CARBON NANOTUBE AND POLYIMIDE INTERACTIONS: A MOLECULAR DYNAMICS STUDY

Qian Jiang,[1,2] Syamal S. Tallury,[2,3] and Melissa A. Pasquinelli[3]

[1]College of Textiles, Donghua University, Shanghai, China;
[2]Materials Science and Engineering, North Carolina State University;
[3]Fiber and Polymer Science/TECS, North Carolina State University
2401 Research Drive, Campus Box 8301, Raleigh, N.C. 27695-8301
Melissa_Pasquinelli@ncsu.edu

Introduction

The discovery of carbon nanotubes (CNTs) has revolutionized the field of polymer nanocomposites.[1] A combination of aspect ratio, mechanical and electrical properties possessed by CNTs renders them as reinforcement materials for polymer based composites.[2]

Polyimides (PI) are a class of polymers having excellent mechanical strength and high thermal resistance.[3] Polyimide-CNT composites are hence promising multi-functional materials for extreme thermal conditions. An understanding of the molecular level interactions between polyimide and carbon nanotubes is necessary to engineer high performance materials. However, due to the size and timescale of such interactions, it is challenging to observe the molecular phenomenon experimentally. Computational approaches such as molecular dynamics[4] and Monte Carlo[5] techniques have been proven useful for studying these molecular characteristics.

Our work aims to explore the interface between polyimide and different types of carbon nanotubes via molecular dynamics (MD) simulations. We have simulated the CNT pullout under constant rate of loading. The critical force required for complete pullout was determined for the PI/CNT system and the interfacial shear stress was calculated for the different CNT types.

Computational Details

Generation of molecular models. The polyimide used in our studies is BPDA-PDA (3,3',4,4'-biphenyltetracarboxylic dianhydride, p-phenlyenediamine) with a molecular structure as provided in **Figure 1**. The polymer chains used contain 150 repeat units. Each simulation cell consisted of 36 polymer chains and one CNT molecule. The molecular weight of the polymer chains in our simulations is comparable to that of the commercially available product. All the simulations were run with head-tail configuration of the polyimide chain and with the isotactic structure.

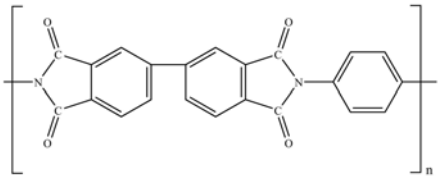

Figure 1. Chemical structure of BPDA-PDA.

All of the molecular structures were generated using Materials Studio package.[6] In this study, we have included three types of carbon nanotubes namely, single-walled, double-walled and triple-walled. The details of the molecular structures studied in our simulations are provided in **Table 1**.

Simulation Details. All simulations were run using the LAMMPS software package.[7] The polyimide/CNT systems were first equilibrated with a 2 ns NPT run under 3 atm pressure and 300 K temperature. The resulting structures were then subjected to pullout simulations for 0.5 ns under NVT ensemble at 300 K. All of the simulations have been performed using the PCFF force field. The simulations had a timestep of 1 fs with 10.0 Å van der Waals cutoff and 8.5 Å Columbic cutoff radii.

Table 1. Properties of the Molecular Structures Utilized In This Study.

CNT Walls	Single (SWNT)	Double (DWNT)	Triple (TWNT)
Type of CNT	(10,10)	(5,5)(10,10)	(5,5)(8,7)(10,10)
Total Number of Atoms	53952	56952	61456
Diameter (Å)	13.6		
Number of PI Repeat Units	150		
CNT Length (Å)	368.9		
Number of PI Chains	36		

Results and Discussion

CNT Pullout Results. We have performed the pull-out simulations of polyimide/CNT systems under different rates of loading. Our results indicate that below a critical laoding rate, the polymer molecules in the system start translating along with the CNT molecule. Figure 2 depicts the interaction energy between the polyimide molecules and the SWCNT molecule as the simulation progresses. It can be observed that below an applied critical force of 20.82 fN, the interaction energy levels off to a fairly constant value. This trend is due to the motion of polymer chains with the SWCNT that maintains a high level of interaction between the two components. Above the critical force of 20.82 fN, we have observed complete pullout occurring with drastic decrease in the interaction energy. This trend was also observed in double-walled and triple-walled CNT systems.

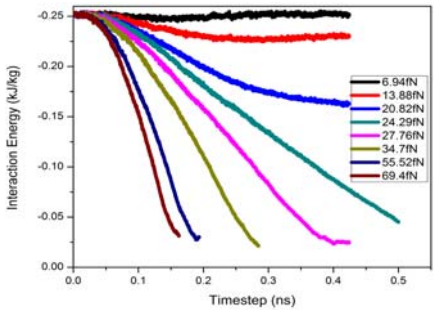

Figure 2. Polyimide/SWCNT interaction energy as a function of simulation time under different loading conditions.

From the simulated systems we were also able to determine the interfacial shear stress (ISS) for the three types of CNTs. We have observed that (not shown) the TWCNT system exhibits a sharp increase in the interfacial shear stress for forces beyond 30 fN. However, it is interesting to observe that the ISS below 30 fN for the TWCNT is well below that of both SWCNT and DWCNT systems.

Conclusions

Our simulations investigate the interfacial characteristics of the polyimide/CNT systems for different types of CNTs. We report the critical loading rate required to obtain a complete pullout for the single, double and triple walled CNTs. The pullout energy was mapped for different applied forces in the simulations. The molecular details and the dynamics of the pullout simulations are extracted from the simulations.

References

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