Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Tunable "soft and stiff", self-healable, recyclable, thermadapt shape-memory biomass polymer based on multiple hydrogen bonds and dynamic imine bondss

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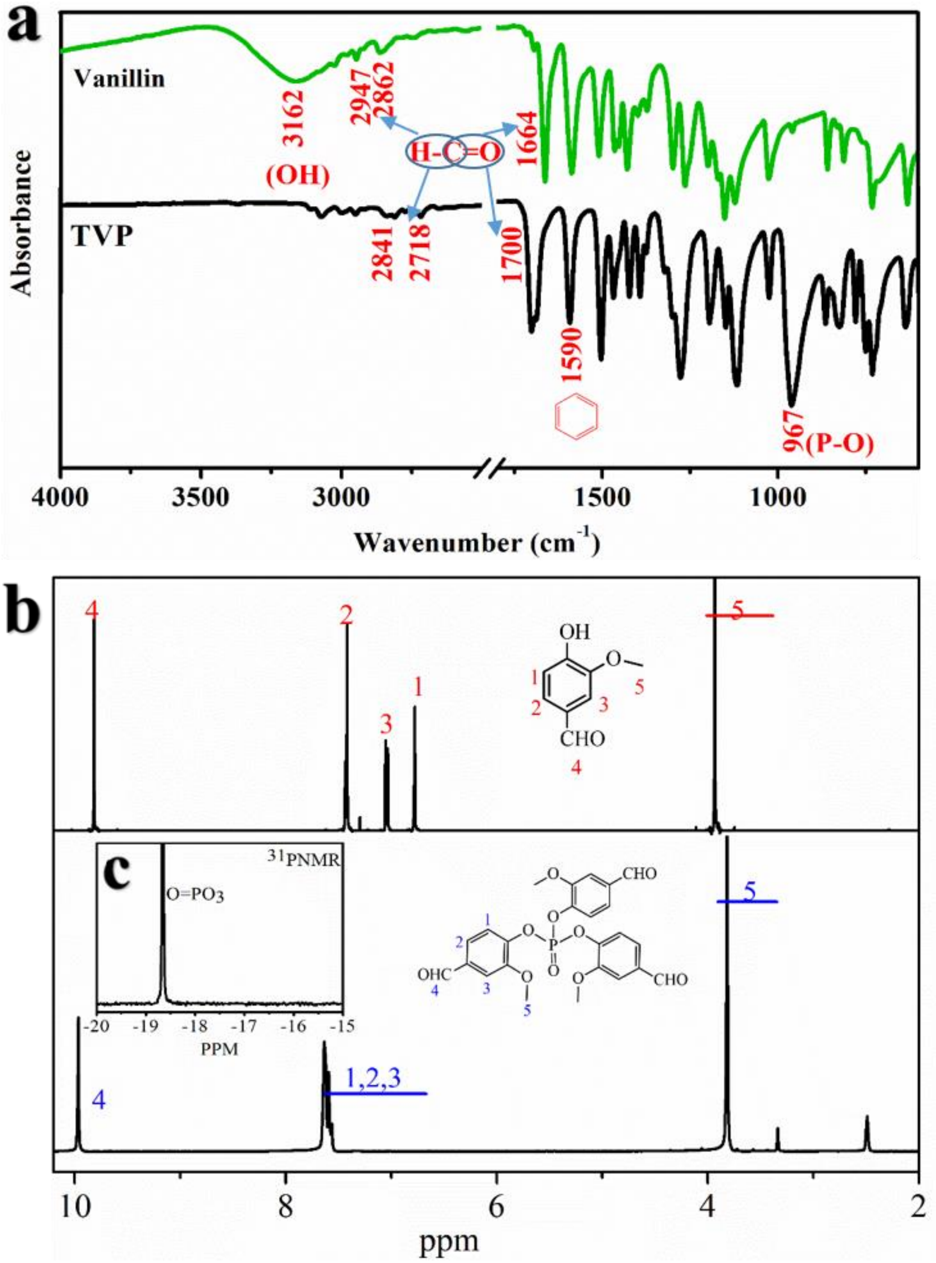

Figure S1. FTIR (**a**) and ^{1}H NMR (**b**) for vanillin and TVP, and ^{31}P NMR (**c**) spectrum for TVP.

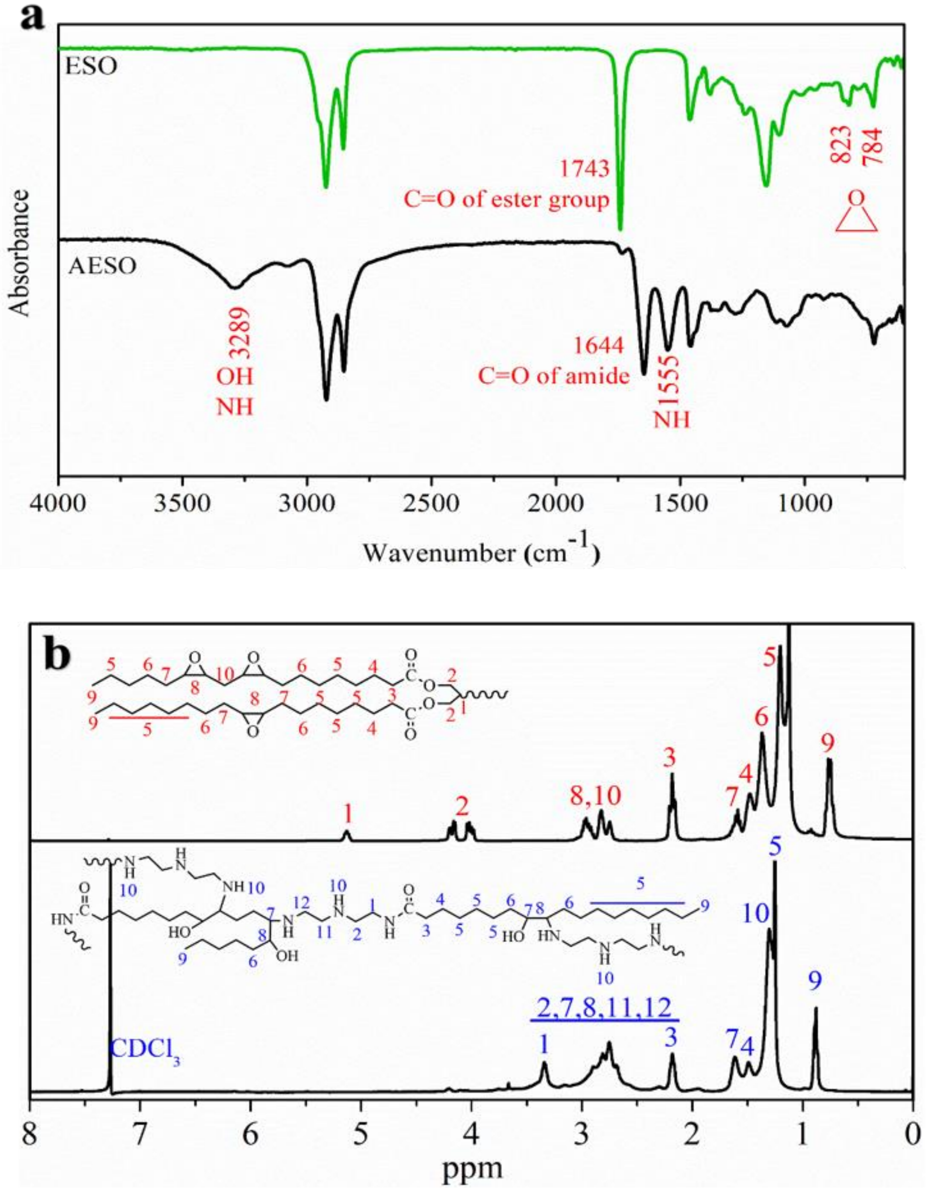

Figure S2. FTIR (**a**) and ^{1}H NMR (**b**) spectrum for ESO and EASO.

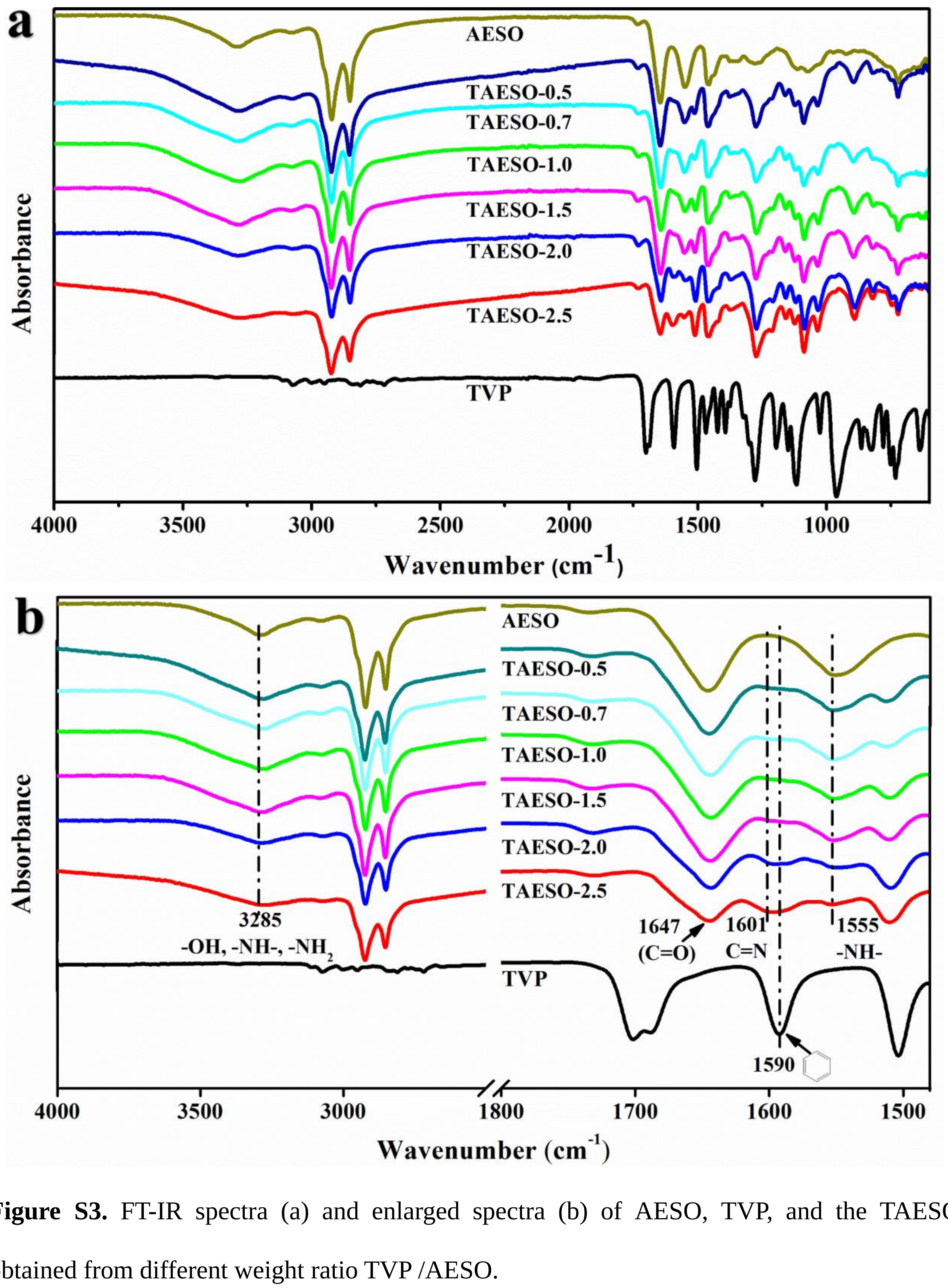

Figure S3. FT-IR spectra (a) and enlarged spectra (b) of AESO, TVP, and the TAESO obtained from different weight ratio TVP /AESO.

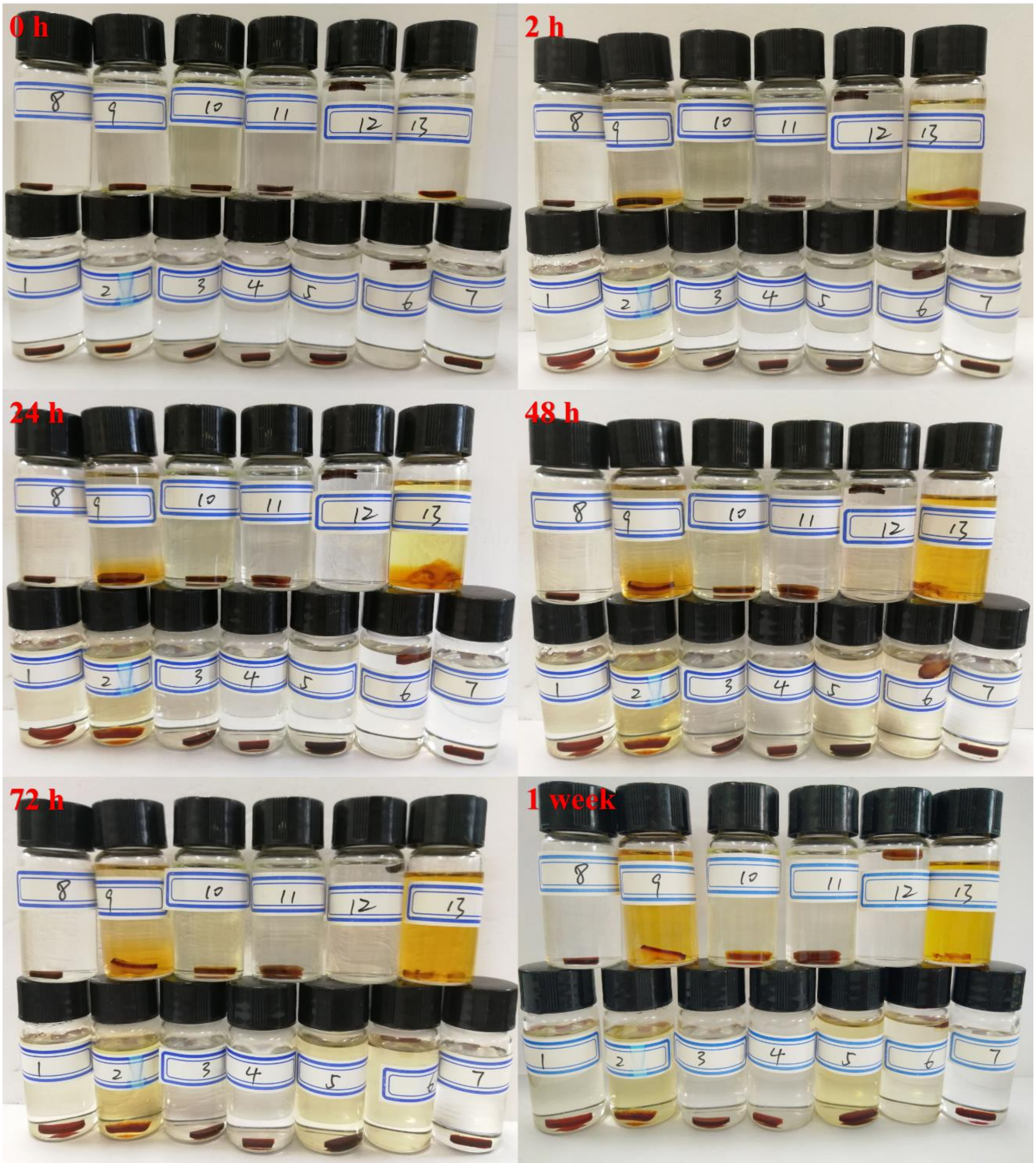

Figure S4. The dissolution experiments for the TAESO-2.5 polymer. It shows that the polymer different levelly swells in solvent 2, 5, 6, 9, 10 and 11, completely dissolve in solvent 13. The corresponding solvents are as followed: 1. Tetrahydrofuran, 2. Dimethyl formamide, 3. Ethyl acetate, 4. Ether, 5. Acetone, 6. Dichloromethane, 7. Toluene, 8. Petroleum ether , 9. Ethanol, 10. Soybean oil 11. 1 mol/L HCl solution, 12. 30 wt % KOH solution, 13. Acetic acid.

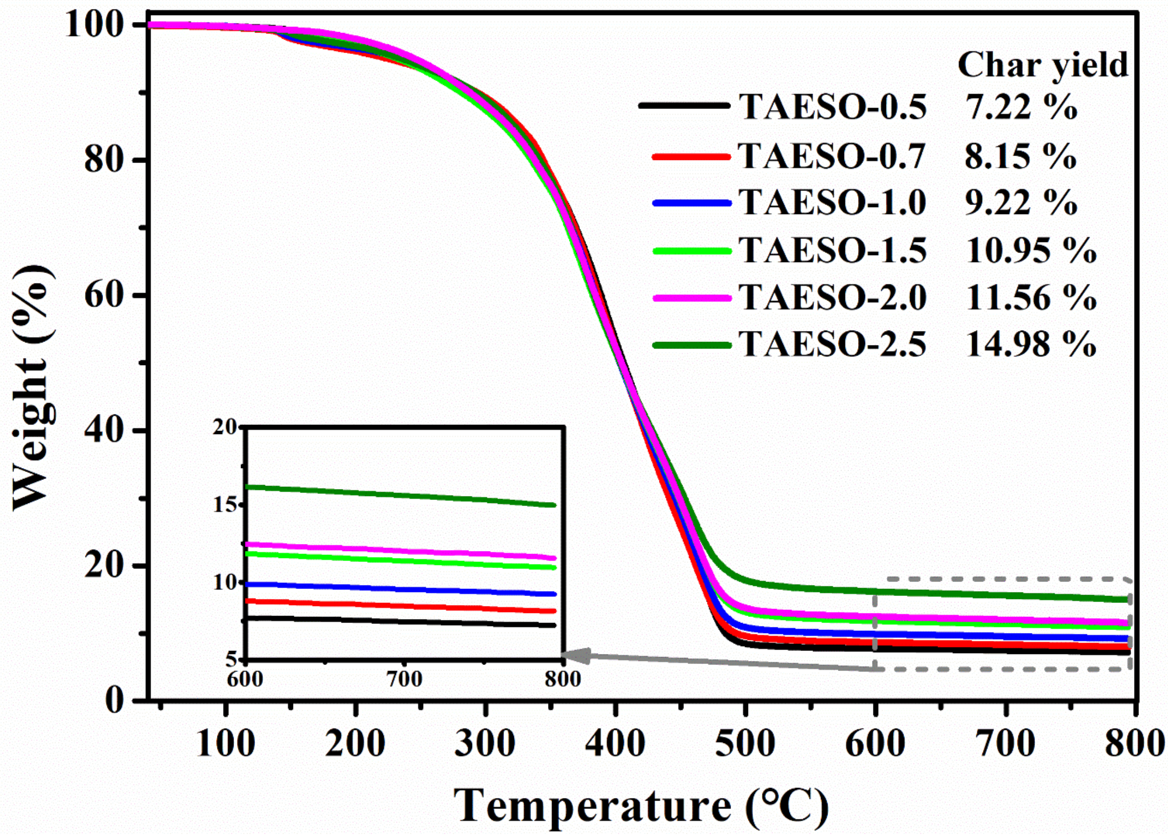

Figure S5. The TGA curves of the TAESO polymer different weight ratio TVP /AESO.

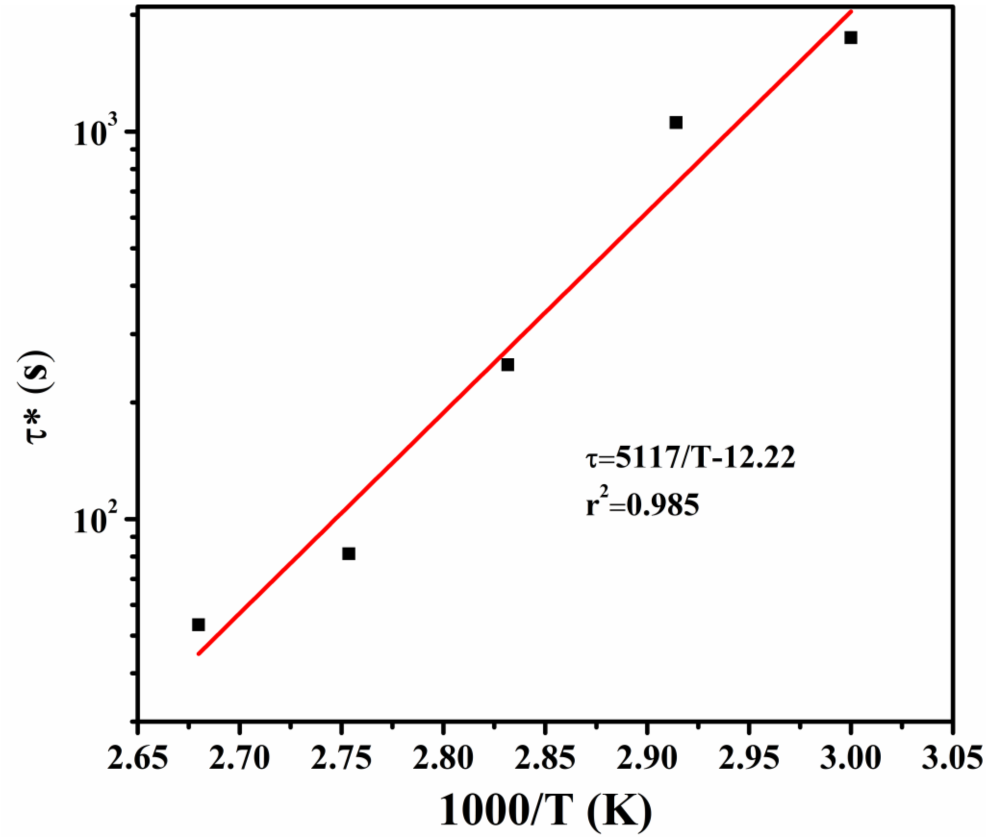

Figure S6. The relaxation time (τ) variation as a function of 1/T, which followed a simple Arrhenius law:

$$\tau^* (T) = \tau_0 \exp(E_a/RT)$$

with τ^* the relaxation time (s), τ_0 a constant (s), E_a the activation energy (J • mol^{-1}), R the ideal gas constant (J • mol^{-1} •K-1) and T the temperature (K). The activation energy E_a, as calculated from the slope (E_a/R), was 43 kJ • mol^{-1}.

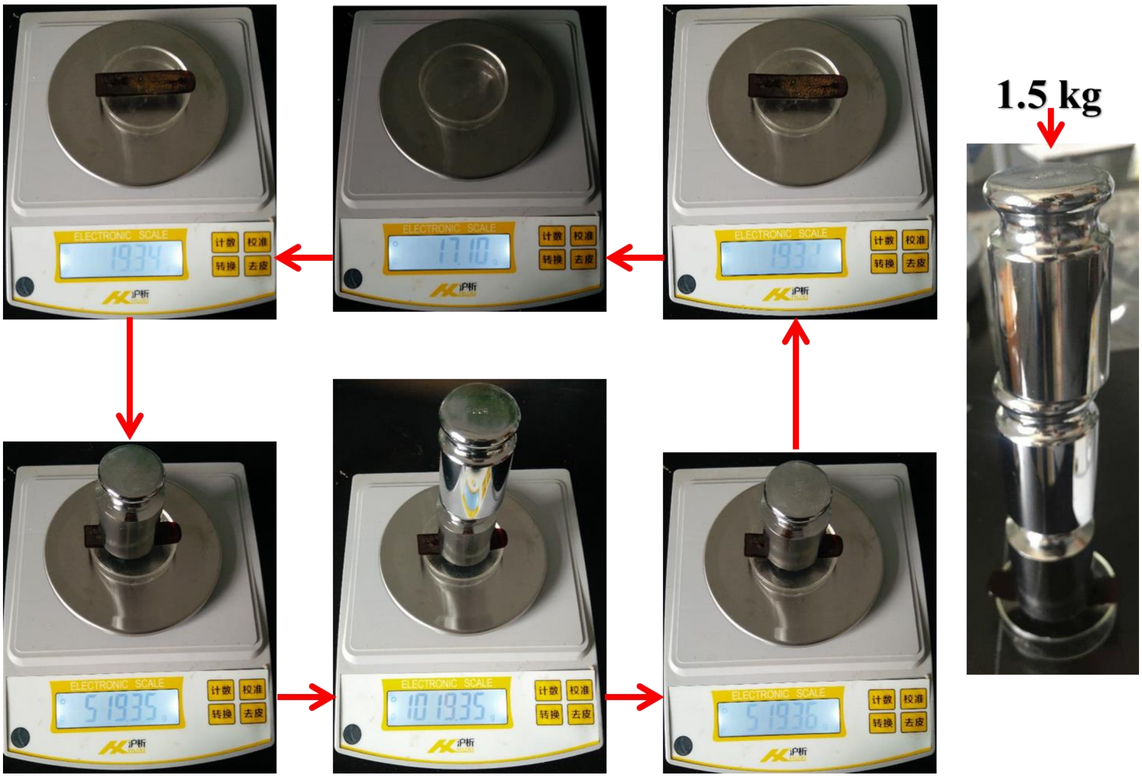

Figure S7. The weight-bearing test of TAESO-2.5.

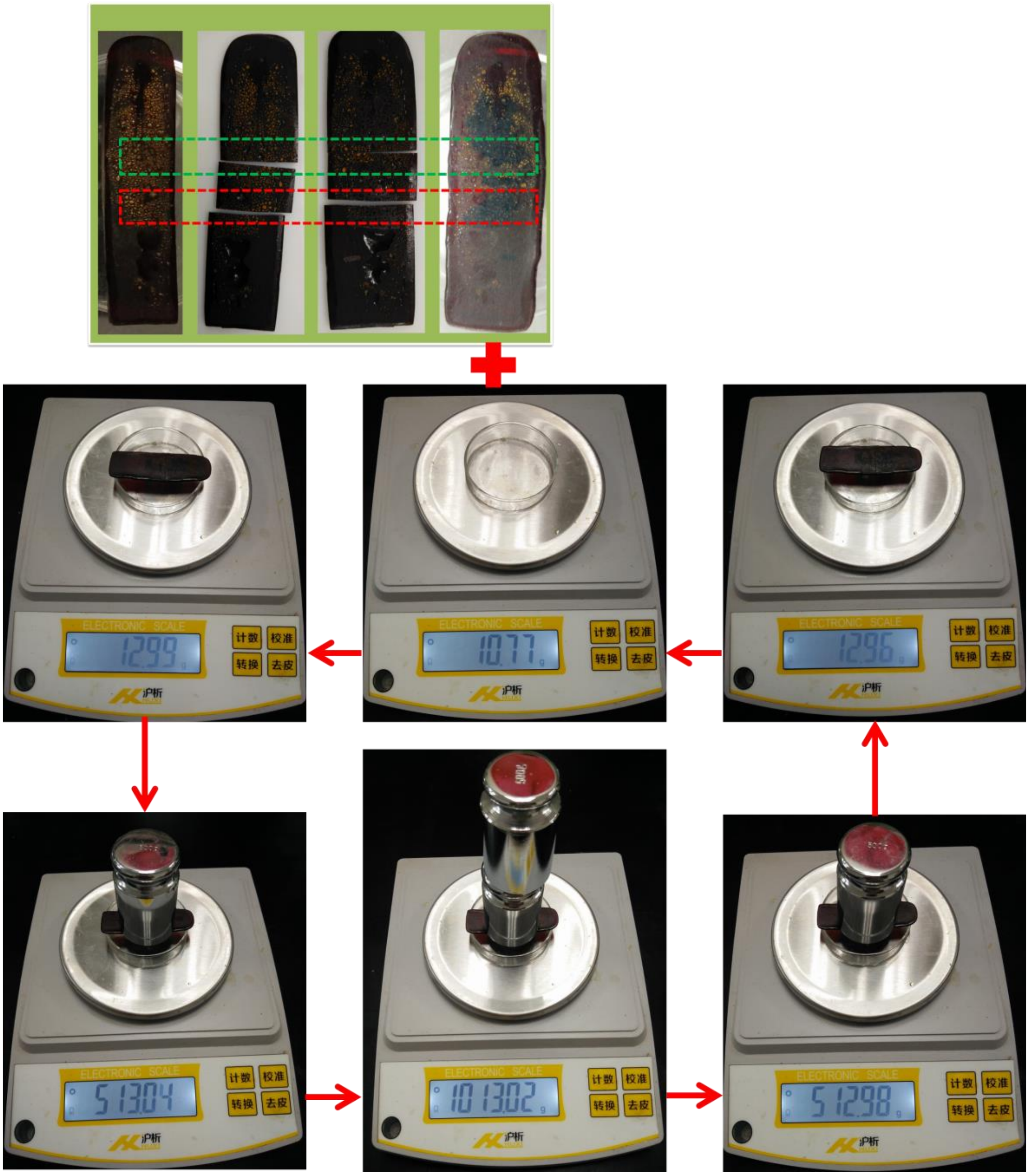

Figure S8. The weight-bearing test of self-healed TAESO-2.5.

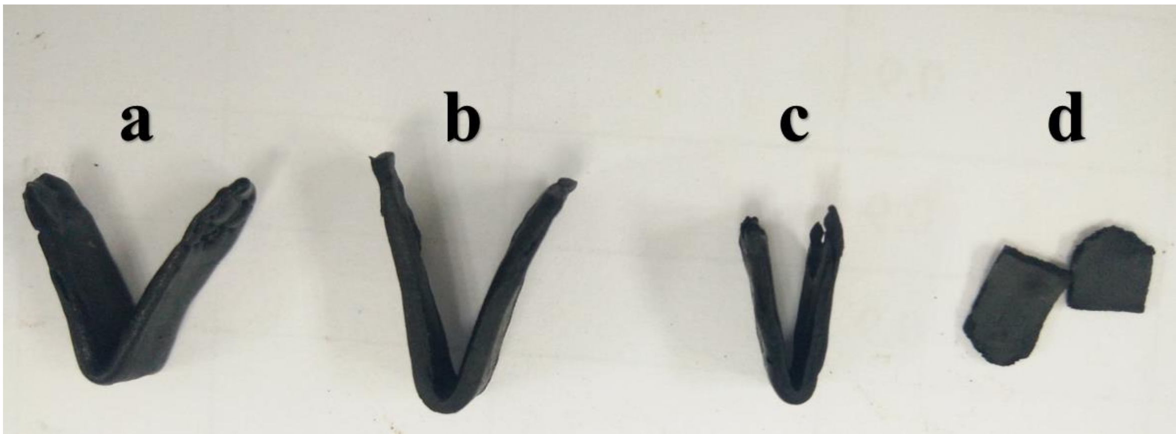

Figure S9. Photographs of composites with different weight ratios of MWCNTs. **a**, 5.0 wt%; **b**, 10.0 wt%; **c**, 15.0 wt%; **d**, 20.0 wt%.

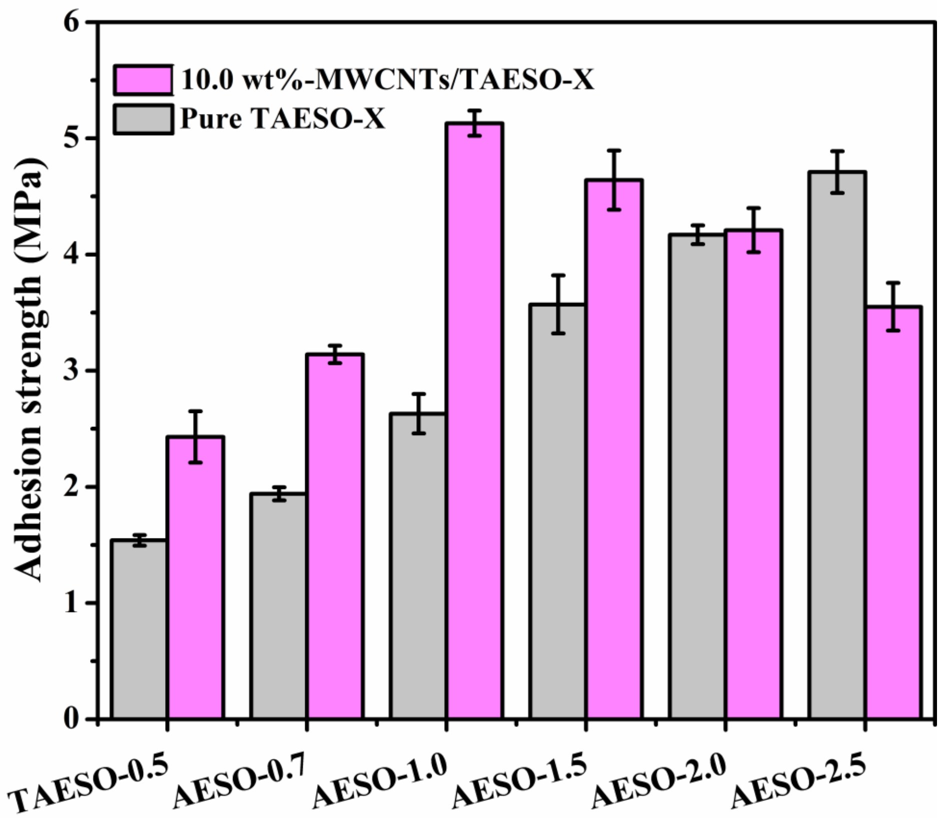

Figure S10. Shear strength of the TAESO-X and 10.0 wt%-MWCNTs/TAESO-X composite.

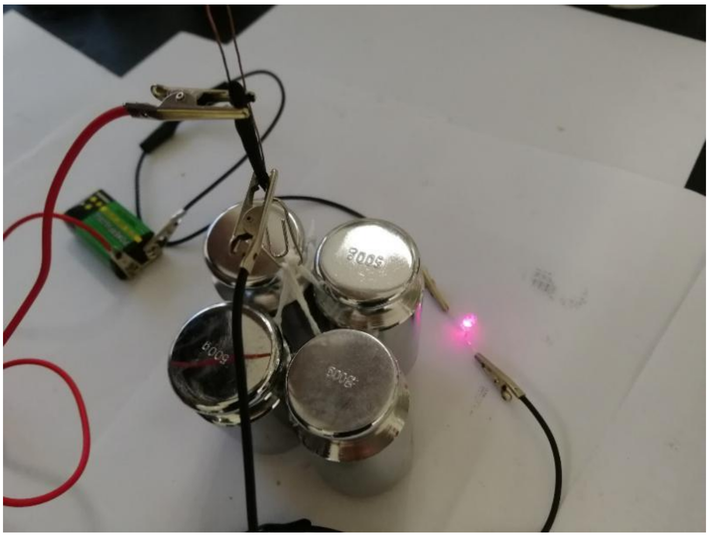

Figure S11. A circuit that glued and conducted by 10.0 wt%-MWCNTs/TAESO-1.0 composite could sustain a weight of 2 kg.

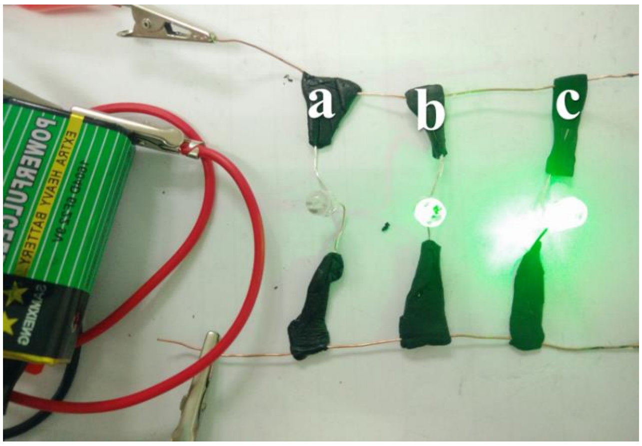

Figure S12. Photographs of the parallel circuit of composites with different weight ratios of MWCNTs. **a**, 5.0 wt%; **b**, 10.0 wt%; **c**, 15.0 wt%.

Table S1. The mechanical properties of TAESO.

	Sample (mean ± s.d., n=4)	Maximal strength (MPa)	Breaking strain (%)
Tensile testing mode	TAESO-0.5	0.63 ± 0.055	>2800
	TAESO-0.7	1.74 ± 0.079	832.57 ± 88.32
	TAESO-1.0	3.20 ± 0.19	464.26 ± 28.17
	TAESO-1.5	7.74 ± 0.33	30.74 ± 5.33
	TAESO-2.0	18.55 ± 0.24	21.29 ± 2.17
	TAESO-2.5	20.50 ± 0.23	12.43 ± 2.49
Bend testing mode	TAESO-1.5	9.77 ± 0.42	2.39 ± 0.26
	TAESO-2.0	20.97 ± 0.79	2.24 ± 0.42
	TAESO-2.5	25.51 ± 2.56	2.03 ± 0.33

Table S2. The healing efficiency of TAESO-2.5 with various healing time and healing temperature.

Healing time (100 °C, h)	**Maximal strength (MPa, mean ± s.d., n=4)**	**Healing Efficiency (%, mean ±s.d., n=4)**
Original	20.50 ± 0.23	-
12	19.11 ± 0.72	93.22 ± 3.50
8	17.87 ± 0.46	87.17 ± 2.22
4	11.20 ± 0.25	54.63 ± 1.25
1	5.77 ± 0.17	28.15 ± 0.84
Healing temperature (12 h, °C)	**Maximal strength (MPa, mean ± s.d., n=4)**	**Healing Efficiency (%, mean ±s.d., n=4)**
Original	20.50 ± 0.23	-
100	19.11 ± 0.72	93.22 ± 3.50
70	17.25 ± 0.48	87.21 ± 2.36
60	12.78 ± 0.71	62.34 ± 3.48
50	6.43 ± 0.32	31.36 ± 1.56
40	3.58 ± 0.15	17.46 ± 0.34

Movie S1. Thermally induced shape recovery of permanent strip sample.

Movie S2. Thermally induced shape recovery of reconfigured permanent spring shape.

Movie S3. Two sanded steel substrates bonded by TAESO-2.5 can withstand a full force of 26.6 kg (approximately equal to 260.9 N).

Movie S4. The parallel circuit of composites with different weight ratios of MWCNTs. From left to right is 5.0 wt%, 10.0 wt% and 15.0 wt%.