Molecular simulations of the interfacial characteristics of polymer nanocomposites

Melissa A. PASQUINELLI* and Syamal S. TALLURY
Fiber and Polymer Science/TECS, North Carolina State University
Raleigh, N.C. 27695-8301
*Melissa_Pasquinelli@ncsu.edu

Nanocomposites that have increased surface area at the interface between the reinforcement and the matrix only need a little amount of reinforcement to achieve a significant enhancement in the overall desired property[1]. Thus, carbon nanotubes (CNTs), with their high surface area, are an attractive option for a composite reinforcement material. CNTs have a very high modulus, ranging up to 1 TPa, and thus can greatly enhance the modulus of nanocomposites to several GPa[2]. In addition, some studies have shown that CNTs can bend more than 120 degrees before breaking and that CNTs have versatile electronic properties ranging from nonmetallic to metallic[3]. An important factor of the desired properties of CNT-based nanocomposites is the molecular interactions at the interface between the CNT reinforcement and the polymer matrix.

The goal of the present study is to explore the interface between single-walled carbon nanotubes (SWCNTs) and polymer chains *in vacuo* via molecular dynamics (MD) simulations. These simulations investigate whether the polymers prefer to wrap the SWCNT, what the molecular details of that interface are, and how the interfacial interaction is affected by the chemical composition and structure of the polymer. A series of MD simulations were performed with polymeric matrices of varying degrees of saturation, aromaticity, and aliphaticity, and will thus complement previous MD simulations[4,5] on a few oligomer types. The results of these simulations can be used to correlate the interfacial behavior between a SWCNT and a polymer matrix to the properties of the nanocomposite, and thus lead to the optimization of the desired properties of nanocomposites comprised of these materials.

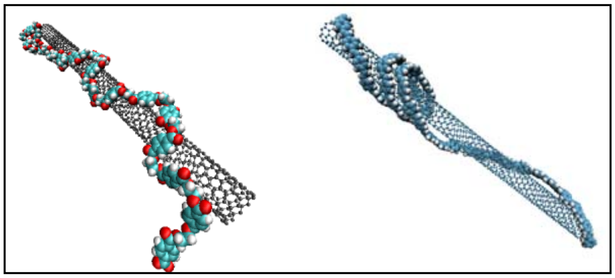

Figure 1: MD snapshots of the interaction between the SWCNT and PET (left) and PPV (right).

MD snapshots for a polymer with a semi-rigid backbone, PET (polyethylene terephthalate), and a rigid backbone, PPV (polyphenylene vinylene), are given in Figure 1. The polymers with a semi-rigid backbone tend to form an S-shaped conformation but never fully wrap completely around the circumference of the SWCNT. The rigid backbone polymers, however, do completely wrap around the SWCNT in a distorted helical conformation. Comparable results were observed for other rigid and semi-rigid polymers in the series. In contrast, flexible backbone chains like nylon-6 wrap the SWCNT in a random configuration, meaning that the polymers wrap completely around the circumference of the SWCNT but no ordered helical structure is observed.

Chemical functional groups such as carbonyls, esters, and amines within the backbone appear to increase the interaction of the polymer chain with the SWCNT. In addition, aromatic groups both within the backbone and on side-chains exhibit much π-π stacking with the SWCNT. For polymers with a flexible background but a bulky side group such as PMMA (polymethylmethacrylate), wrapping was not observed. Other trends and the correlation of these features to the properties of nanocomposites are currently being explored.

References

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