Enhanced dielectric property and energy density in poly(vinylidene fluoride-chlorotrifluoroethylene) nanocomposite incorporated with graphene functionalized with hyperbranched polyethylene-graftpoly(trifluoroethyl methacrylate) copolymer

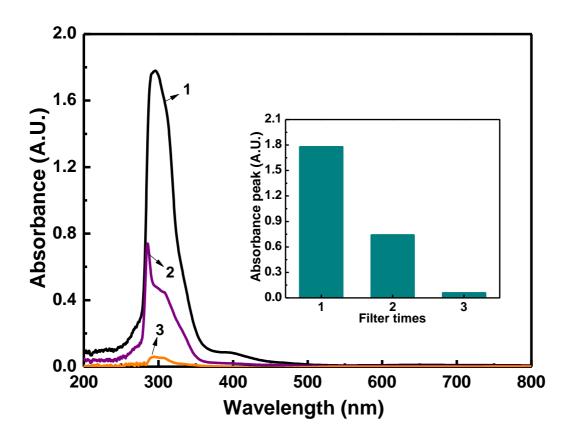
Huijian Ye, Xuanhe Zhang, Chunfeng Xu, Bo Han, Lixin Xu\*

College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China

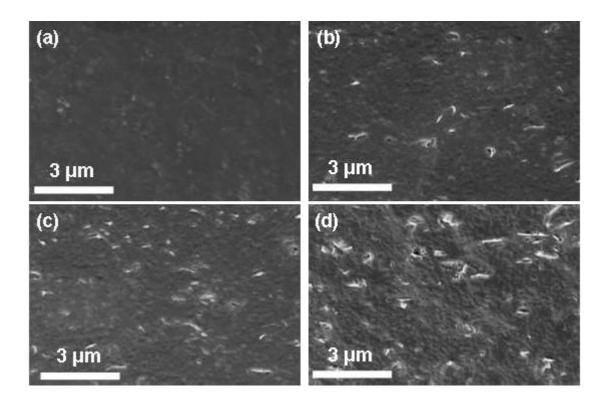
\* To whom correspondence should be addressed. Tel: 86-571-88871519; E-mail: gcsxlx@zjut.edu.cn.

## The polymerization for the hyperbranched polyethylene copolymer

Hyperbranched polyethylene-graft-poly(trifluoroethyl methacrylate) copolymer (HBPE-g-PTFEMA) was prepared by atom transfer radical polymerization (ATRP) of TFEMA with HBPE-Br as macro-initiator, which is analogous with hyperbranched polyethylene preparation [1]. Specifically, 0.6 g of HBPE-Br, 10 g of TFEMA, 0.26 g of bipyridine, and 80 mL of toluene was injected into the 100-mL Schlenk bottle under the protection of N<sub>2</sub>, which was frozen and de-frozen under vacuum for 3 times. Then, 0.12 g of CuBr was injected into mixture and heated to 80 °C under stirring. Subsequently, the polymerization solution was poured into 20 mL of THF and stirred for 1 h to quench the polymerization. The solution was stirred for 2~4 h to dissolve the residuals. After purification by dissolution in THF and precipitation with methanol for three cycles, the polymer product was dried under vacuum at 50~80 °C for 24 h. The THF (AR) and methanol (AR) used herein were purchased from Shanghai Ling Feng Chemical Reagent Co. and Hangzhou Chemical Reagent Co., respectively.



**Fig. S1** UV-Vis curves for graphene filtrate after (1) first filtration, (2) second filtration and (3) third filtration. Inst is the representation of absorbance peak for graphene filtrate. The very low absorbance after washing procedure illustrates that the polymer concentration in the final filtrate has become extremely low, indicating that the free copolymer has been thoroughly removed.



**Fig. S2** SEM images of fracture-surface for (a) pure P(VDF-CTFE), (b) 0.2 vol% nanocomposite, (c) 0.5 vol% nanocomposite, (d) 0.8 vol% nanocomposite

## References

- [1] L. X. Xu, J. W. Mcgraw, F. Gao, M. Grundy, Z. B. Ye, Z. Y. Gu and J. L. Shepherd,
- J. Phys. Chem. C, 2013, 117, 10730–10742.