

Supporting Information

Carbon-nanotubes-filled Catalyst-free Thermoset Polyurea Composites toward Recyclability, Weldability and Permanent Shape Reconfiguration by Multiple stimuli

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1. Synthesis and characterization of small molecule compounds

Synthesis of the hindered amide (HDLA-100): The hindered amide was synthesized *via* the reactions between HD-100 and LA with the molar ratio of 1:3 at 25 °C under nitrogen atmosphere for 4 h and then reacted at 80 °C for 4 h. The reaction product was degassed under vacuum for 30 min. The synthesized HDLA-100 was characterized and used in subsequent syntheses.

Fourier transform infrared spectroscopy (FTIR): The structure of LA, HD-100 and HDLA-100 were characterized using an Infrared Spectrophotometer (Nicolet-560, Nicolet Co.,USA) from 4000-400 cm^{-1} with a resolution setting of 4 cm^{-1} . The KBr pressed disc technique (about 1 mg KBr of sample and 100 mg of KBr) was used to test the liquid samples.

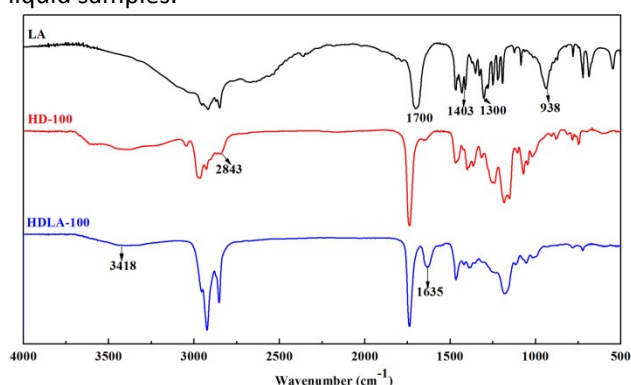


Fig. S1 FTIR spectra of LA, HD-100 and HDLA-100.

Nuclear magnetic resonance (NMR): Hydrogen nuclear magnetic resonance (^1H NMR) spectra of HDLA-100 were recorded on a Bruker Avance II-600 MHz spectrometer by utilizing CDCl_3 as the solvent and tetramethylsilane (TMS) as the internal standard at ambient temperature.

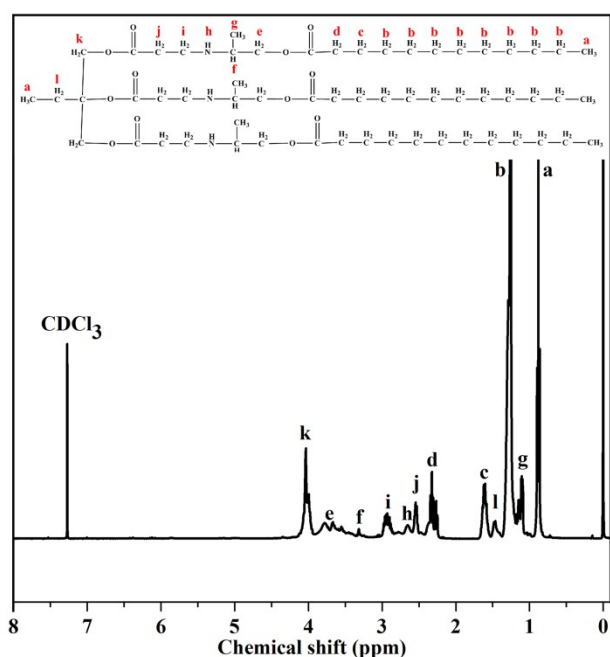


Fig. S2 1H NMR spectra of HDLA-100.

Gel Permeation Chromatography (GPC): The average molecular weight of HDLA-100 was determined by using Agilent HPLC 1200 Infinity Series. The sample of HDLA-100 was dissolved in DMF used as carrier solvent at a rate of 1 mL/min.

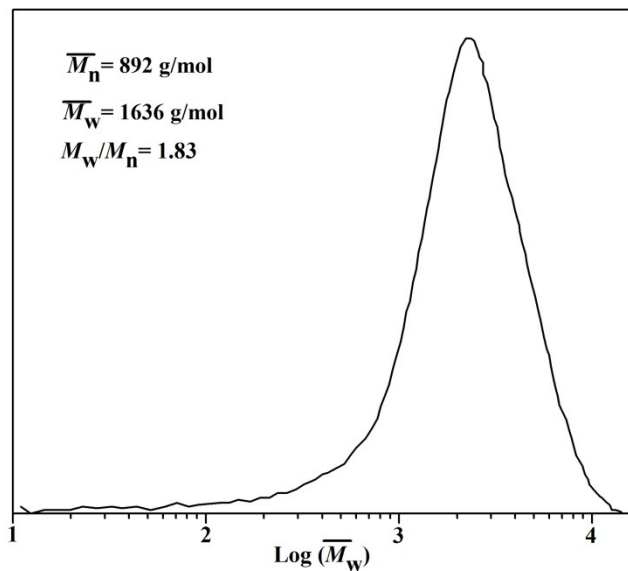


Fig. S2 The average weight of the HDLA-100.

2. Figure and caption

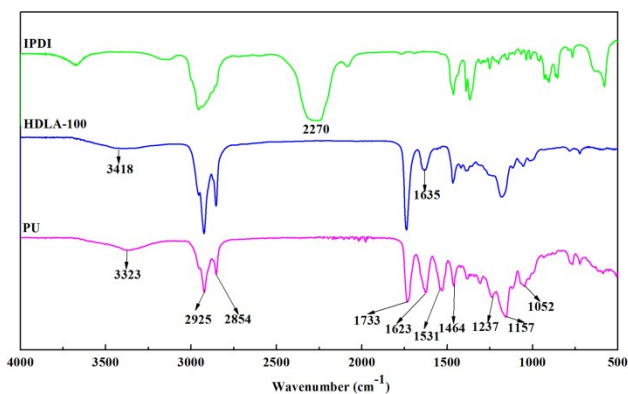


Fig. S3 FTIR spectra of IPDI, HDLA-100 and thermoset polyureas (PU).

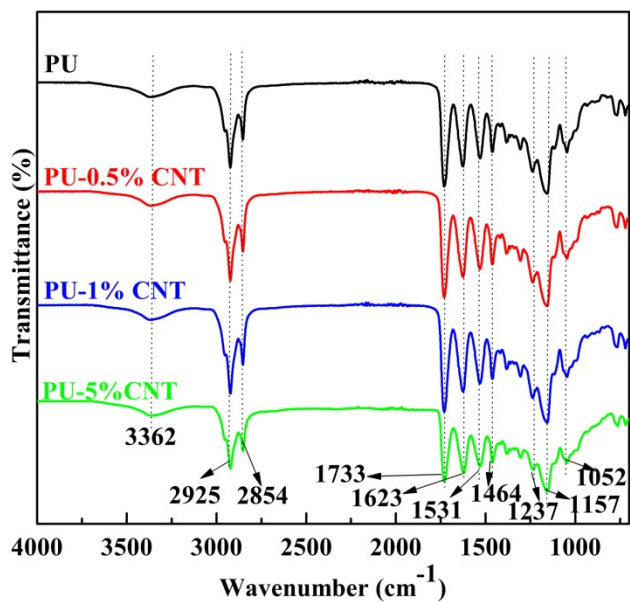


Fig. S5 FTIR spectra of thermoset PU-x % CNT composites.

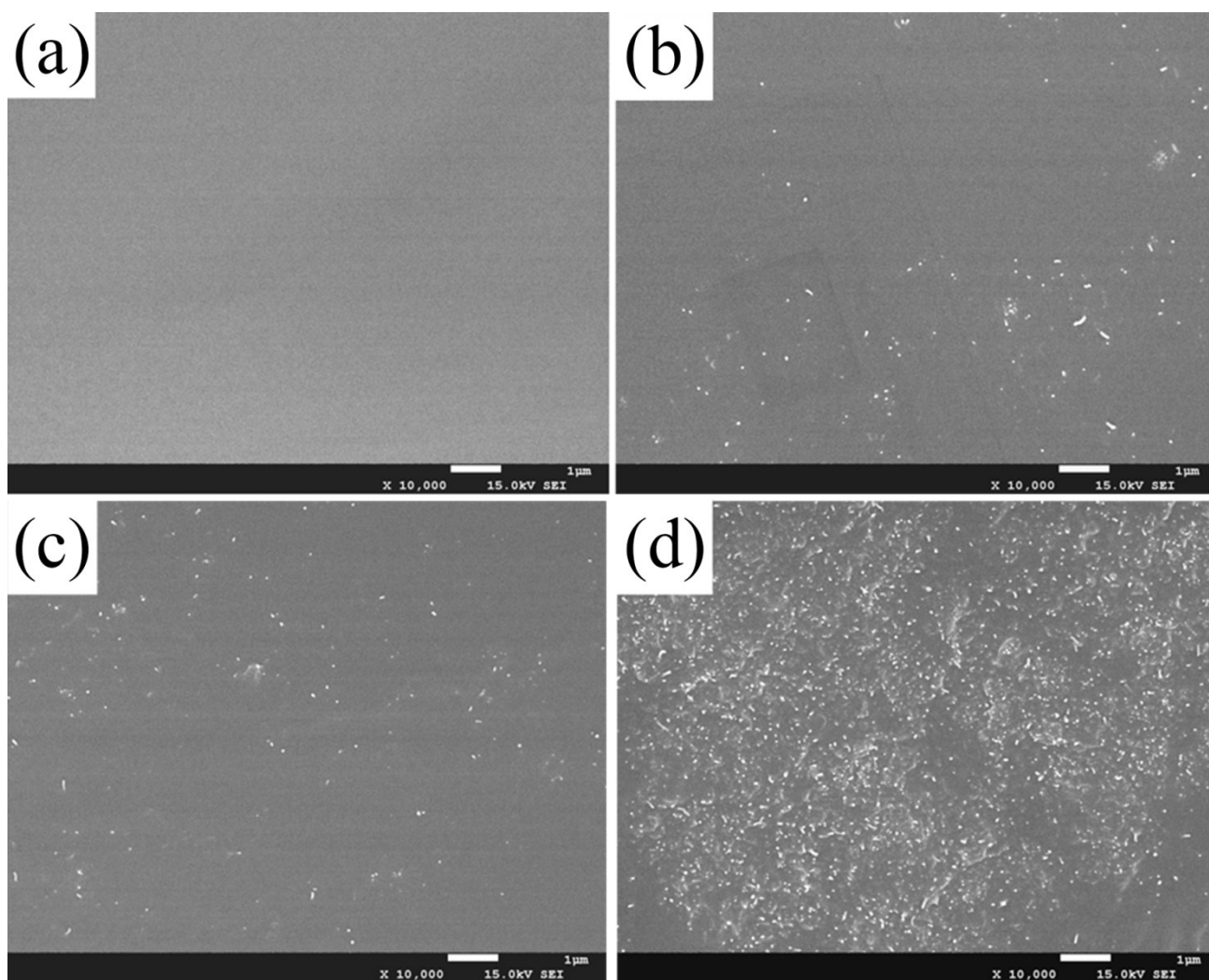


Fig. 6 Low-resolution SEM images of cryofractured surfaces for (a) PU, (b) PU-0.5% CNT, (c) PU-1% CNT, (d) PU-5%

CNT.

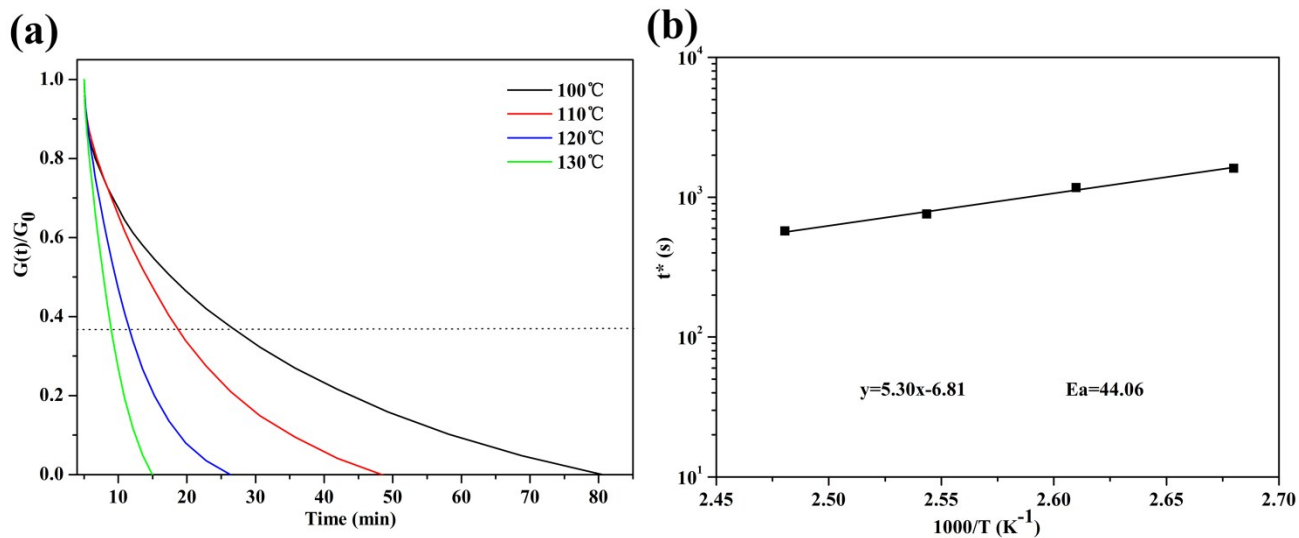


Fig. S7 (a) Stress relaxation analysis of thermoset PU-5% CNT at various temperatures. (b) Fitting of relaxation time (τ) to an Arrhenius equation.

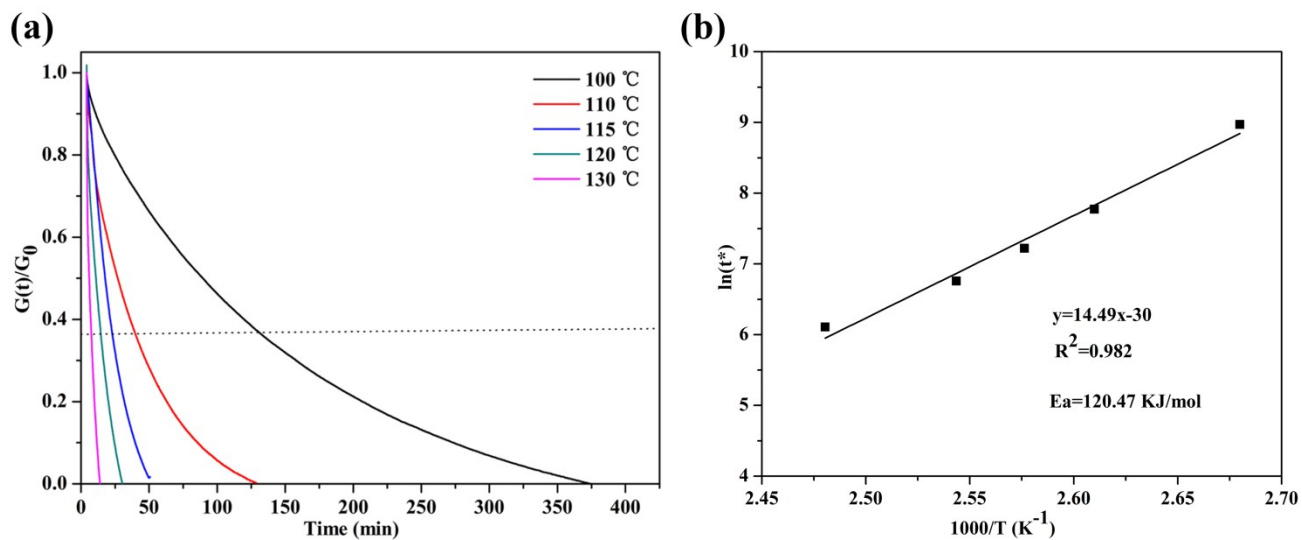


Fig. S8 (a) Stress relaxation analysis of pristine thermoset PU at various temperatures. (b) Fitting of relaxation time (τ) to an Arrhenius equation.

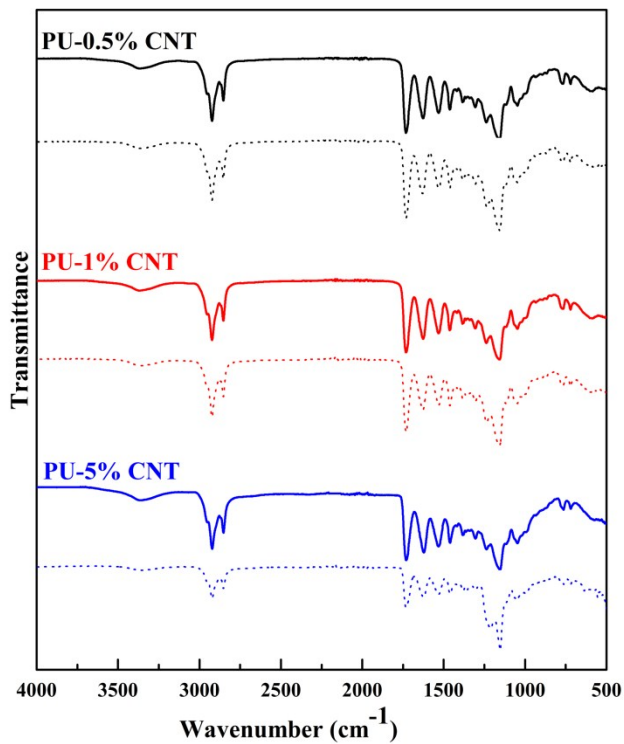


Fig. S9 FTIR spectra of original thermoset PU-x % CNT composites (solid line) and recycled thermoset PU-x % CNT composites (dash dot line).

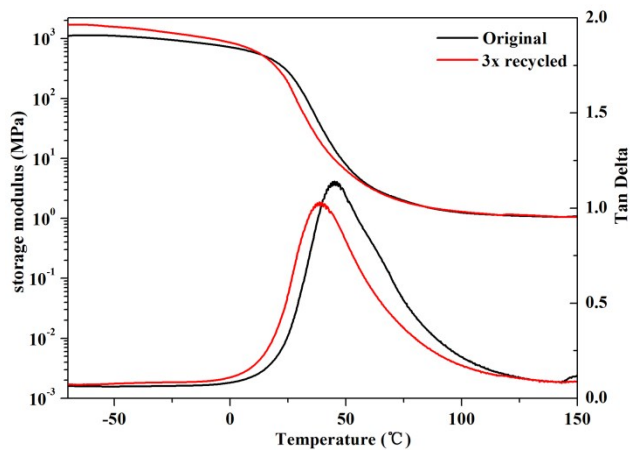


Fig. S10 Storage Modulus and tan delta curves of original thermoset PU-5 % CNT composites (black solid line) and three time recycled thermoset PU-5 % CNT composites (red solid line).

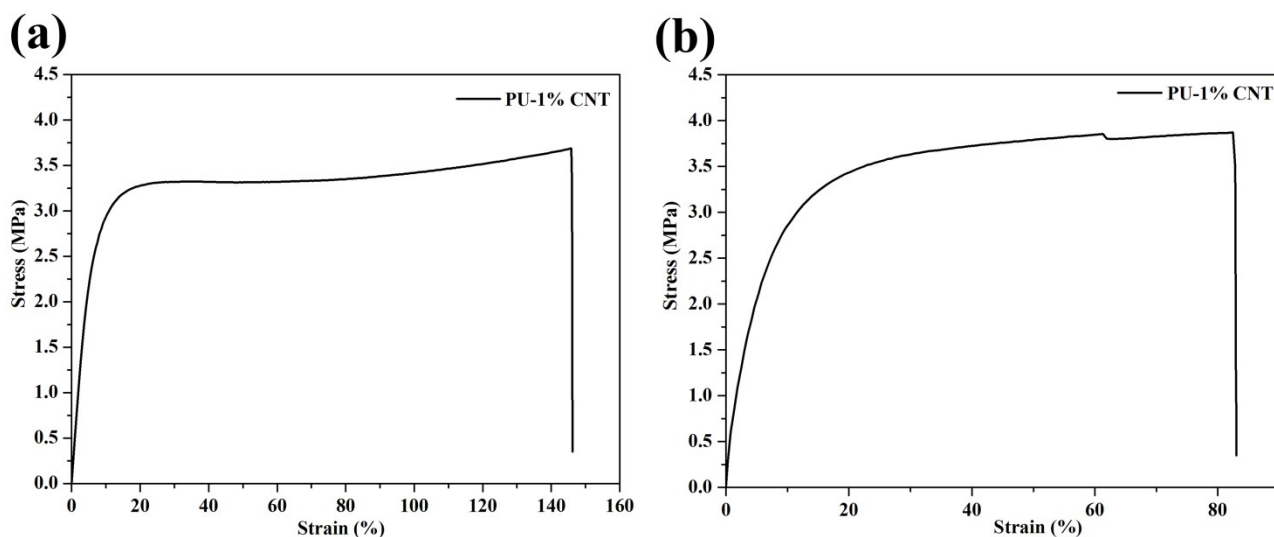


Fig. S11 Stress-strain curves of welded samples thermoset PU-1% CNT for (a) Welding at 110 °C 30 min without load, (b) Welding at IR irradiation (1.14 W cm⁻²) for 2 min.

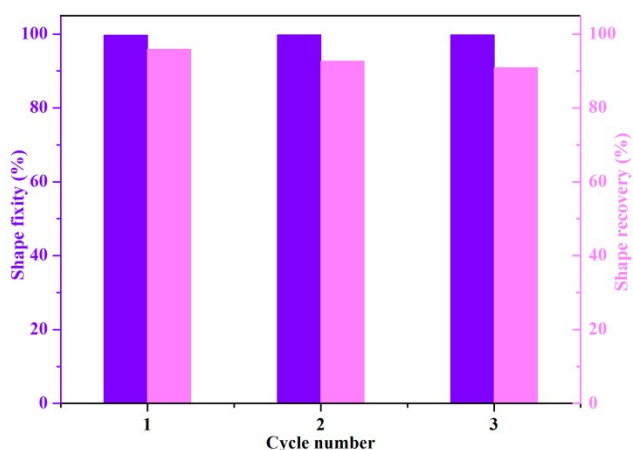


Fig. S12 Consecutive elastic shape memory cycles of the shape fixity and shape recovery.

3. Video

Video S1 The welded sample of PU-1% CNT loading 1 Kg weight.