## **Supporting information**

## Tailored Pyroresistive Performance and Flexibility by Introducing a Secondary Thermoplastic Elastomeric Phase into Graphene Nanoplatelet (GNP) Filled Polymer Composites for Self-regulating Heating Devices

Yi Liu,<sup>a,c</sup> Han Zhang,<sup>a,c,\*</sup> Harshit Porwal, <sup>a,c</sup> Wei Tu, <sup>c</sup> Kening Wan, <sup>a</sup> Jamie Evans, <sup>b</sup> Mark Newton, <sup>b</sup> James J.C. Busfield, <sup>a</sup> Ton Peijs, <sup>a,c</sup> and Emiliano Bilotti <sup>a,c,\*</sup>

- a. School of Engineering and Materials Science, Queen Mary University of London, Mile End Road, London E1 4NS, UK
- LMK Thermosafe Ltd., 9-10 Moonhall Business Park, Helions Bumpstead Rd, Haverhill, Suffolk CB9 7AA, UK
- c. Nanoforce Technology Ltd., Joseph Priestley Building, Queen Mary University of London, Mile End Road, E1 4NS London, UK



Figure S1. Neat HDPE, SEBS, EPR and TPU of a) the complex viscosity as a function of frequency. SEBS displays the highest complex viscosity, and there was no Newtonian behaviour observed in the whole experimental frequency range. EPR shows a Newtonian plateau at lower frequencies and shear thinning behaviour at high frequency. HDPE and TPU has similar complex viscosity at low frequency and not an obvious shear thinning behaviour over all the frequency range. b) Storage modulus – frequency relationship, SEBS shows the highest storage modulus and lowest influence of frequency change. While the other three show similarly trend of modulus change.



Figure S2. a) Illustrates the morphology of the fine morphology blend SEBS/HDPE/GNP (50 wt.% SEBS), where the GNP dispersion condition is very similar to the case of the HDPE/GNP composite; b) In the co-continuous EPR/HDPE/GNP blend (50 wt.% EPR), the GNP particles are mostly present in the HDPE phase rather than in the EPR phase; c) For TPU/HDPE/GNP blends (50 wt.% TPU) ,a large proportion of the GNP fillers are located at the interface between the HDPE and TPU phase, while still a smaller proportion of the GNPs in both the HDPE and TPU phase.



Figure S3. a) DSC curve of neat HDPE, indicating the melting temperature of HPDE is around 130 °C. b) The crystallinity degree of TPE/HDPE/GNP composites shows no obvious change at different TPE content.



Figure S4. a) The dependence of storage modulus (E') on the temperature for neat HDPE, SEBS, EPR and TPU. b) Tan delta as a function of temperature, the glass transition temperature (Tg) can be pointed out and correct to the phase change of the polymer matrix. The Tg of HDPE is around -110 °C while the other three sit between -50 °C to -20 °C.

Table S1. Interfacial energy as calculated using Geometric mean equations.

Materials	HDPE/GNP	SEBS/GNP	HDPE/SEBS	EPR/GNP	HDPE/EPR	TPU/GNP	HDPE/TPU
Interfacial energy by Geometric mean equation (mJ/m <sup>2</sup> )	9.56	18.16	19.04	10.58	0.24	10.39	2.22

Table S2 Young's modulus, tensile strength, and strain-at-break of TPE/HDPE/GNP composites.

Composites		Young's Modulus (MPa)	Ultimate Tensile Strength (MPa)	Elongation at Break (%)	
HDPE/GNP	MB	1104.6 (±284.1)	22.3 (±0.9)	5.8 (±0.6)	
	10 wt.% SEBS	1023.3 (±105.8)	18.5 (±0.8)	8.7 (±1.5)	
SEBS/HDPE/GNP	20 wt.% SEBS	1002.7 (±346.0)	18.6 (±0.7)	8.7 (±1.5)	
	35 wt.% SEBS	406.2 (±18.4)	9.9 (±0.1)	283.7 (±22.9)	
	50 wt.% SEBS	235.6 (±27.4)	9.7 (±0.5)	309.6 (±45.6)	
	10 wt.% EPR	892.4 (±61.5)	17.7 (±0.9)	7.5 (±1.4)	
	20 wt.% EPR	843.9 (±182.7)	12.9 (±0.4)	7.3 (±0.8)	
EFK/HDFE/GNP	35 wt.% EPR	462.1 (±113.2)	6.5(±0.3)	8.3 (±0.5)	
	50 wt.% EPR	240.2 (±43.9)	3.7 (±0.1)	30.9 (±2.0)	
	10 wt.% TPU	1021.1 (±87.3)	18.7 (±0.9)	8.7 (±0.5)	
TDU/HDDE/CND	20 wt.% TPU	906.8 (±173.3)	17.2(±1.7)	7.7 (±1.6)	
TPU/HDPE/GNP	35 wt.% TPU	547.8 (±14.6)	9.9 (±1.2)	3.2 (±0.4)	
	50 wt.% TPU	332.5 (±39.7)	7.6 (±0.7)	4.5 (±0.2)	