**Electronic Supplementary Information** 

## Smart conducting polymer composites having zero temperature

## coefficient of resistance

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**Fig. S1** Normalized resistance as a function of increasing temperature for CNTs with different aspect ratio. Multiwalled carbon nanotubes (MWNTs) with an outer diameter of 10 nm outer diameter and length of 10  $\mu$ m were purchased from Nanocyl. MWNTs with outer diameter and length of 10–20 nm and 50–200  $\mu$ m (and 10-20nm and 50-100  $\mu$ m), respectively, were purchased from Hanwa Nanotech Inc. The average aspect ratios of the MWCNT samples were characterized by SEM to be ~1000, 2000–6000, and ~10000. Therefore, the degree of NTC of resistance originating from the interconnection resistance between the CNTs could be enhanced by using CNTs with higher aspect ratio.



**Fig. S2** Other configurations of the bi-layered composite with the corresponding circuit diagrams. (a) A layer with PTC of resistance with higher resistance is connected with one Cu electrode, while the other Cu electrode is connected to the NTC layer. (b) Both Cu electrodes are connected to the NTC layers. ( $R_T$ : total resistance of the bi-layer composite,  $R_N$ : resistance of the NTC layer,  $R_P$ : resistance of the PTC layer, and  $P_I$ : contact resistance between the CB/PDMS (PTC) and CNT/PDMS layers (NTC))



**Fig. S3** (a) SEM image of uniformly dispersed CNT/PDMS composite (CNT weight fraction of 12%) with low resolution. (b) Dependence of DC conductivity on the number of three roll passes (CNT weight fraction of 12%). When only a few roll passes were performed, dispersion was poor resulting in low  $\sigma_{DC}$  and a high error bar. On the other hand, a large number of roll passes guarantees uniform dispersion leading to a small error bar (within 2%) and very consistent results, but it slightly reduces the CNTs resulting relatively decreased conductivity. (c) Raman spectroscopy of the CNT composite (12 wt%) with varying three-roll milling passes. (at a wavelength of 514.5 nm to determine changes in the D band (defect and disorder) and the G band (carbon structure)). The inset shows a constant ratio of the intensities of the D-band and G-band peaks (I<sub>D</sub>/I<sub>G</sub>).





(c)

Fig. S4 Numerically calculated normalized resistance of the bi-layer composite with different thickness ratio as a function of temperature. ( $T_P/T_N = 0.3$ , 0.85, and 1.5). At  $T_P/T_N = 0.85$ , the bi-layer composite showed a stable zero TCR, while NTC (PTC) was dominant at  $T_P/T_N = 0.3$  ( $T_P/T_N = 1.5$ ).



**Fig. S5** The change resistance of the bi-layer composite during the electric heating cycling test. In cycling test, 30 second heating and 30 second cooling process (as inset figure) is repeated for 50 hours. During 50 hours, the composite showed nearly constant resistance values with less than 3% deviation of the normalized resistance.



**Fig. S6** Streamline and arrow plots of the current densities of the bi-layer composites with other configurations at a voltage of 11 V. (a) A layer with a PTC of resistance with higher resistance is connected with one Cu electrode, while the other Cu electrode is connected with the NTC layer. (b) Both the Cu electrodes are connected with the NTC layers. In both cases, NTC dominant behaviors were observed at room temperature and 200 °C which are not efficient for zero TCR.

