## Fabrication of High-k Epoxy Composites with Low Dielectric Loss Based on Polymer Shell-coated Multiwalled Carbon Nanotubes

Yaotian Su,<sup>a</sup> Ye Ren,<sup>b</sup> Guang-Xin Chen,<sup>a,b,\*</sup> and Qifang Li<sup>b,\*</sup>

a. Key Laboratory of Carbon Fiber and Functional Polymers, Ministry of Education, Beijing University of Chemical Technology, Beijing 100029, P. R. China.

b. College of Material Science and Engineering, Beijing University of Chemical Technology, Beijing 100029, P. R. China.

## Supplementary information



Fig. S1. FT-IR spectra of (a) pristine MWCNTs, (b) poly(DVB)@MWCNTs-1, (c) poly(DVB)

The FT-IR spectra of pristine MWCNTs, poly(DVB)@MWCNTs-1 and poly(DVB) obtained through in situ free-radical polymerization are shown in Fig. S1. It can be clearly seen that pristine MWCNTs don't show any characteristic peaks before coating with poly(DVB). After functionalization, peaks at around 703, 993, and 3018 cm<sup>-1</sup>, corresponding to C-H stretching and hydrogen bending vibrations on the benzene ring of the DVB segment are observed. A peak indicating –CH<sub>2</sub> stretching vibrations on the benzene ring appears at around 2923 cm<sup>-1</sup>. The absorption band ranging from 1466 cm<sup>-1</sup> to 1605 cm<sup>-1</sup> is characterized as the C=C stretching vibrations of benzene. All peaks illustrated above are consistent with spectra curve of poly(DVB), which demonstrates the existence of poly(DVB) shells on MWCNTs surfaces.



Fig. S2. Raman spectra of (a) pristine MWCNTs, (b) poly(DVB)@MWCNTs-1,

The MWCNTs before and after radical functionalization are characterized by Raman spectroscopy (Fig. S2). D and G bands, which are associated with defects/disorder-induced modes and the vibrations of sp<sup>2</sup> bonded carbon atoms in a 2D hexagonal lattice, respectively, are clearly detected at around 1337 and 1568 cm<sup>-1</sup> in pristine MWCNTs samples. Compared with that of pristine MWCNTs ( $I_D/I_G=0.7987$ ), the  $I_D/I_G$  ratios of the poly(DVB)@MWCNTs-1 shown on Fig. S2 is 0.8108 and the two peak positions are all shifted to high wave-numbers. The evidence illustrated above confirms the interactions between poly(DVB) shells and MWCNTs is covalent instead of physical absorption.



Fig. S3. SEM and TEM images of pristine MWCNTs (a and c) and poly(DVB)@MWCNTs-1(b and d)

TEM and SEM images of morphology and microstructure of pristine MWCNTs and poly(DVB)@MWCNTs-1 are shown in Fig. S3. By comparing SEM images of pristine MWCNTs(Fig. S3.a)and poly(DVB)@MWCNTs-1(Fig. S3.b), we can clearly see that the surfaces of poly(DVB)@MWCNTs-1 are much rougher than that of MWCNTs and the average diameter of the tubes increase obviously. As is shown in TEM images, large number of randomly oriented tubes with poly(DVB) coating layers could be observed (Fig. S3.d) and the "core-shell" structure is evident compared with pristine MWCNTs (Fig. S3.c). The evidence illustrated above demonstrates that we successfully prepared poly(DVB)-coated MWCNTs.