

## Supplementary Information

### Repeatable and highly conductive carbon nanotube/thermoplastic polyurethane composite with enhanced segregated structure for electrically driven heater

Wen-Jin Sun,<sup>a</sup> Chang-Ge Zhou,<sup>a</sup> Li-Chuan Jia,<sup>a</sup> Yue-Yi Wang,<sup>a</sup> Yun-Peng Zhang,<sup>a</sup> Ting Wang,<sup>a</sup> Ding-Xiang Yan,<sup>a,b\*</sup> and Zhong-Ming Li<sup>a\*</sup>

<sup>a</sup>College of Polymer Science and Engineering, State Key Laboratory of Polymer Materials Engineering, Sichuan University, Chengdu 610065, China

<sup>b</sup>School of Aeronautics and Astronautics, Sichuan University, Chengdu 610065, China

\*Corresponding Authors: [yandingxiang@scu.edu.cn](mailto:yandingxiang@scu.edu.cn) (D.-X. Yan) and [zmli@scu.edu.cn](mailto:zmli@scu.edu.cn) (Z.-M. Li)

#### 1. The DSC curves of the TPU granules.

LTPU and HTPU exhibited different melting behavior, which is investigated via differential scanning calorimeter (DSC). Fig. S1 shows typical the DSC heat flow curves for the HTPU (solid line) and LTPU (dotted line).

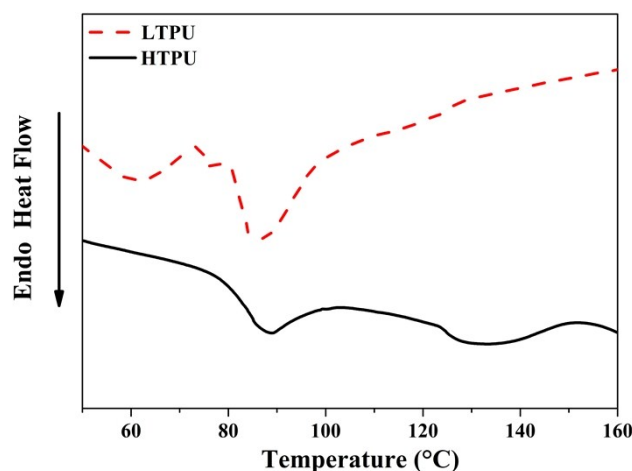


Fig. S1 The DSC curves of the TPU granules.

## 2. SEM micrographs of ESSC and SSC after remolded processing.

The SEM micrographs of ESSC and SSC after remolded processing were supplemented in Fig. S2

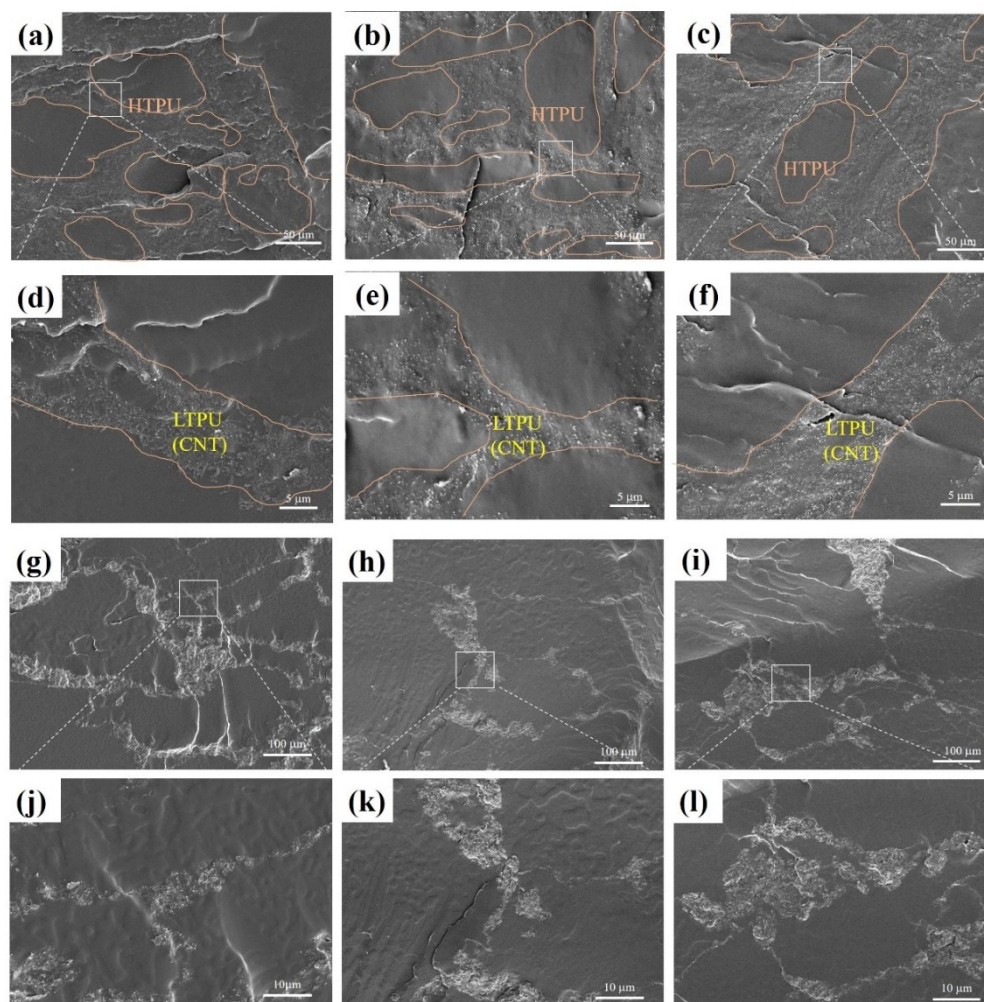


Fig. S2 SEM micrographs of (ESSC) (a) 4C, (b) 6C, (c) 8C, and (d, e and f) are high magnification, respectively; SEM micrographs of (SSC) (g) 4C, (h) 6C, (i) 8C, and (j, k and l) are high magnification, respectively.

## 3. The rheological behavior of HTPU and LTPU at 130 °C

The rheological behavior was measured by a torque rheometer (XSS-300, Shanghai kechuang rubber & plastic equipment co. LTD) (100 rpm/min, 130 °C), as shown in Fig. S3.

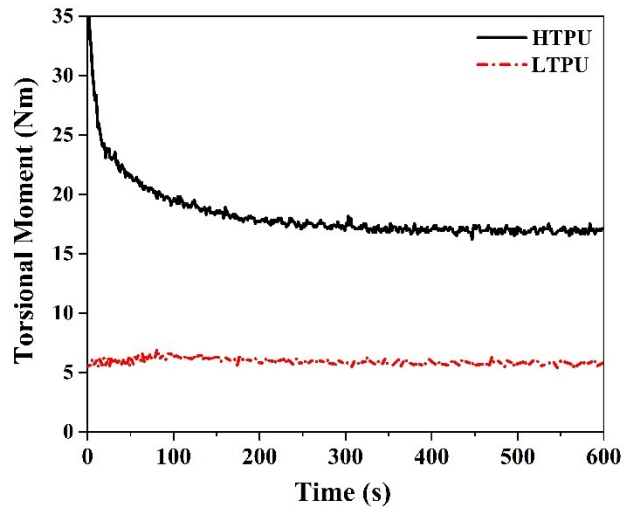


Fig. S3. The rheological behavior of HTPU and LTPU.

#### 4. The schematic of the bending experiment

In order to understand the bending process, we supplement the schematic of the bending experiment, as shown in Fig. S4.

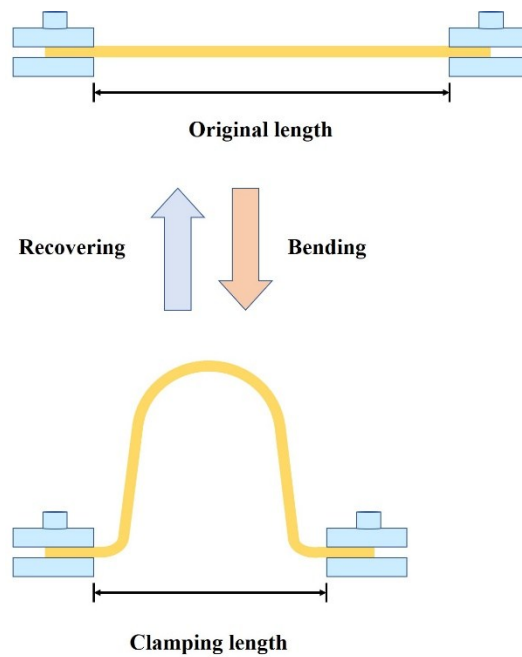


Fig. S4. The schematic of the bending experiment.

#### 5. Time-dependent temperature changes of the CNT/TPU composites under different applied voltages.

In order to represent the electrical heating performance of the CNT/TPU composites, the time-dependent temperature changes of all composites are measured under multiple voltage, as shown in Fig. S5. The temperatures of all CNT/TPU composites with segregated networks raise significantly at an applied voltage below 9 V. As the ratio of LTPU decreases, the voltages that is required for the composites to reach a certain temperature decrease. The CNT/TPU with the ratio of 4/6 obtain 60 °C under applied voltage of 7.5 V. In comparison, the voltage required for the CNT/TPU with the ratio of 1/9 to obtain 60 °C is reduced to 6V. However, all ESSC exhibit better electrical heating properties than CNT/TPU composite with CNT randomly distributed, which obtain 60 °C under applied voltage close to 15 V. The composite that consists of LTPU and HTPU with a ratio of 1/9 not only exhibits excellent conductivity and electrical heating performance close to those of the SSC, but also the repeatability and superior mechanical properties is proved.

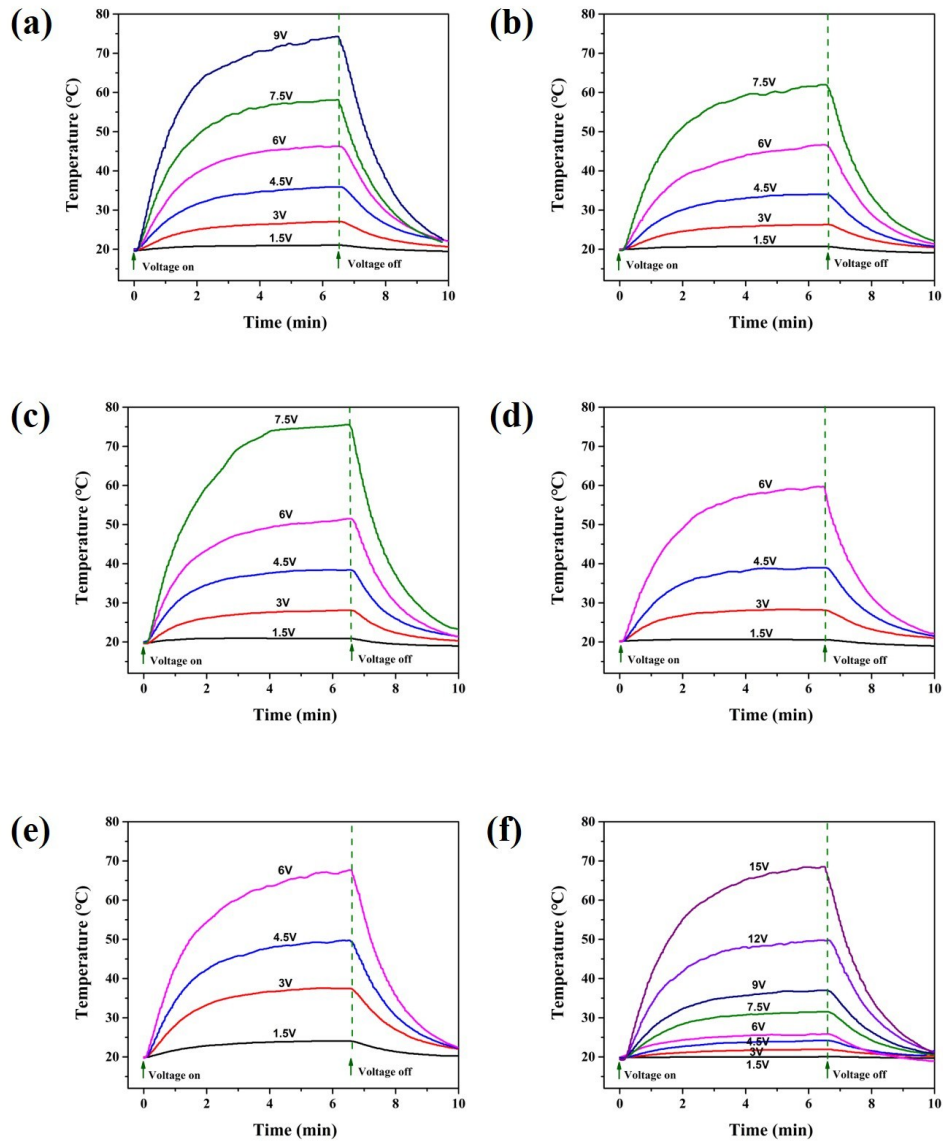


Fig. S5 Time-dependent temperature changes of the CNT/TPU composites under different applied voltages of (a) 4/6, (b) 3/7, (c) 2/8, (d) 1/9, (e) 0/10 and (f) 1/9-R.

### 6. $T_{MAX}$ of the CNT/TPU composites under different voltages and electrical power input per unit volume.

The relationship between  $T_{MAX}$  and applied voltage is described to show the electrical heating of CNT/TPU composites. As we previously reported, the  $T_{MAX}$  is quite dependent on the applied voltage (Fig. S6a). The  $T_{MAX}$  of an electrically driven heater can be calculated by the equation (1).

$$T_{MAX} = \frac{P - Q_d}{Cm} + T_0 = \frac{\frac{V^2}{R} - Q_d}{Cm} + T_0 \quad (1)$$

Where  $T_0$  is the room temperature,  $C$  is the heat capacity,  $m$  is the mass, and  $Q_d$  is the total heat loss. The positively related increment of  $T_{MAX}$  with the applied voltage proves that the electrical power applied to the CNT/TPU composites are effectively converted into thermal energy. In order to theoretically predict the  $T_{MAX}$ , the electrical power input per unit volume ( $P_{in}$ ,  $W\ cm^{-3}$ ) –  $V$  curves are shown in Fig. S6b. Clearly, the  $T_{MAX}$  increases linearly with the  $P_{in}$  of all the CNT/TPU composites. For all CNT/TPU composites,  $T_{MAX}$  can be efficiently acquired by adjusting  $P_{in}$  according to the relation  $T_{MAX} \sim 4.74P_{in}$ .

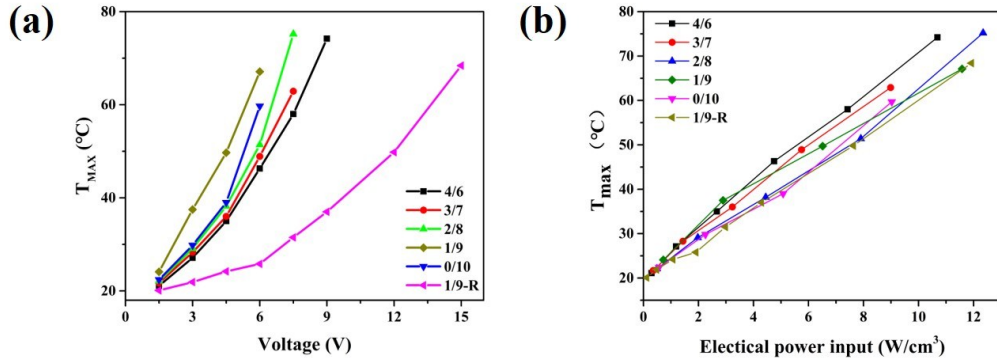


Fig. S6. (a)  $T_{MAX}$  of the CNT/TPU composites under different voltages. (b)  $T_{MAX}$  of the CNT/TPU composites as functions of electrical power input per unit volume ( $P_{in}$ ).