Supporting Information

Solvent-Induced Microstructure Disruption and Catalytic Activation Synergy Toward Mild and Rapid Degradation of Epoxy Thermosets

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Supplementary Figures and Table

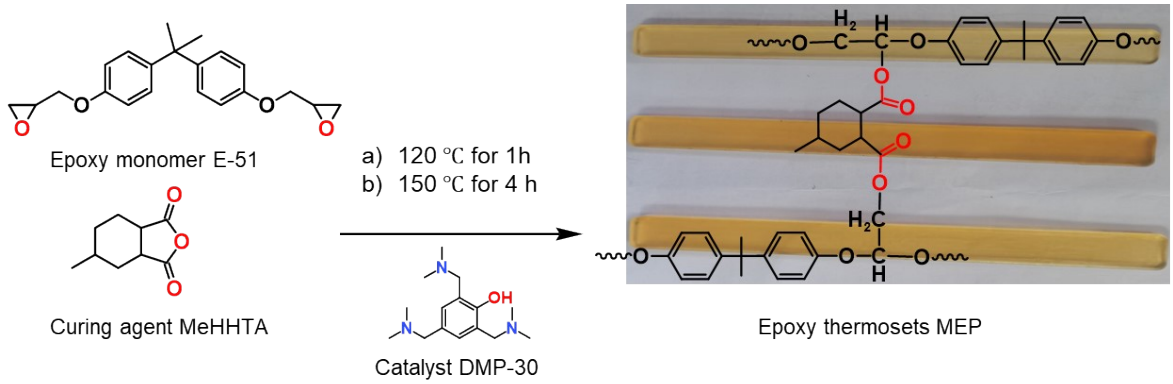

Fig.S1 The curing reaction of epoxy thermosets.

Table S1 Comparison of the catalytic degradation system (this work vs. literature)

System type		Degradation system	Degradation temperature (°C)	Degradation time (min)
Alcoholysis	This work	NaOH/EG/THF	60	30
	Ref 17	K_3PO_4/ethanol	110	180
	Ref 18	NaOH/EG	150	60
	Ref 19	K_3PO_4/PEG400	130	60
	Ref 20	DBU/EG	140	120
	Ref 21	$Zn(Ac)_2$/EG	170	>240
	Ref 22	$Zn(Ac)_2$/2-ethyl-hexanol	170	~150
	Ref 23	NaOH/PEG200	180	50
	Ref 24	4-(Dimethylamino) pyridine/EG	180	60
Amination	This work	NaOH/DETA/THF	60	30
	Ref 25	Diethylenetriamine	130	55
	Ref 26	Hydrazine hydrate	120	60
	Ref 27	Aminoethanol	160	240
Hydrolysis	This work	NaOH/H_2O/THF	60	70
	Ref 28	$FeCl_3$/H_2O	170	30
	Ref 29	Surfactants/H_2O	170	~720
	Ref 30	DBSA/H_2O	170	~660
	Ref 31	HPW/H_2O	190	150

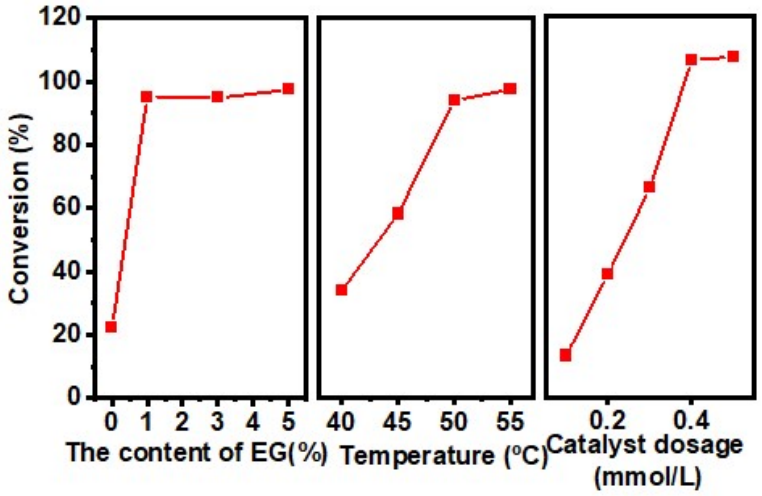

Fig.S2 The effect of reaction conditions on alcoholysis degradation conversion.

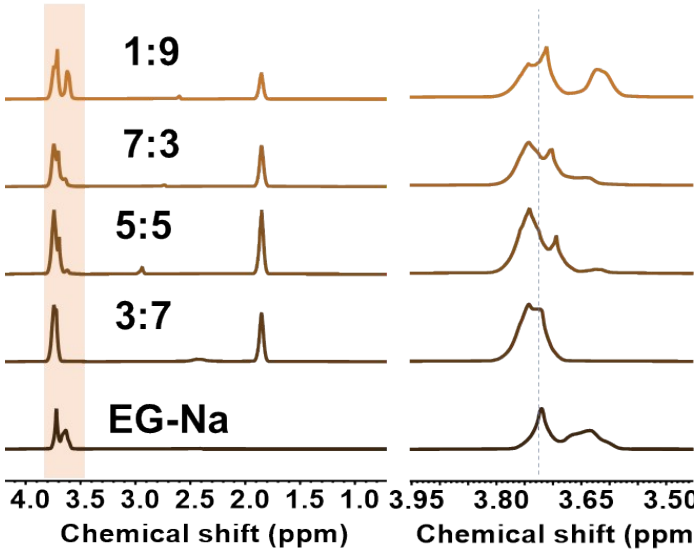

Fig.S3 ^{1}H-NMR spectra of ethylene glycol sodium salt/THF mixtures at varied ratios.

Calculation Methods and Modeling Description

• [Computational Software]:

This work is based on Gaussian16 C.01 software for calculations.

• [Relevant convergence accuracy, simulation steps, and other specific parameters]

Description of calculation methods: Calculations were carried out using the Gaussian 16 software package [1]. Geometry optimizations were performed at the B3LYP[2]-D3(BJ)[3]/ def2svp [4,5] level. Single-point energy calculations and ESP calculations were performed at the B3LYP[2]-D3(BJ)[3]/ def2tzvp [4,5] level to obtain more accurate energetics. The ESP is obtained by Multiwfn 3.8 Dev [6-8] program and the ESP molecular surfaces are visualized by VMD[9].

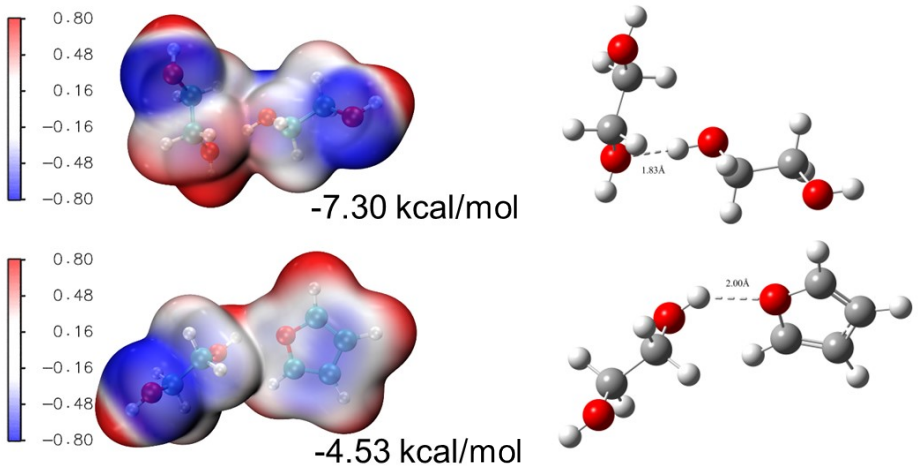

Fig.S4 Electrostatic potential, binding energy and Hydrogen bond length of two molecules of ethylene glycol and ethylene glycol/tetrahydrofuran.

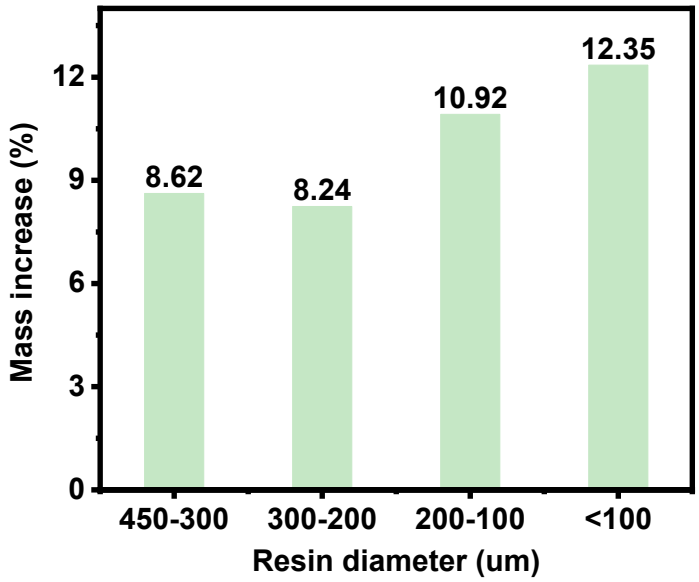

Fig.S5 The influence of different particle size on the mass increase of epoxy thermosets.

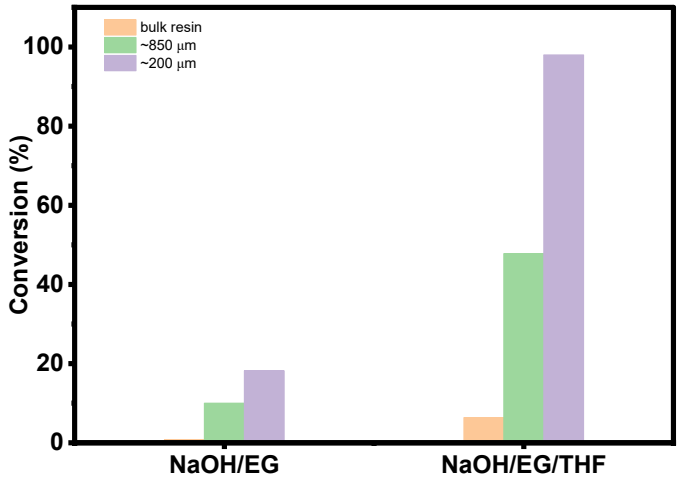

Fig.S6 The effect of particle size on conversion in different degradation systems.

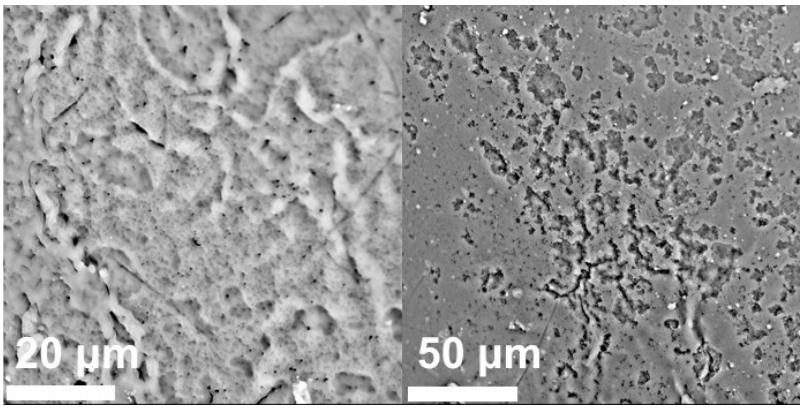

Fig.S7 SEM of epoxy thermoset via THF assisted alcoholysis.

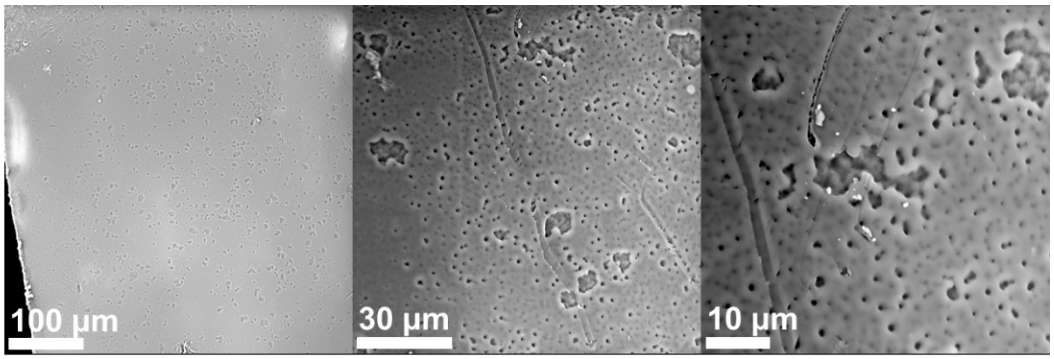

Fig.S8 SEM of epoxy thermoset via DMF assisted alcoholysis.

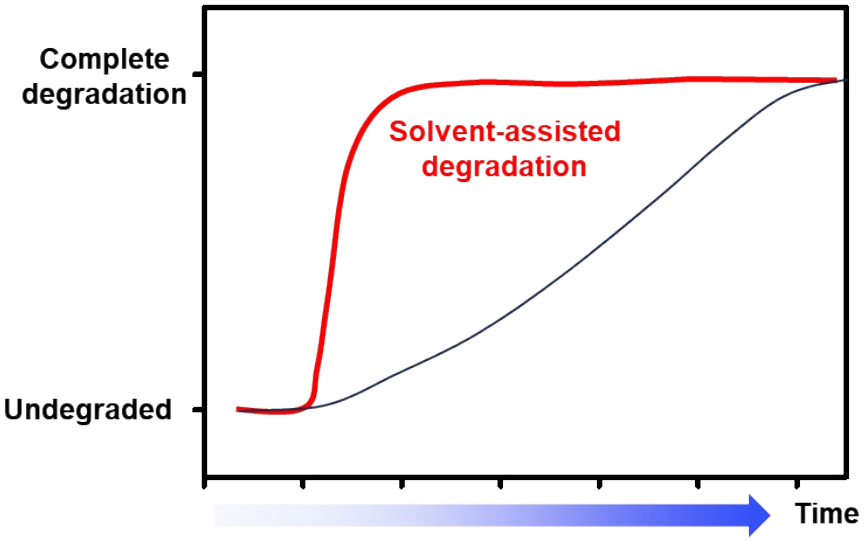

Fig.S9 Curve model of degradation of THF/EG/NaOH system.

The calculation of purity:

$$\text{Purity}(\%) = \left(\frac{A_{sample}/N_{H,sample}}{A_{IS}/N_{H,IS}}\right) \times \frac{W_{IS}}{W_{sample}} \times \frac{MW_{sample}}{MW_{IS}} \times P_{IS} \times 100\%$$

A_{sample}: Integral area of the selected characteristic peak of the analyte (sample). $N_{H,sample}$: Number of protons producing the characteristic peak of the sample. A_{IS}: Integral area of the selected characteristic peak of the internal standard. $N_{H,IS}$: Number of protons producing the characteristic peak of the internal standard. W_{IS}: Mass of the internal standard (mg or g). W_{sample}: Total mass of the sample to be tested (mg or g). MW_{sample}: Molecular weight of the sample (g/mol). MW_{IS}: Molecular weight of the internal standard (g/mol). P_{IS}: Purity of the internal standard (0.98, i.e., 98%).

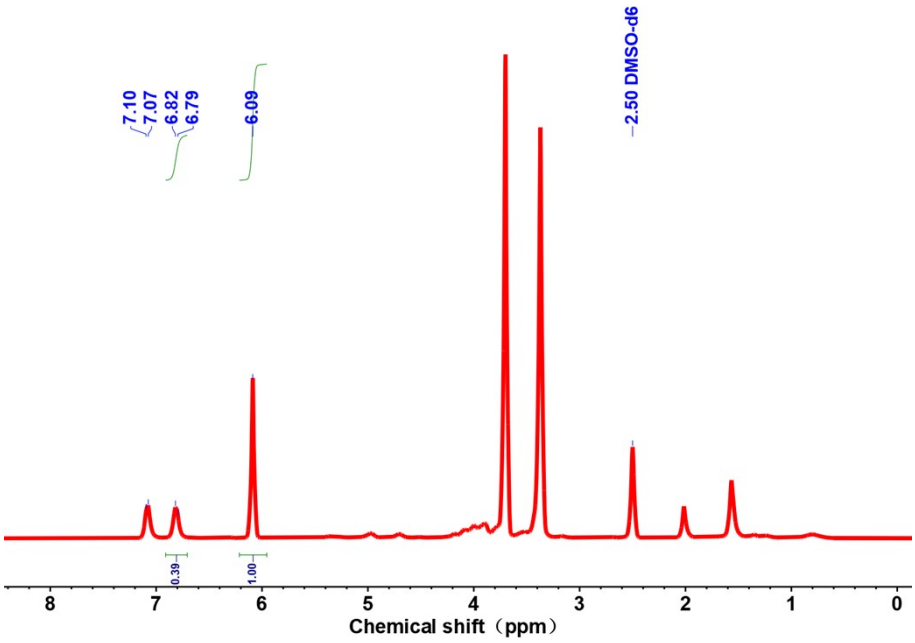

Fig.S10 ^{1}H-NMR spectrum of DEP-1 and 1,3,5-Trimethoxybenzene.

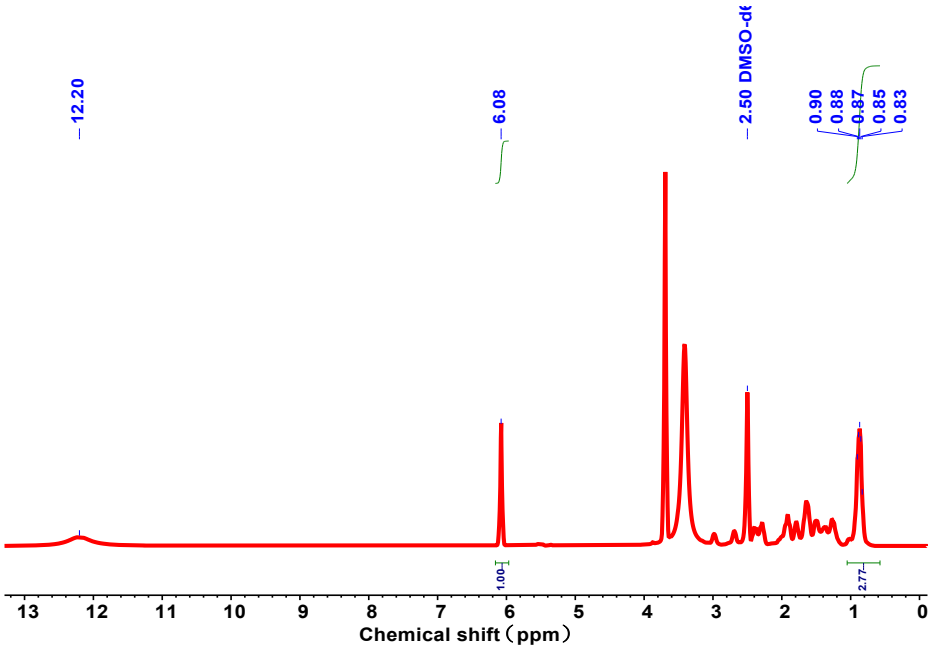

Fig.S11 ^{1}H-NMR spectrum of DEP-2 and 1,3,5-Trimethoxybenzene.

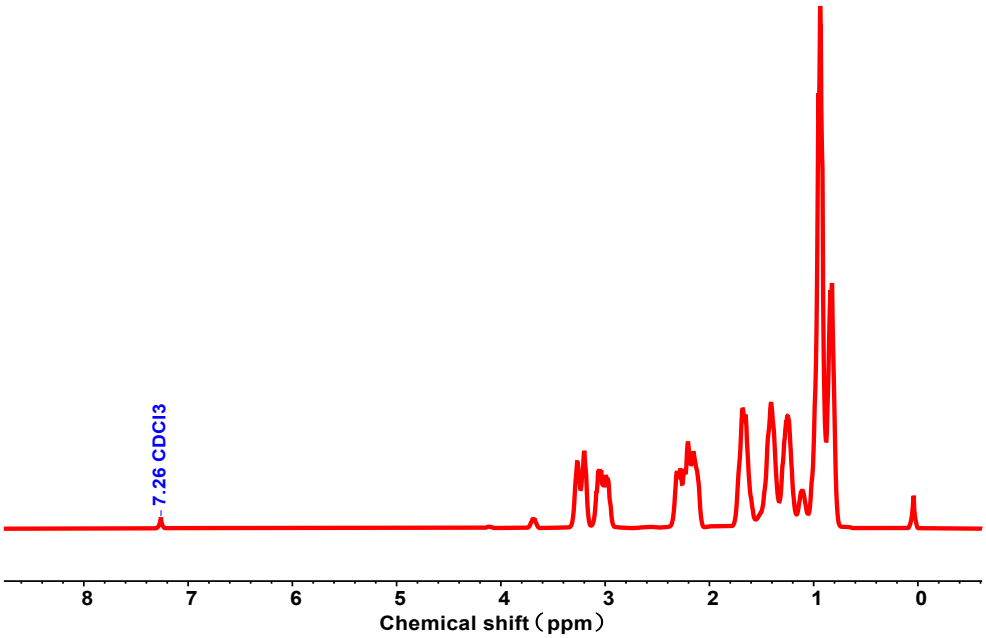

Fig.S12 ^{1}H-NMR spectrum of methylhexahydrophthalic anhydride (MeHHPA).

The calculation of E-factor:

The E-factor was defined as the mass of waste generated per unit mass of the target product. According to the calculation method based on literature, during the degradation process, the waste amount was determined by subtracting the product mass and the mass of recovered solvent or catalyst from the input mass, and the calculation formula is provided below.

$$E-factor=\frac{W_{waste}}{W_{product}}=\frac{W_{input}-W_{recycled}-W_{product}}{W_{product}}$$

The calculation of Energy-saving:

Based on the literature method for calculating energy consumption, the energy consumption of the degradation process is calculated as the theoretical energy required for the reaction mixture to reach a specific temperature from room temperature.

$$Q=mc_p\Delta T=(m_{THF}c_{THF}+m_{EG}c_{EG})\Delta T=(V_{THF}\rho_{THF}c_{THF}+V_{EG}\rho_{EG}c_{EG})\Delta T$$

where m represent the mass of solvent (g); c_p represent the specific heat capacity of solvent, respectively ($Jg^{-1}°C^{-1}$); ΔT is the temperature change; V and ρprepresents the volume (mL) and density (g/mL) of the solvent.

The total energy-saving coefficient (η) of this work can be calculated as follows, compared with the energy required for similar works in the literature.

$$\eta=\frac{Q_{literature}-Q_{this\ work}}{Q_{literature}}\times 100\%$$

In cases where the literature did not specify the exact amount used, it was estimated. If the product yield was not provided, it was calculated based on the theoretical maximum yield.

Table S2 compares the E-factor and energy consumption with similar works in the literature

Degradation system	Matter	Temperature	Recovered	E-factor	Energy
NaOH/EG/THF	EP:0.75 g NaOH:0.5 g EG:1 g THF:9 g	60℃	Product:0.45 g THF:8.5 g NaOH:0.45 g	**4.1**	**0.641 kJ** **0.855 kJ (1g EP)**
NaOH/EG	EP:3 g NaOH:1 g EG:20 mL	150℃	Product:2 g	11	6.533 kJ 2.178 kJ (1g EP)
NaOH/PEG	EP:2 g NaOH:0.8 g PEG:40 mL	180℃	Product:2 g	20.4	16.216 kJ 8.108 kJ (1g EP)

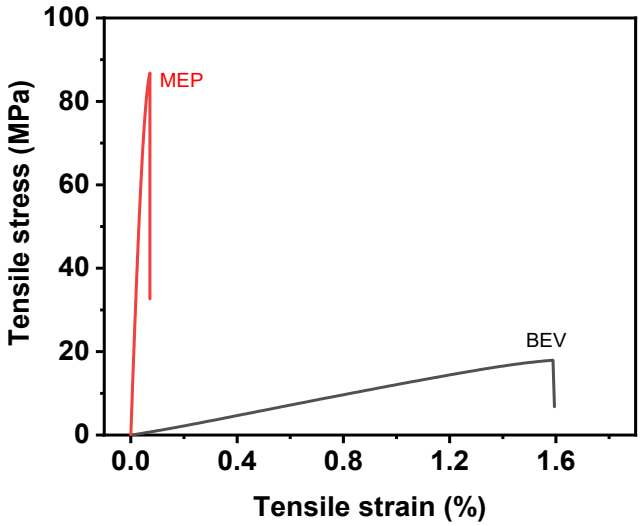

Fig.S13 The stress-strain curves of MEP and BEV.

Table S3 Comparison of the properties/parameters of MEP and BEV

Performance/Parameters	MEP	BEV
Tensile strength（MPa）	83.56	17.9
Elastic modulus（MPa）	1753.57	1259.8
Elongation at break（%）	0.07	1.59
Shape memory	x	√
Healing	x	√
Physical recycling	x	√

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