

## Detailed simulation of role of functionalized polymer chains on structural, dynamic and mechanical properties of polymer nanocomposites

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For the rod-like NP filled system, according to the RDF in Figure S1, the adsorption amount of terminal groups increases significantly with  $\varepsilon_{np}$ . In Figure S2 (a) and (b), we probe the fraction of adsorbed terminal-groups and adsorbed chains with the two terminal-groups in each chain being adsorbed at the same time, respectively. It can be seen that the fractions increase with  $\varepsilon_{np}$ , and are the smallest for the spherical case.

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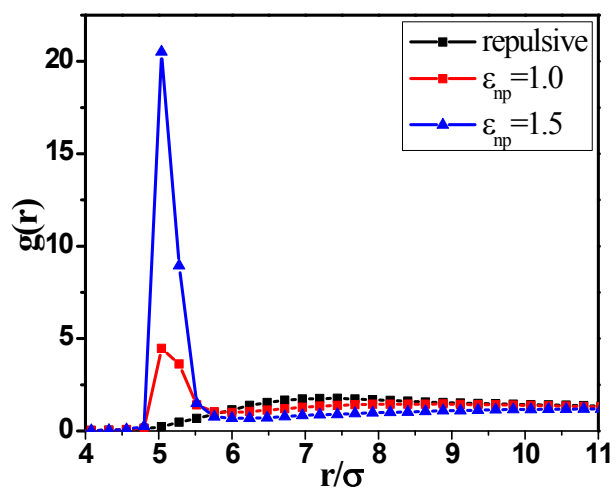
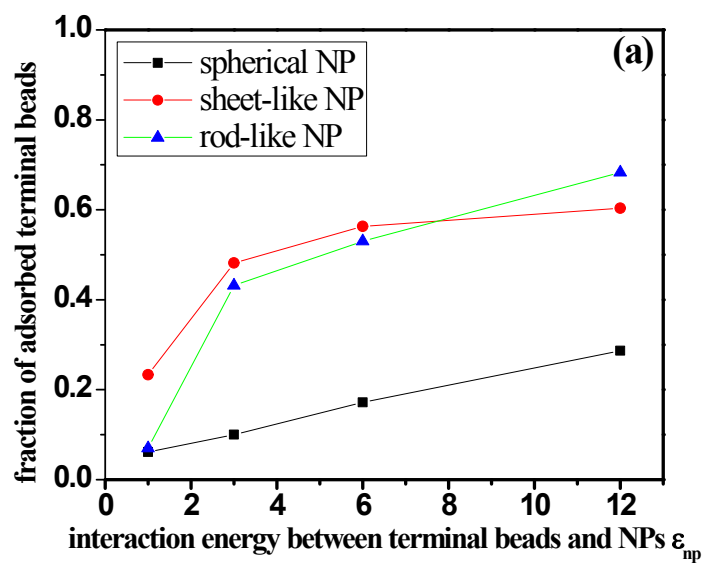


Figure S1. The radial distribution function of terminal-groups around the rod-like NP.



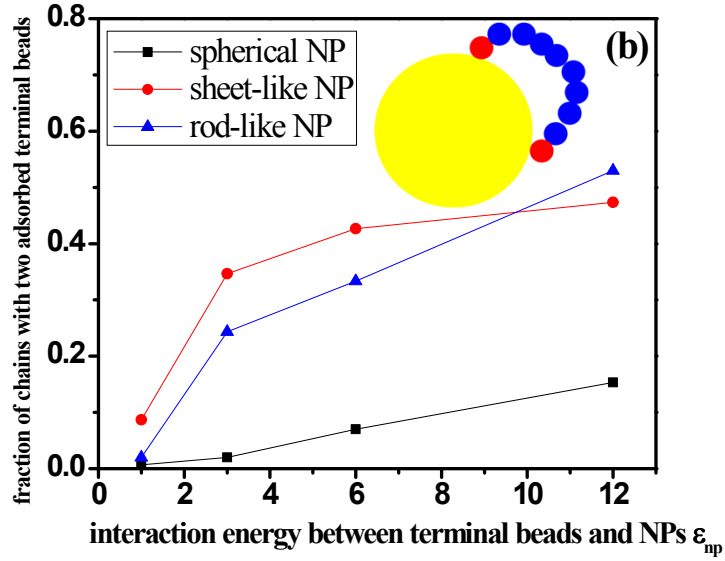
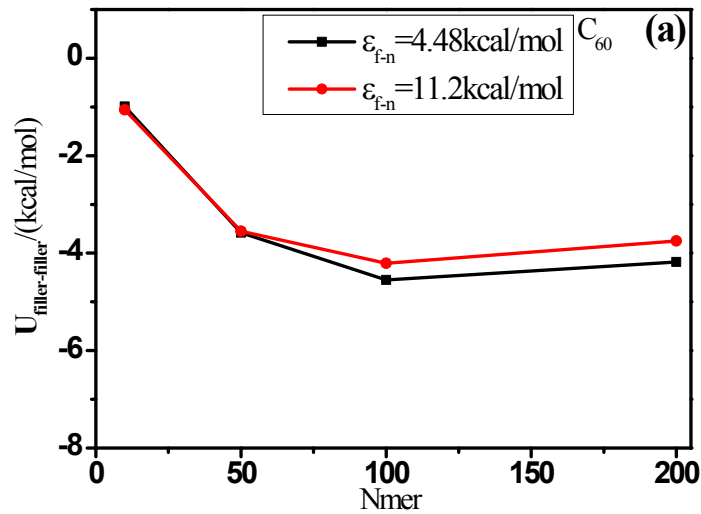


Figure S2. (a) The fraction of adsorbed terminal beads and (b) the fraction of chains with two terminal beads both adsorbed on the surface of NPs as a function of the interaction energy between terminal beads and NPs with different geometries.

We set the length of polymer chains equal to 10, 50 and 100, respectively, while keeping the total number of polymer atoms fixed. Obviously, with the decrease of the chain length, the number of end-functionalized beads increase. We plot the equilibrium value of  $U_{filler-filler}$  with respect to the chain length  $N_{mer}$  with  $\epsilon_{f-n} = 4.48$  and  $11.2kcal/mol$  for  $C_{60}$ , CNT and graphene in Figure S3. With the decrease of the chain length, the absolute value of  $U_{filler-filler}$  decreases, indicating the filler becomes more separated. Meanwhile, the influences of the chain flexibility and temperature on the final dispersion state are also presented in Figure S4. Note that here we set the interaction strength between end-functionalized beads and polymer chains to be  $\epsilon_{f-n} = 11.2kcal/mol$ , and each chain contains ten atoms. We change the parameters of the angle potential as shown in eq. (5b) to tune the flexibility of polymer chains with the following four different cases: (1)  $K_{\theta} = 60, \theta_0 = 109.5$ ; (2)  $K_{\theta} = 60, \theta_0 = 180$ ; (3)  $K_{\theta} = 300, \theta_0 = 180$ ; (4)

$K_\theta = 600, \theta_0 = 180$ . Figure S4(a) indicates that the equilibrium value of  $U_{filler-filler}$  is nearly independent of  $K_\theta$  and  $\theta_0$ . In Figure S4(b) the change of the temperature ( $T = 350K, 400K, 500K$  and  $600K$ ) also seems to have negligible effect on the equilibrium value of  $U_{filler-filler}$ . Namely, here our simulated results indicate that the chain flexibility and temperature have no obvious effect on the dispersion of  $C_{60}$ , CNT and graphene for end-functionalized polymer chains.



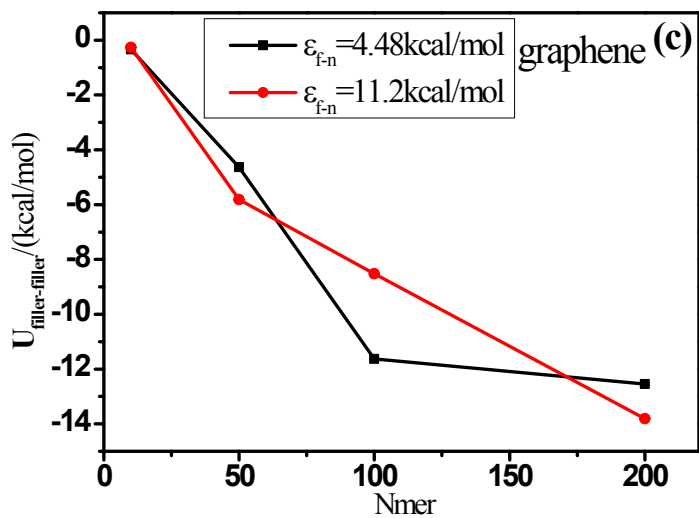
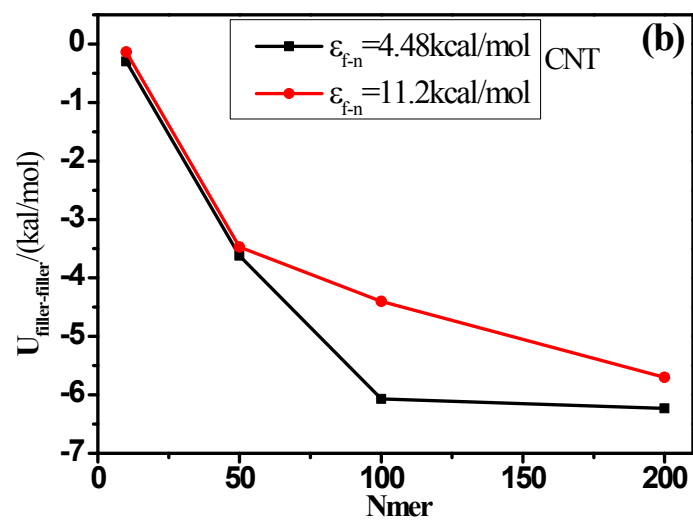


Figure S3. The change of the total filler-filler interaction energy as a function of the length of polymer chains for (a)  $C_{60}$  (b)CNT and (c)graphene in the cases of the interaction strength between functionalized beads and filler.

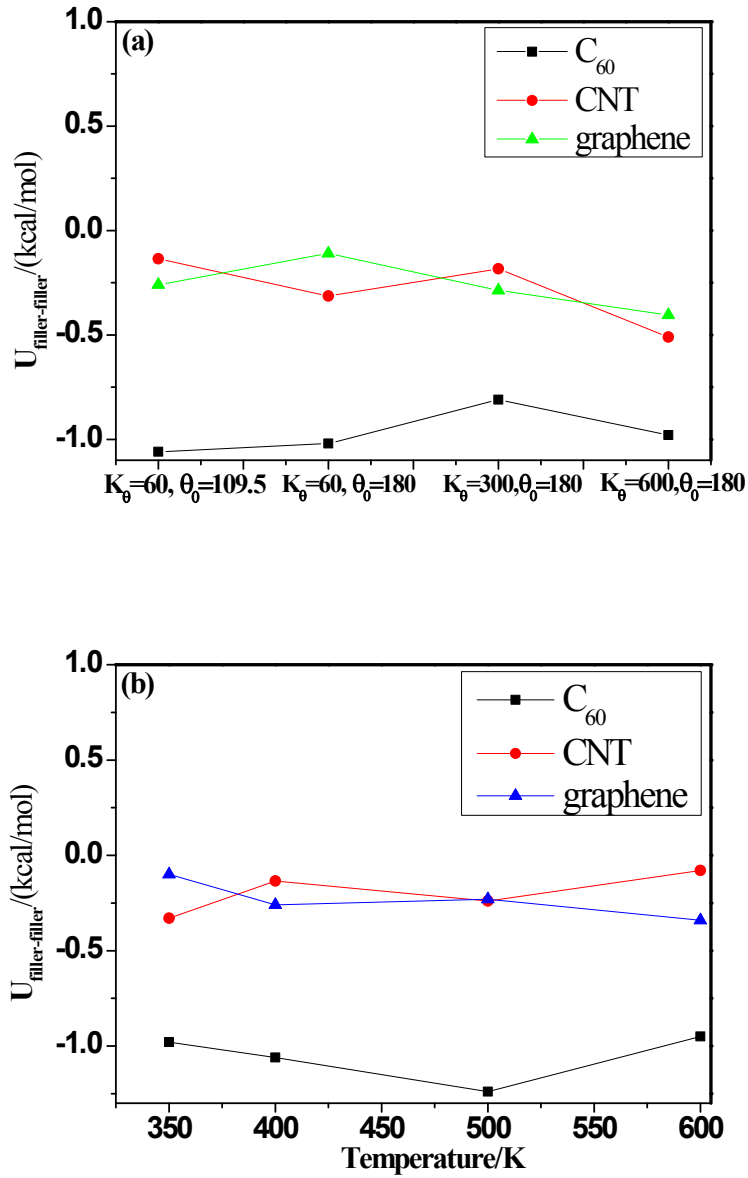


Figure S4. The change of the total filler-filler interaction energy as a function of (a) the effect of the chain stiffness and (b) temperature for  $C_{60}$ , CNT and graphene nanoparticles. Note that  $k$  and  $\theta$  represent the parameters of the angle potential energy of polymer chains. The interaction strength between functionalized beads and polymer chains are set to  $\varepsilon_{f-n} = 11.2 \text{ kcal/mol}$ .