

## **Polydopamine-Coated Graphene as Multifunctional Nanofillers in Polyurethane**

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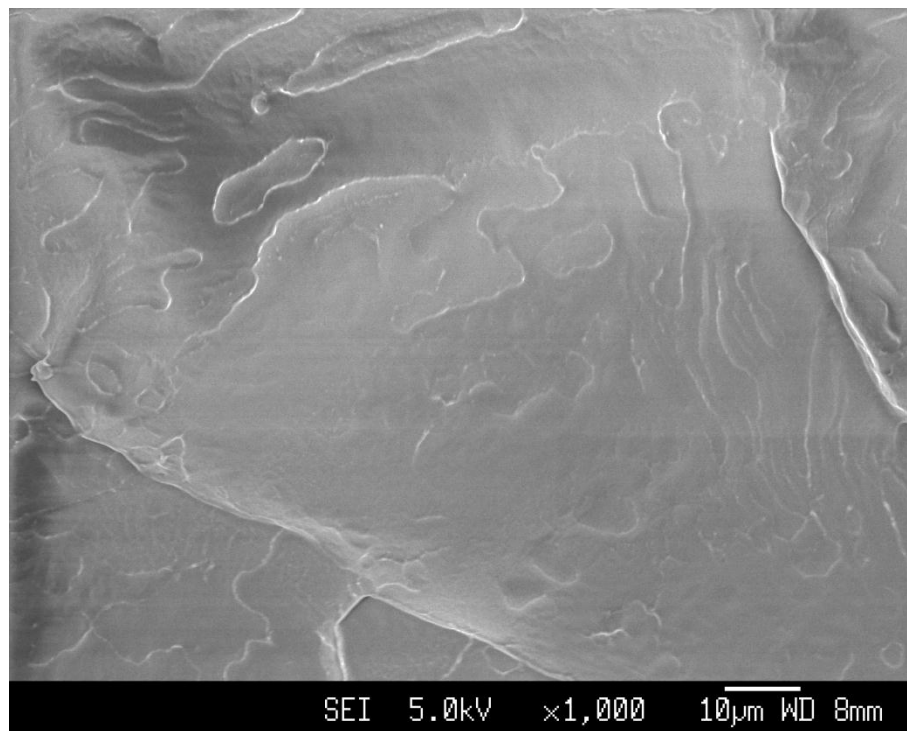
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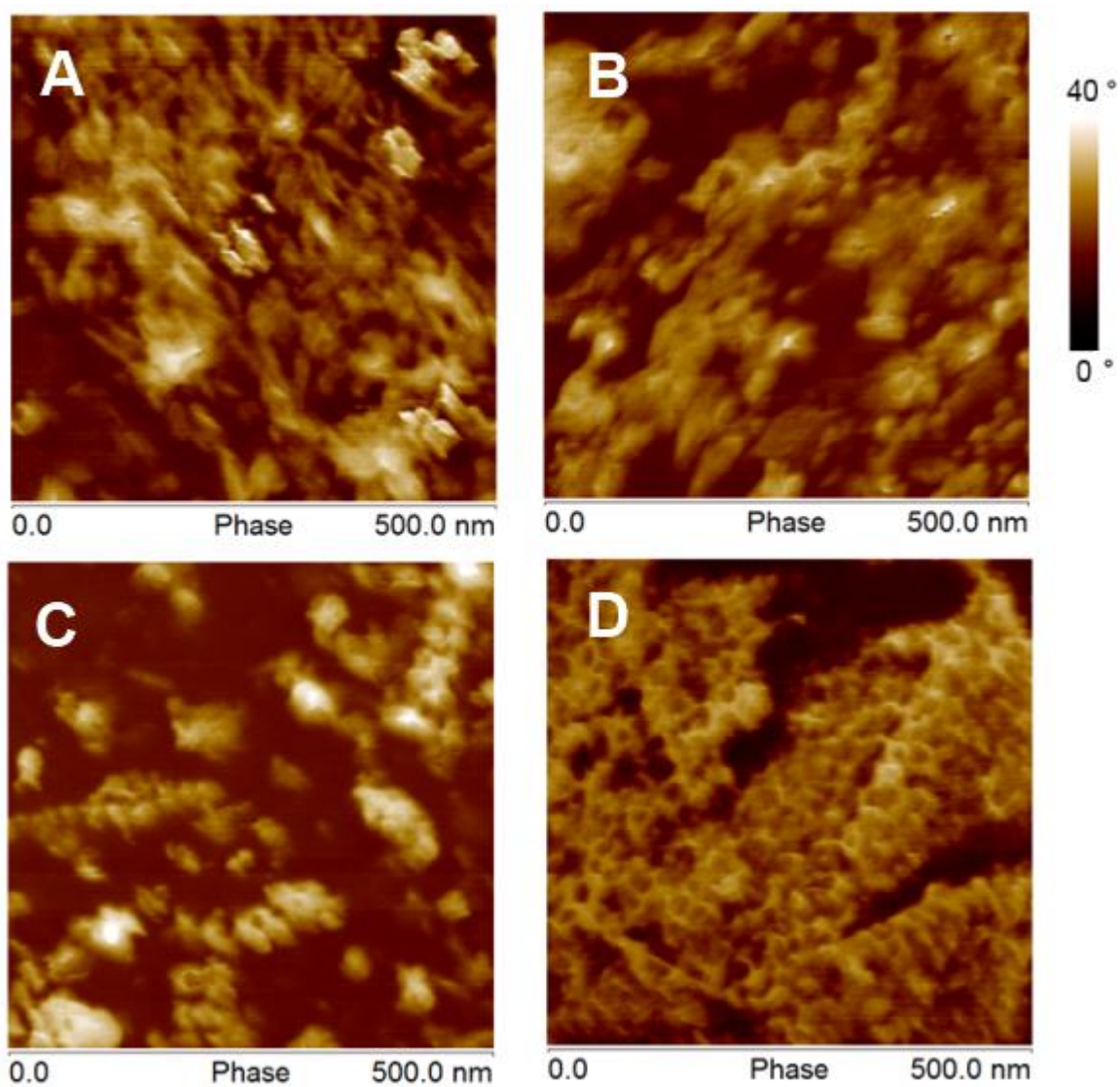
**Keywords:** Graphene, dopamine, polyurethane, nanocomposite, interface



**Figure S1.** Digital photograph of a PU/D-Graphene nanocomposite film sample containing 0.94 vol.% D-Graphene sheets (100mm × 100mm × 2mm).



**Figure S2.** SEM images showing fractured surface of pure PU.



**Figure S3.** Tapping mode AFM phase images of (A) pure PU film, (B) PU/D-Graphene nanocomposite film with 0.24 vol. % D-Graphene and (C) PU/D-Graphene nanocomposite film with 0.47 vol. % D-graphene and (D) PU/D-Graphene nanocomposite film with 2.75 vol. % D-graphene. The hard-segment and soft-segment domains are represented by brighter and darker areas, respectively.

## The Halpin–Tsai model used for predicting the theoretical moduli values of filler-reinforced nanocomposites with unidirectional or randomly distributed fillers

The mechanical reinforcement of filler-reinforced polymer composites depends on the size, shape and orientation of the fillers and the load transfer efficiency between the filler and polymer.<sup>1</sup> To have a better comprehension of the effects of interfacial interactions and filler orientation on elastic moduli of the PU/D-Graphene nanocomposites, the Halpin–Tsai model used for predicting the moduli of filler-reinforced nanocomposites with unidirectional or randomly distributed fillers was adopted to estimate the moduli of the nanocomposites.<sup>1</sup>

For randomly oriented and 2D aligned graphene sheets in the polymer matrix, the nanocomposite modulus  $E_R$  or  $E_A$  are given by equation (1) and (2), respectively,

$$E_R/E_m = 3/8 * (1 + \eta_L \zeta V_G) / (1 - \eta_L V_G) + 5/8 * (1 + 2\eta_T V_G) / (1 - \eta_T V_G) \quad (1)$$

$$E_A/E_m = (1 + \eta_L \zeta V_G) / (1 - \eta_L V_G) \quad (2)$$

in which  $\eta_L$  and  $\eta_T$  are described by the following equations, respectively,

$$\eta_L = (E_G/E_m - 1) / (E_G/E_m + \zeta)$$

$$\eta_T = (E_G/E_m - 1) / (E_G/E_m + 2)$$

where  $E_R$ ,  $E_A$ ,  $E_G$ ,  $E_m$  represent the Young's modulus of the nanocomposite with randomly distributed D-Graphene, the Young's modulus of the nanocomposite with 2D aligned D-Graphene parallel to the surface of the sample film, the Young's modulus of the D-Graphene, and the Young's modulus of polymer matrix, respectively.  $\zeta$  is the aspect ratio of D-Graphene, and  $V_G$  is the volume fraction of graphene in the nanocomposites. The Young's modulus of the chemically reduced graphene sheet was previously measured as around 250 GPa,<sup>2</sup> the Young's modulus of pure PU was 9.10 MPa based on our experimental data. The density of the PU matrix

is  $1.1 \text{ g cm}^3$ , and the density of graphene is  $2.2 \text{ g cm}^3$ .<sup>3</sup> The average size and thickness of D-graphene sheet are about  $1.0 \text{ }\mu\text{m}$  and  $1.25 \text{ nm}$ , respectively, as determined by AFM analysis in ref.<sup>4</sup> Young's modulus of the PU nanocomposites with different graphene dispersion and orientations are calculated by substituting these parameters into the equations.

1. J. J. Liang, Y. Huang, L. Zhang, Y. Wang, Y. F. Ma, T. Y. Guo and Y. S. Chen, *Advanced Functional Materials*, 2009, 19, 2297-2302.
2. C. Gomez-Navarro, M. Burghard and K. Kern, *Nano Letters*, 2008, 8, 2045-2049.
3. H. Kim, A. A. Abdala and C. W. Macosko, *Macromolecules*, 2010, 43, 6515-6530.
4. L. P. Yang, W. A. Yee, S. L. Phua, J. H. Kong, H. Ding, J. W. Cheah and X. H. Lu, *Rsc Advances*, 2012, 2, 2208-2210.