

**A new approach to construct segregated structure in thermoplastic
polyolefin elastomer towards improved conductive and mechanical
properties**

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Support Information

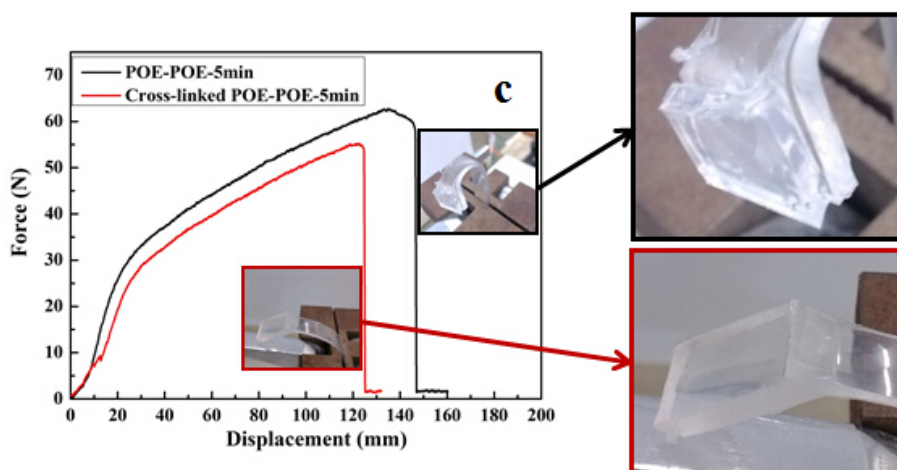
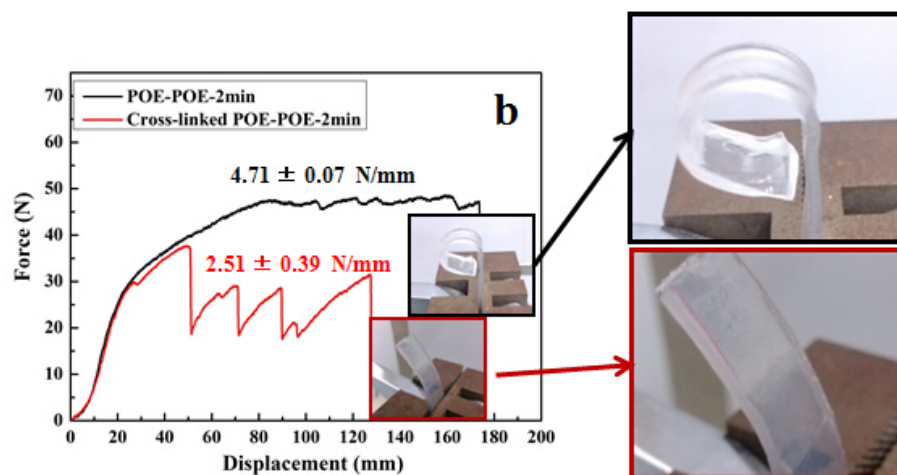
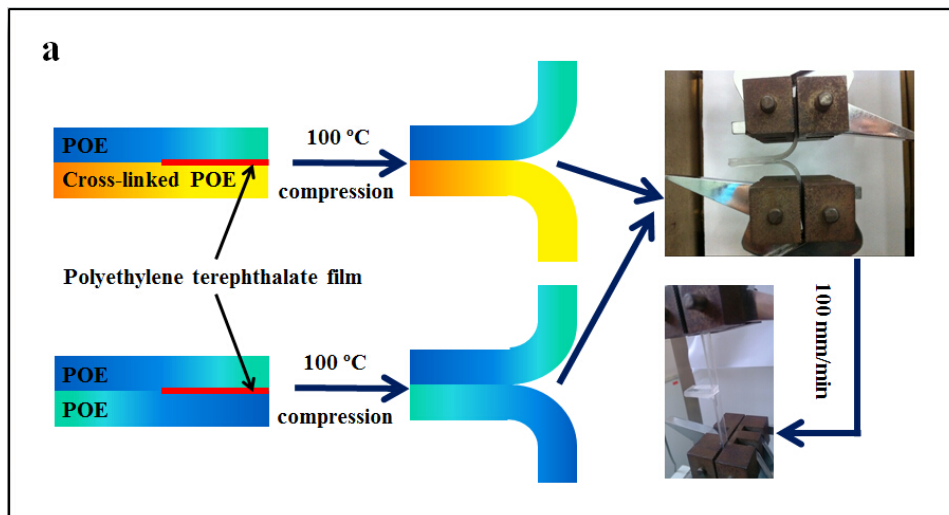


Fig. S1 (a) Schematic for the peeling tests for sample POE-cross-linked POE and POE-POE, (b) force-displacement curves of samples experienced 2 min heat compression at 100 °C and (c) force-displacement curves of samples experienced 5

min heat compression at 100 °C. Digital photographs in (b) and (c) are the final appearance of the samples after peeling test.

To direct prove the interfacial adherence of POE towards cross-linked granules exists, peeling tests were conducted according to a widely adopted peeling procedure as shown in Fig. S1a. For the samples experienced only 2 min heat compression at 100 °C, the peeling strength of sample POE-cross-linked POE and sample POE-POE is 2.51 ± 0.39 N/mm and 4.71 ± 0.07 N/mm, respectively, calculated according to the force-displacement curves presented in Fig. S1b. For the samples experienced 5 min heat compression at 100 °C as can be seen in Fig. S1c, both samples were broken before they were peeled off. Although the interfacial interaction maybe mainly originated from interfacial adherence of POE towards its homogeneous cross-linked granules, instead of interfacial interaction between the two components, the peeling test still clearly indicates that the interfacial adherence of POE molecular chains towards cross-linked granules is excellent, especially when the processing time is longer than 5min at a temperature as low as 100 °C.

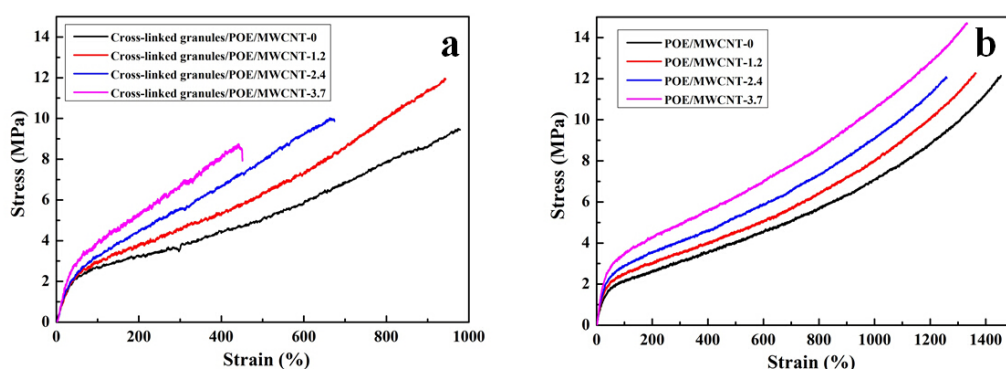


Fig. S2 The representative uniaxial tensile stress-strain curves of (a) cross-linked granules/POE/MWCNT composites, (b) POE/MWCNT composites.

Tensile strength and tensile modulus were discussed in the main text; we mainly discuss the difference of elongation here. Numerous reports revealed that the extensibility of conductive elastomeric composites are decreased when compared with the parent polymer because of the negative influence of rigid conductive fillers¹, so incorporation of MWCNT is one of the reason why the elongation reduced.

After addition of cross-linked granules, while the inherent three-dimensional networks of the granules translate to the impediments of the movement of molecular chains under large deformation, which is in accordance with the investigation of the un-ductile surface of tensile fracture samples in Fig. 6a and previous report², the extensibility decreased obviously and finally affected the whole composite through interface, and this maybe the main reason for the decline of elongation. The interfacial adherence is mainly from POE molecular chains to cross-linked granule as mentioned above, rather than interfacial interaction from each other, so this would be another reason. However, All cross-linked granules/POE/MWCNT elastomeric composites retained high extensibilities greater than **400%**, which is as high as other literature reports on filled elastomeric polymer composites, especially when the loading content of filler is high,^{3,4} resulting in balanced mechanical performance.

Different from cross-linked granules/POE/MWCNT composites, POE/MWCNT composites show a ductile surface as shown in Fig. 6b, indicating the molecular chains of POE/MWCNT slippage severely under large strain, which corresponds to unrestricted movement of molecular chains, and can be considered as the reason why the elongation of POE/MWCNT is better.

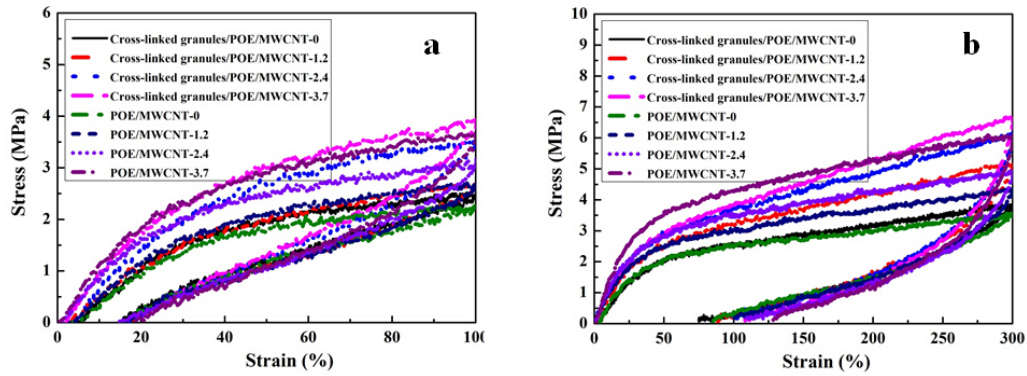


Fig. S3 The representative extension-relaxation curves of samples after (a) Exp100, (b) Exp300.

Reference

- 1 B. X. Yang, K. P. Pramoda, G. Q. Xu and S. H. Goh, *Adv. Funct. Mater.*, 2007, **17**, 2062 -2069.
- 2 K. Sirisinha and D. Meksawat, *J. Appl. Polym. Sci.*, 2004, **93**, 1179-1185.
- 3 M. Z. Seyedin, J. M. Razal, P. C. Innis, R. Jalili and G. G. Wallace, *Adv. Funct. Mater.*, 2015, **25**, 94–104.
- 4 Y. Zhan, M. Lavorgna, G. Buonocore and H. Xia, *J. Mater. Chem.*, 2012, **22**, 10464-10468.