

Simulation of chemically bonded nanotube-polyethylene composites subjected to mechanical loading

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Abstract: Carbon nanotube polymer nano-composites are promising new materials with a combination of good mechanical, electrical and radiation detection properties. This paper reports the influence of covalent bonds at the carbon nanotube–polyethylene interface under different mechanical loading conditions. Interface covalent bonds were generated by incorporating carbon nanotubes into polyethylene matrix with radical centers on the polymer chains. The resulting nano-composite was then subjected to nano-impact and nano-sliding with the aid of molecular dynamics analysis. It was found that the load carrying capacity of the nano-composite was higher and the bending deformation of carbon nanotube was elastic. In addition, the covalent bonds at the interface were stable during the loading processes.

Keywords: mechanical loading, molecular dynamics, nanotube, polyethylene.

1 Introduction

Carbon nanotubes (CNTs), either single-walled (SWCNTs) or multi-walled (MWCNTs), have been used in making nano-composites due to their superior properties. The reinforcement by carbon nanotubes depends on the ability to transfer load from the matrix to the nanotubes. In order to have high stress transfer, strong bonding between the CNTs and polymer chains is necessary. With the change of polymer matrix and processing conditions, the interfacial bonding properties of CNT-polymer composites can vary significantly. The acceptability of nano-composites in industrial applications is determined by the load carrying capacity, wear life, coefficient of friction, etc.

There have been a number of investigations in the literature on the preparation and testing of various CNT-polymer composites¹⁻³. Tang et al.¹ discussed the suitability and difficulty of various methods and used a melt processing method to make MWCNT/HDPE composite films with various percentages of nanotube content by weight. From a punch test, they found that the stiffness, the yield strength and the fracture toughness all increased with the increasing percentage of CNTs. Xue et al.⁴ studied the tribological behaviour of ultra-high molecular weight polyethylene (UHMPE), high density polyethylene (HDPE) blends reinforced with MWCNTs and showed that CNTs can improve the wear performance of the composite. They found the specific wear rate of the composite decreased with increasing CNTs content. Recently Zhang et al.⁵ and Zarudi et al.⁶ studied CNTs in an epoxy composite subjected to contact sliding stresses using a pin-on-disk machine. Zhang et al.⁵ showed that the surface coverage area of CNTs plays a significant role in the wearability of the composites. They found that the wear rate reduced with high surface coverage area and concluded that this was due to the CNTs exposed to the sliding surface which protected the epoxy matrix. Zarudi et al.⁶ reported a new structural evolution of CNTs in epoxy composites during contact sliding under an applied pressure of 8.36 GPa.

Previous quantum mechanical studies by the authors⁷⁻⁹ have demonstrated that CNT-polymer interface chemical bonds could be formed via functionalizing CNTs, introducing deformations on CNTs, or generating radical centres on the polymer chains. In this work we incorporate CNTs into polyethylene matrix with radical centres and study the effect of chemical bonding on nano particle impact and nano sliding with the aid of molecular dynamics analysis.

2 Simulation methodology

Two CNT-PE models containing one and three (17,0) SWCNTs embedded in PE matrix with covalent bonds at the CNT-PE interface were generated as follows. Some PE chains were placed around an SWCNT and radical centres were created on the PE chains by removing some hydrogen atoms along the chains. The energy of the system was minimized (Figure 1(a)) by conjugate gradient method. Then a crystalline PE matrix was placed around this unit and the energy was again minimized by

conjugate gradient method. This produced a model (I) with one SWCNT embedded in PE matrix via covalent bonds. A spherical diamond nano particle was projected about 5 Å above the surface of model (I) as shown in Figure 1(b). Its impact on, and its retraction from the composite were studied at 300 K. As in most simulation studies, the CNT and PE atoms on the sides and two layers of the PE matrix at the bottom are taken as boundary atoms and all other atoms are taken as thermostat atoms to ensure heat conduction.

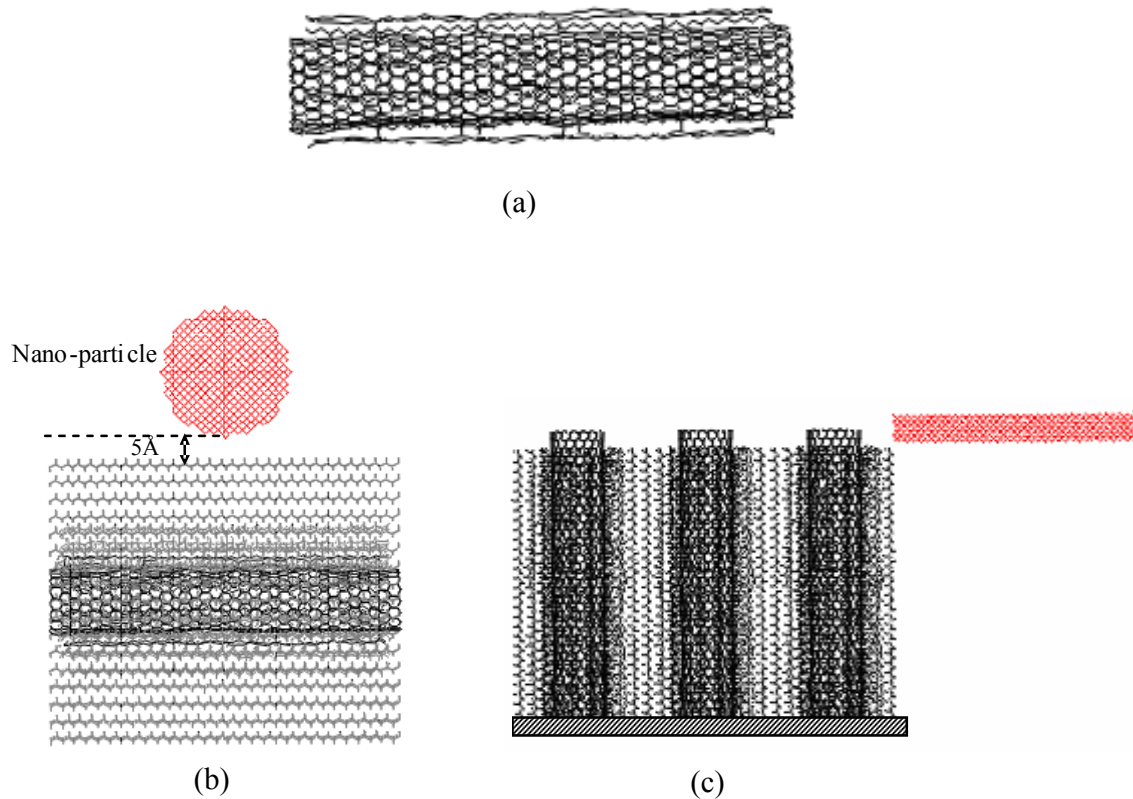


Figure 1: (a) CNT with covalently bonded PE chains (b) side view of the composite nano-particle impact model (c) side view of the composite sliding model.

The above procedure of making model (I) was repeated to produce model (II) with three CNTs embedded in the PE matrix via chemical bonding. A rectangular diamond piece was projected 2 Å above the surface of model (II) as shown in Figure 1(c). Two atomic layers of the nanotubes and the polymer matrix at the bottom were fixed and then relaxed. To simulate the nano-sliding process the diamond piece was moved first vertically by 3 Å and then laterally in steps of 0.001 Å. Nano impact and nano sliding processes were also simulated on pure PE matrices having the same CNT-PE composite sizes, respectively.

The inter atomic interactions were described by a three-body Tersoff-Brenner potential^{10,11}. The non-bonded interactions within the PE matrix and between the polymer matrix and the nanotube were modeled with the Lennard-Jones potential¹².

3 Results and Discussion

The formation of covalent bonds at the CNT-PE interface and their characterization has been demonstrated and explained elsewhere¹³.

3.1 CNT-PE composite subjected to nano-impact

As the nano-particle approached the PE surface, there was initial attraction between the particle and the PE chains. As a result PE chains near the particle moved up and this is reflected in the initial portion of the particle load-displacement curve of Figure 2. The same behaviour was observed in various indentation simulations^{14,15}. As loading continues the force on the nano-particle increased until the CNT fails near the fixed ends. A comparison of the mechanical response of composite with that of the pure PE shows that the composite loading curve took a different path when the CNT started flattening. In addition, it was observed that a higher load is required for the impact and penetration of a particle into the composite showing that the covalent bonded CNT reinforces the polymer.

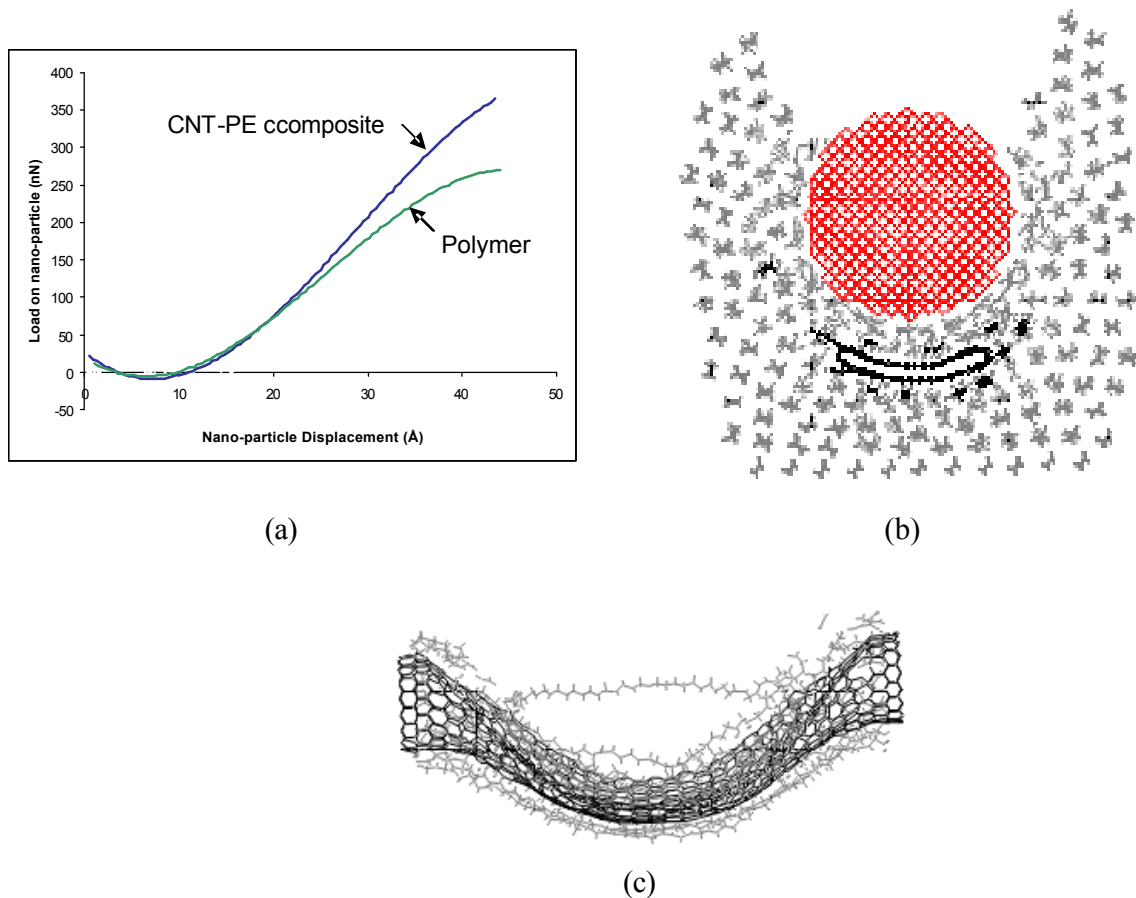


Figure 2: (a) The nano-particle load-displacement curves (b) Cross-sectional view of the CNT-PE composite and (c) CNT and the inner covalently bonded polymer chains on particle penetration by about 45 Å.

As the particle penetrated further into the composite, the PE matrix around the particle got compressed and the CNT first flattened and then bent as shown in Figure 2(b). However, most of the inner polymer chains were stretched and intact with the CNT (Figure 2(c)), showing that the chemical bonds play an important role in reinforcing the composite. On retracting, i.e. on releasing the load from a point slightly before the failure of the CNT, the CNT recovered its cylindrical shape without any structural defect. However, the unloading curve took a different path to the loading curve due to the inelastic deformation of polymer.

3.2 CNT-PE composite subjected to nano-sliding

On sliding, the CNTs bent and buckled near to their fixed ends. The load applied not only caused the nearby CNT to deform but also influenced other CNTs in the polymer matrix to bend and buckle (Figure 3) well before the tool reaches their positions. This demonstrates the load transfer between the CNTs and the polymer matrix. Even though the CNTs deform significantly, the interface covalent bonds between the CNTs and the PE matrix were stable. During the sliding process some polymer chains got destroyed and worn out. The polymer matrix in between the CNTs that were not covalently bonded to the CNT got compressed. This illustrates the protruding effect of CNTs in the composite on sliding as demonstrated by Zhang et al.⁵ in their wear test on CNT-epoxy composite. After the sliding process the CNTs recovered their cylindrical shape and the polymer chains were still covalently bonded to the CNT.

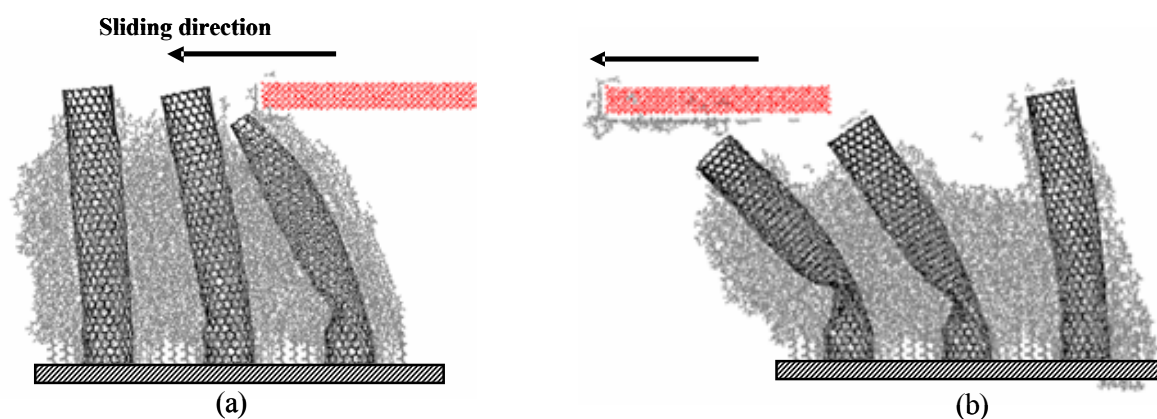


Figure 3: Configuration of a CNT-PE composite (a) on sliding the tool by 39.5 Å and (b) on sliding the tool by 150.0 Å.

In their experiment, Zhang et al. observed that when a short part of a CNT was exposed to the surface it oriented randomly; but when a long part of a CNT was exposed it aligned along the sliding direction. These exposed CNT parts would act as a solid lubricant at the sliding interface and protect the polymer matrix from wear.

4 Conclusion

The study demonstrates that chemical bonds between CNTs and PE can be formed by generating radical centres on the polymer chains. The presence of chemically bonded CNTs in the composite increases the load carrying capacity of the composites subjected to particle impact. The chemical bonds at the CNT-PE interfaces are stable during the deformation processes studied. On sliding the protruding CNTs can act as solid lubricant at the sliding interface and reduce the wear of the composite.

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