This is the author's version of a work that was submitted/accepted for publication in the following source:

Zhan, Haifei, Zhang, Gang, Tan, Vincent B. C., Cheng, Yuan, Bell, John M., Zhang, Yong-Wei, & Gu, Yuantong (2016) Diamond nanothread as a new reinforcement for nanocomposites. Advanced Functional Materials, 26(29), pp. 5279-5283.

This file was downloaded from: https://eprints.qut.edu.au/95798/

© Copyright 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

Notice: Changes introduced as a result of publishing processes such as copy-editing and formatting may not be reflected in this document. For a definitive version of this work, please refer to the published source:

https://doi.org/10.1002/adfm.201600119
Supporting Information

Diamond nanothread as new reinforcement for nanocomposites

Haifei Zhan\textsuperscript{1,2,3}, Gang Zhang\textsuperscript{2,*}, Vincent BC Tan\textsuperscript{3}, Yuan Cheng\textsuperscript{2}, John M. Bell\textsuperscript{1}, Yong-Wei Zhang\textsuperscript{2}, and Yuantong Gu\textsuperscript{1,*}

\textsuperscript{1}School of Chemistry, Physics and Mechanical Engineering, Queensland University of Technology (QUT), Brisbane QLD 4001, Australia
\textsuperscript{2}Institute of High Performance Computing, Agency for Science, Technology and Research, 1 Fusionopolis Way, Singapore 138632
\textsuperscript{3}Department of Mechanical Engineering, National University of Singapore, 9 Engineering Drive 1, Singapore 117576

S1. The energy change during the pull-out simulation for the PE composite with DNT-4

The total energy change ($\Delta E_{\text{PE}}$) increases with the sliding distance and saturates to a certain value ($\sim 9.5$ eV) when the DNT is fully pulled-out from the PE matrix. As shown in Figure S1, the total energy change for the whole system or the PE matrix show relative large fluctuations after the DNT is fully pulled-out, whereas, the profile of the total vdW energy change $\Delta E_{\text{vdW}}$ or potential energy change of the DNT $\Delta E_{\text{DNT}}$ appear relatively flat. In fact, the pull-out of the DNT will release a free space inside the polymer, which will induce free surface inside the polymer and thus allows for a free relaxation of the polymer matrix. Such relaxation will induce fluctuation to the potential energy of the PE matrix, and result in a total energy change trajectory with relatively large fluctuations as seen in Figure S1.

![Figure S1](image-url)

**Figure S1** The potential energy change of the whole PE composite ($\Delta E_{\text{PE}}$), the polymer matrix ($\Delta E_{\text{poly}}$) and the DNT ($\Delta E_{\text{DNT}}$), and the total vdW energy change ($\Delta E_{\text{vdW}}$), as a function of the sliding distance. The potential energy change for the DNT while it is taken as a rigid body is also presented for comparisons ($\Delta E_{\text{DNT}}(R)$).
S2. Estimation of the interfacial shear strength

Simplifying the PE matrix as a continuum media and the DNT as a solid cylinder with a diameter of $D$, the ISS can be derived as:

$$\tau = \frac{2 \Delta E_{vdW}}{\pi DL^2} \quad \text{(S1)}$$

Here $\Delta E_{vdW}$ is the total potential energy change, which approximates to the total vdW energy change. $D$ is the diameter of the DNT, which is approximated as the distance between exterior surface hydrogens, i.e., $\sim 0.5$ nm, following Roman et al. $L$ is the sliding distance.

For the results presented in Figure 2 (in the paper) for the composite with DNT-4, $\Delta E_{vdW}$ is about 9.5 eV, $L$ is about 5.8 nm. Substitute these to Eq. S1, we obtain an ISS of $\sim 58$ MPa at the PE/DNT-4 interface. Similarly, the ISS is estimated as $\sim 52$ MPa for the interface between (4,0) CNT and the PE matrix. We should note that due to the presence of fluctuations, it is hard to get a precise sliding distance or the total vdW energy change when the DNT/CNT is fully pulled-out. As illustrated in Figure S2, adopting different sliding distance would not induce a big difference between the ISS at the PE/DNT and PE/CNT interface. Overall, comparing with the experimentally measured interfacial shear strength, i.e., $\sim 33$ MPa at the PE/CNT(10,10) interface, our estimations suggest that the DNT has comparable interfacial shear strength with PE matrix comparing with that of CNT.

Figure S2 The estimated ISS as a function of the approximated sliding distance.

S3. Composite with different DNTs, different density and with $–C_2H_5$ functional groups

Figure S3a shows the total vdW energy change of the composite with different DNTs, including DNT-1, DNT-2 and DNT-4. As is seen, the composites with DNT-1 and DNT-2 have a similar vdW energy change ($\sim 10.5$ eV), and the other counterpart with DNT-4 appears to have a lower $\Delta E_{vdW}$ (about 9.5 eV). The lower $\Delta E_{vdW}$ for the sample with DNT-4 is actually due to the factor that the initial PE model with DNT-4 is shorter in the length.
direction, i.e., contains less atoms and therefore possesses smaller total vdw energy. As pointed out in Figure S3a, if we consider a same sliding distance, the vdw energy change is similar in all three models. According to Eq. S1, these results indicate that the structure of the DNT exerts insignificant influence to the interfacial shear strength.

Figure S3 The total vdw energy change of the PE composites with: (a) different embedded DNTs, (b) different initial PE densities, and (c) 5% -C2H5 functional groups.

In comparison, \( \Delta E_{\text{vdw}} \) increases from 10.5 to 12 eV when the PE density increases from 0.92 to 1.00 g/cc, signifying an increase in the interfacial shear strength (Figure S3b). Such observation is understandable, as increasing the PE density will increase the non-covalent interaction sites and thus enhance the interfacial shear strength. Further, the total vdw energy change is found to receive an obvious increase when the -C2H5 functional groups are introduced to the embedded DNT. From Figure S3c, \( \Delta E_{\text{vdw}} \) increases from 10.5 to 12 eV, suggesting that the functional groups can effectively enhance the interfacial shear strength.

References
(1) Liao, K.; Li, S. Appl. Phys. Lett. 2001, 79, 4225.
(2) Xiong, Q. L.; Meguid, S. A. Eur. Polym. J. 2015, 69, 1.
(3) Roman, R. E.; Kwan, K.; Cranford, S. W. Nano Lett. 2015, 15, 1585.
(4) Zheng, Q.; Xia, D.; Xue, Q.; Yan, K.; Gao, X.; Li, Q. Appl. Surf. Sci. 2009, 255, 3534.