

## Supplementary Information

Synergistic effect of Cu<sup>2+</sup>-coordinated carbon nanotube/graphene network on the electrical and mechanical properties of polymer nanocomposites

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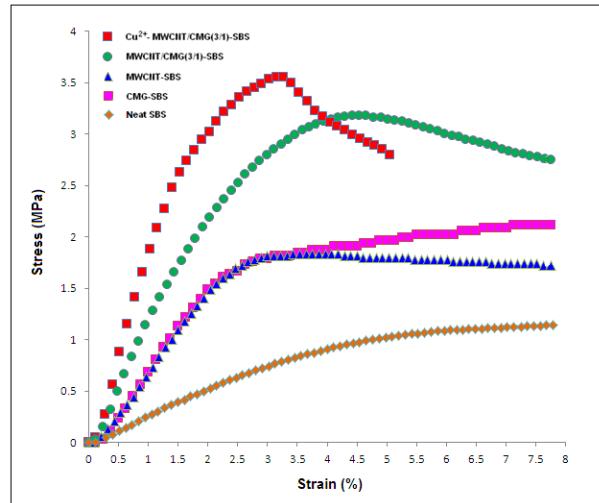
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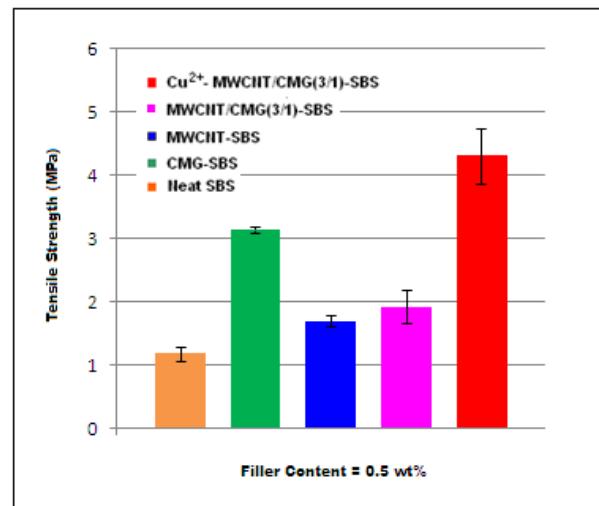
E-mail address: [xxm-dce@mail.tsinghua.edu.cn](mailto:xxm-dce@mail.tsinghua.edu.cn)

## Mechanical property comparison

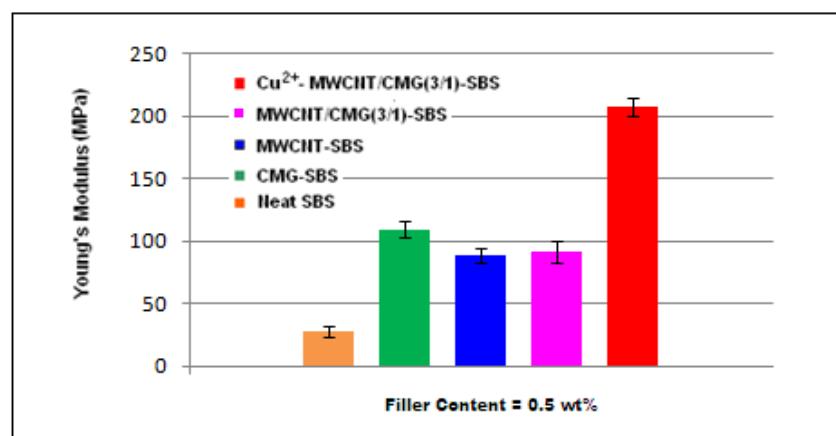
The details of the mechanical properties of neat SBS and the SBS composites filled with CMG, MWCNTs, MWCNT/GO(3/1) and Cu<sup>2+</sup>-coordinated MWCNT/GO(3/1) (filler content = 0.5 wt%) are provided here, including the representative stress-strain curves ([Fig. S1](#)), a comparison among the tensile strength ([Fig. S2](#)), and a comparison among the Young's moduli ([Fig. S3](#)). When the strain is within 2%, the stress-strain relationship is quasi-linear, corresponding to the so-called elastic deformation zone. Seen from the figures, both tensile strength and Young's modulus of the SBS composites filled with MWCNTs or CMG are significantly higher than those of neat SBS, exhibiting remarkable reinforcing effects of both nanofillers. The strength and modulus increases of MWCNTs and CMG are 43% *vs.* 166% and 216% *vs.* 295%, respectively. The better reinforcing effect of CMG is derived from its more excellent mechanical properties and larger specific surface area than MWCNTs [\[1\]](#). A similar phenomenon was observed in a previous study, which revealed that graphene could outperform both SWCNTs and MWCNTs in terms of mechanical properties in the epoxy composites (filler content = 0.1±0.002 wt %) [\[2\]](#). The strength and modulus increases of MWCNT/GO(3/1) are 62% and 227%, once again proving there is no synergistic effect between the two nanofillers by simple physical blending. However, the strength and modulus increases of Cu<sup>2+</sup>-coordinated MWCNT/GO(3/1) are as high as 262% and 653%. The excellent reinforcing effect of a Cu<sup>2+</sup>-coordinated MWCNT network on a metal-matrix composite has already been observed [\[3\]](#). In the present case, the Cu<sup>2+</sup>-coordinated hybrid nanofiller network can effectively confine the polymer chains from deformation or slippage. Besides, the Cu<sup>2+</sup>-coordinated hybrid nanofiller network itself is rigid and robust due to the presence of chemical bonding. The two factors can thus work together to contribute to significantly enhanced tensile strength and Young's modulus at an extremely low filler content.



**Fig. S1** – Representative stress-strain curves of neat SBS and the SBS composites filled with CMG, MWCNTs, MWCNT/GO(3/1) and Cu<sup>2+</sup>-coordinated MWCNT/GO(3/1) network (filler content = 0.5 wt%).



**Fig. S2** – A comparison among the tensile strength of neat SBS and the SBS composites filled with CMG, MWCNTs, MWCNT/GO(3/1) and Cu<sup>2+</sup>-coordinated MWCNT/GO(3/1) network (filler content = 0.5 wt%).



**Fig. S3** – A comparison among the Young's moduli of neat SBS and the SBS composites filled with CMG, MWCNTs, MWCNT/GO(3/1) and Cu<sup>2+</sup>-coordinated MWCNT/GO(3/1) network (filler content = 0.5 wt%).

## Supplementary references

- [1] T. Kuilla, S. Bhadra, D. Yao, N. H. Kim, S. Bose and J. H. Lee, *Prog. Polym. Sci.* 2010, **35**, 1350.
- [2] M. A. Rafiee, J. Rafiee, Z. Wang, H. Song, Z.-Z. Yu and N. Koratkar, *ACS Nano* 2009, **3**, 3884.
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