'$ and $|\eta*|$. The ultrasonic treatment effect in Design 1 appears to improve dispersion of CNTs to a greater extent than the other designs.

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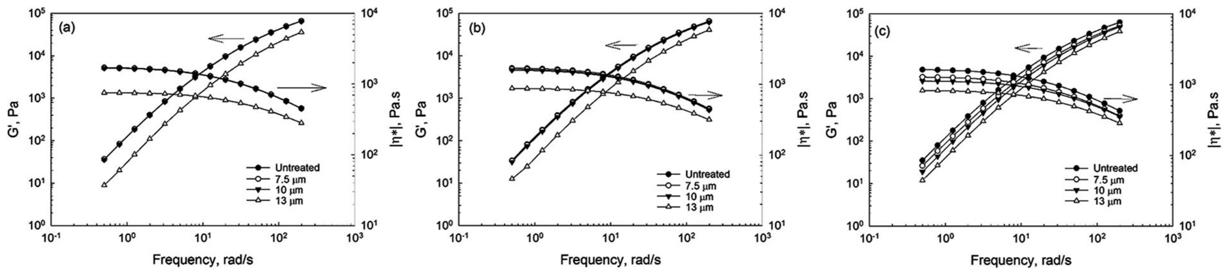

Figure 3. Storage modulus (G′) and complex viscosity (|η ∗ |) of untreated and treated (at amplitudes of 7.5, 10, and 13μm) pure polypropylene (PP) in three screw designs: (a) Design 1, (b) Design 2, and (c) Design 3.

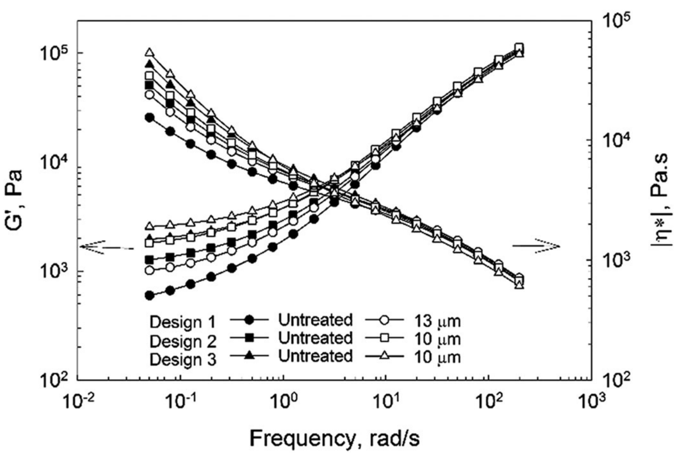

Figure 4. G′ and |η ∗ | for untreated composites and composites prepared using Design 1 at an amplitude of 13μm and with Designs 2 and 3 at an amplitude of 10μm.

Figure 5 shows the stress-strain behavior of PP and PP/CNT composites. For pure PP samples, the yield stress is about the same for all screw designs. The ultrasonic treatment does not significantly affect the yield stress. In Designs 1 and 2, however, the elongation at break increases with ultrasonic treatment at an amplitude of 13μm, while remaining unchanged at an amplitude of 10μm. In Design 3, the elongation at break increases with ultrasonic treatment at amplitudes of 10 and 13μm compared with that of untreated PP. Comparing the elongation at break with different screw designs, the Design 3 and Design 1 composites show the highest and lowest elongation at break, respectively. These results correspond well with the dispersion level implied from the rheological behavior of composites. Our findings indicate that the screw Design 3 is the best design for dispersion. An increase in elongation at break for PP/3wt% CNTs with ultrasonic treatment is seen only with Design 2. Specifically, ultrasonic treatment at an amplitude of 10μm causes an increase in elongation from 247% for the untreated sample to 320% for the treated one.

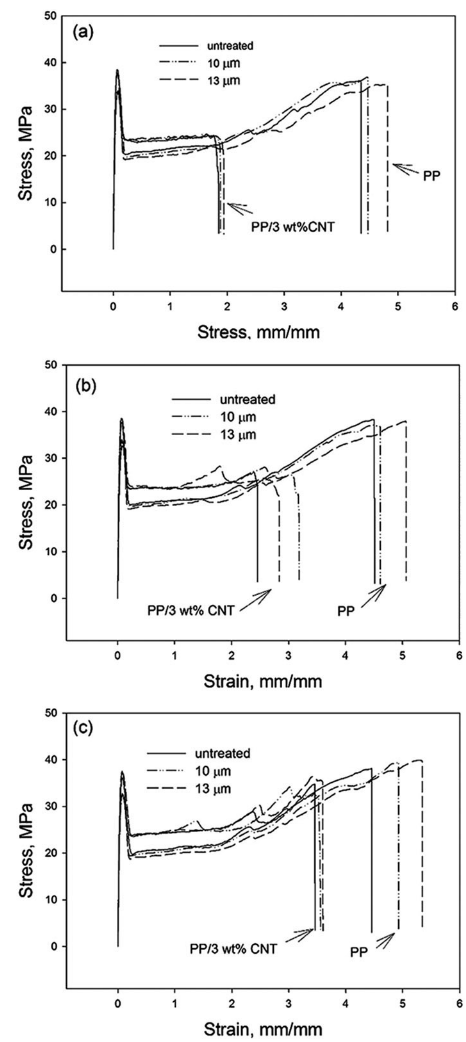

Figure 5. Stress-strain curves of PP and PP/3wt% composites prepared using (a) Design 1, (b) Design 2, and (c) Design 3. CNT: Carbon nanotube.

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In summary, we used an ultrasonic co-rotating twin-screw extruder, with different screw configurations containing kneading blocks that provide different residence times, to compound PP/CNT composites at different ultrasonic amplitudes. A variation of the screw configuration did not induce changes in the rheological and mechanical properties of untreated PP, whereas the ultrasonic treatment induced PP degradation. The degradation was related to the ambient pressure, with a higher pressure leading to less degradation. The screw configuration of Design 3 showed the best dispersion of CNTs, although the residence time was lower. The ultrasonic treatment improved the dispersion in all screw designs, as indicated by a higher storage modulus and complex viscosity. The most significant improvement of the dispersion was observed in Design 1. The mechanism of ultrasonic cavitation in polymer composites is complex, and our future investigations will be directed to further understanding the effect of ultrasonic treatment. The results of the present study open up the possibility for the scale-up of ultrasonic twin-screw extrusion for industrial applications.

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References

1. R. Ramasubramaniam, J. Chen, and H. Liu, *Homogeneous carbon nanotube/polymer composites for electrical applications*, **Appl. Phys. Lett. 83**, p. 2928, 2003.
2. E. W. Wong, P. E. Sheehan, and C. M. Lieber, *Nanobeam mechanics: elasticity, strength, and toughness of nanorods and nanotubes*, **Science 277**, p. 1971, 1997.
3. S. Berber, Y.-K. Kwon, and D. Tománek, *Unusually high thermal conductivity of carbon nanotubes*, **Phys. Rev. Lett. 84**, p. 4613, 2000.
4. J. W. G. Wildoer, L. C. Venema, A. G. Rinzler, R. E. Smalley, and C. Dekker, *Electronic structure of atomically resolved carbon nanotubes*, **Nature 391**, p. 59, 1998.
5. M. Šupová, G. S. Martynková, and K. Barabaszová, *Effect of nanofillers dispersion in polymer matrices: a review*, **Sci. Adv. Mater. 3**, p. 1, 2011.
6. J. Zhong, A. I. Isayev, and K. Huang, *Influence of ultrasonic treatment in PP/CNT composites using masterbatch dilution method*, **Polymer 55**, p. 1745, 2014.
7. J. Zhong and A. I. Isayev, *Ultrasonic treatment of PP/CNT composites during twin-screw extrusion: effect of screw configuration*, **Polym. Compos.**, 2015. doi:10.1002/pc.23867