Supporting Information

Experimental

Materials

CNTs (NC7000), Nanocyl, Belgium; Methyl Methacrylate (MMA), Butyl Acrylate (BA), Azodiisobutyronitrile (AIBN): Chengdu Kelong Chemical Reagent Company, China; Ethylene dimethacrylate(EGDMA), 98% Acros Organics, USA.

Apparatus

The ultrasound equipment employed in this research was a 20 kHz ultrasonic generator (VCF-1500, Sonics & Materials Inc, USA) with a standard titanium horn of diameter 22 mm and a replaceable flat stainless steel tip. The output power of ultrasonic generator was 900 W in the experiment. The ultrasound reaction apparatus is described previously [16].

Preparation of CNTs/P(MMA-BA) electroactive shape-memory composites

The CNTs/P(MMA-BA) electroactive shape-memory composites were prepared according to the following steps. Firstly, MMA (39.6g), BA (20.4 g), EGDMA (0.6 g), AIBN (0.3g) and a certain amount of CNTs were introduced into the reaction vessel. Secondly, the mixture was deoxygenated by bubbling with oxygen-free nitrogen for 2 min in the reaction vessel, and water was circulated to maintain room temperature. Then ultrasonic irradiation was carried out with the probe of the ultrasonic horn immersed directly into the mixture. After a certain time of irradiation, the mixture was injected into a mold composed of two glass plates sealed by silicone rubber (2 mm thick), which were kept at 65 ℃ for 24 h. Then the P(MMA-BA)/CNTs sheets were taken out from the mold and kept at room temperature.

Post thermal treatment

Rectangle sample strips obtained through ultrasound assisted *in-situ* bulk polymerization were thermally treated by heating from room temperature to 180℃ during 30 minutes using an oven while the online electrical resistance of samples was measured by a simple two-point measurement with a picoameter.

Characterization

The electrical conductivity of all samples was measured by a simple two-point measurement with a picoameter (Keithley 2400). Electrodes were painted onto the rectangle sample strip using silver epoxy paste. The measured volume resistance (Rv) was converted to volume resistivity (ρ_v) and conductivity (σ_v) according to the standard (ASTM D4496 and D257) using the formula:

Where *A* is effective area of the measuring electrode and *t* is the thickness of specimen.

Tensile tests were carried out on a universal testing machine (Instron 5567, UK) at 20℃ and a crosshead speed of 50 mm·min^{-1} according to the standard (ASTM D638).

The cryogenically fractured surfaces of the specimens coated with a conductive gold layer, were observed by an Inspect FEI Scanning Electro Microscope (SEM).

The differential scanning calorimetry (DSC) measurements were conducted on a TA Q20 DSC instrument thermal analyzer with a heating rate of 10℃/min and a dynamic nitrogen flow of 50 ml/min.

Dynamical mechanical analysis (DMA) was performed on a TA Q800 DMA instrument at a frequency of 1 Hz from -80 ℃ and 140 ℃. The heating rate was set at 3 ℃/min. The dimension of the rectangle sample strips was 20×10×2 mm.

An infrared video camera (FLIR Thermovision A20) was used to monitor the temperature distribution and shape recovery simultaneously.

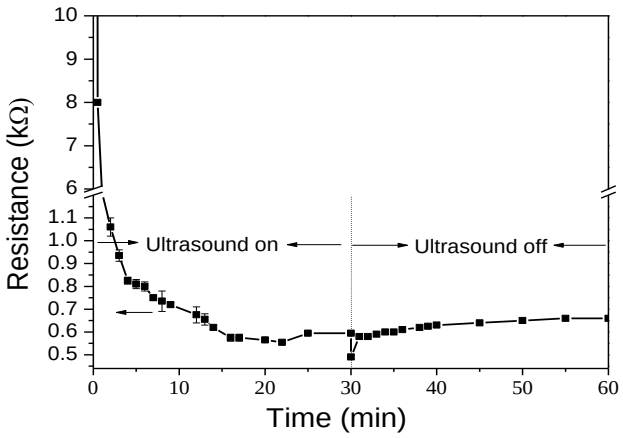

Figure S1. The electrical resistance change of CNTs/MMA/BA/EGDMA liquid mixture during ultrasonic dispersion. (The volume of the liquid mixture dispersion is 25 ml)

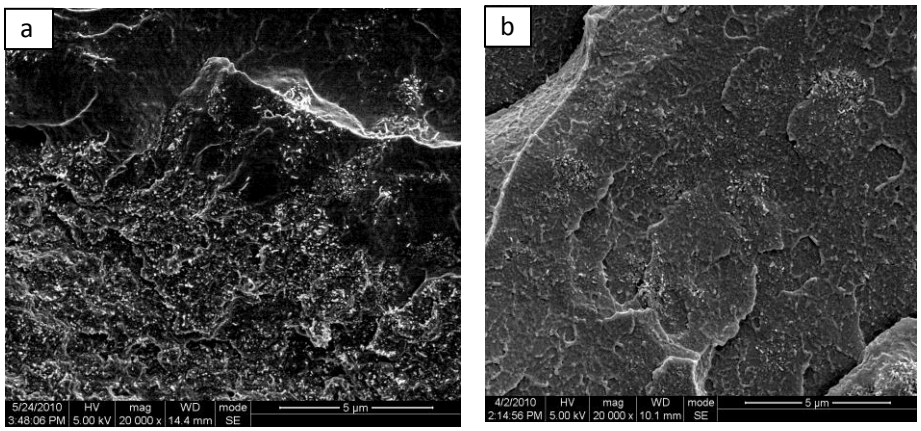

Figure S2. SEM images of cryogenically fractured surface in liquid nitrogen for electro-active shape memory composites. (a) mechanical stirring (b) ultrasonic irradiation (ultrasonic dispersion time: 15 min, CNTs content: 0.5 wt%.)

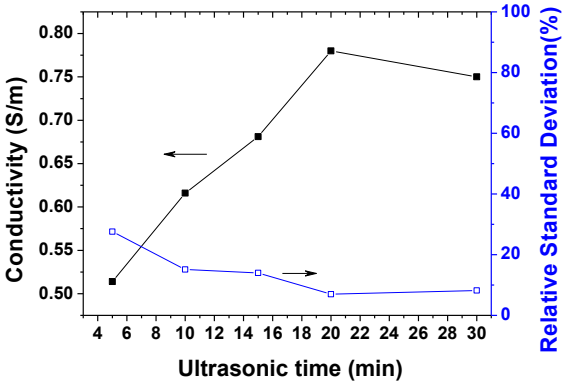

Figure S3. Effect of ultrasonic time on the electrical conductivity of electro-active shape memory composites. (0.5 wt% CNTs)

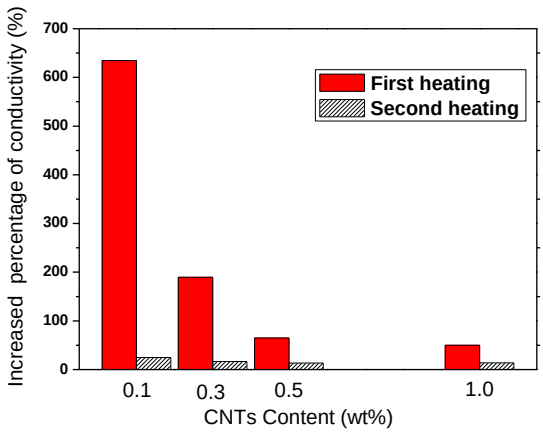

Figure S4. The increased percentages of conductivity of SMP composites when P(MMA-BA)/CNTs composites obtained through ultrasound assisted *in-situ* polymerization were heated up from 25 to 180 ℃.

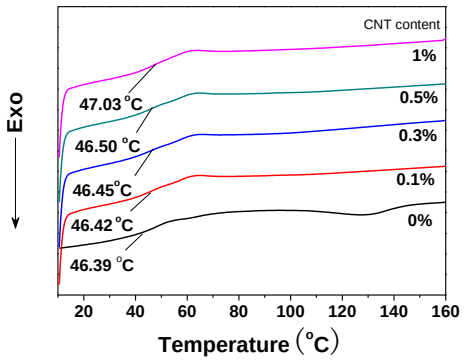

Figure S5 DSC curves of shape memory composites with different CNTs content

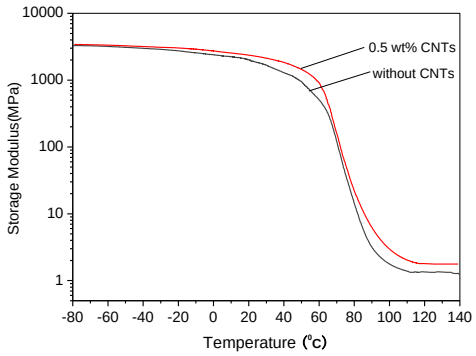

Figure S6. Storage modulus (E') of electro-active shape memory composites as a function of temperature.

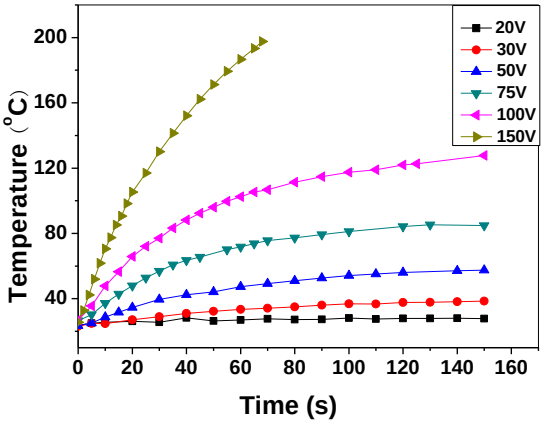

Figure S7. The temperature rise at different voltages with the time for electro-active shape memory composites（Electrical resistance (R): 5.626kΩ, CNT content: 0.5 wt%）

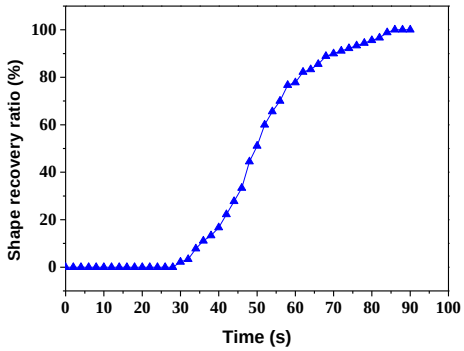

Figure S8. Shape recovery ratio with the time for electro-active shape memory composites with a right angle temporary shape at an electrical power of 2.5 W.

Mechanical behaviors

The influence of CNTs contents on the mechanical properties of electro-active shape memory composites are shown in Table S1. Compared with blank P(MMA-BA) copolymer, CNTs loaded P(MMA-BA)/CNTs composites has a better tensile strength and elongation at break. So, CNTs plays double roles in the electro-active SMP materials. On the one hand, it remarkably enhances the electrical conductivity of the shape-memory composites, and therefore endows the materials electro-active shape memory capability; on the other hand, it also serves as reinforcing filler, leading to the improvement in the mechanical properties of SMP.

Table S1 Effect of CNTs contents on mechanical properties of electro-active shape memory P(MMA-BA)/CNTs composites (ultrasonic dispersion time: 15 min）

CNTs Content (wt%)	Tensile strength (MPa)	Elongation at break (%)	Yield strain (%)
0	37.3	39	5.7
0.1	40.6	39	6.7
0.3	42.0	43	5.3
0.5	39.2	47	5.5
1	39.1	45	6.2

*The testing temperature is 20℃