Supporting Information

A fully degradable epoxy resin based on a nontoxic triphenol derived from diphenolic acid and eugenol

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1. Characterization

Nuclear magnetic resonance (NMR) spectra were recorded on a Bruker Avance AV400 spectrometer (400 MHz, Bruker Corp., Switzerland) using DMSO-d6 as solvent and tetramethylsilane (TMS) as reference. Fourier transform infrared (FTIR) spectra were obtained on a Nicolet 5700 FTIR spectrometer (Nicolet Corp., USA) with a resolution of 4 cm^{-1} and a scanning range of 400 to 4000 cm^{-1}. Elemental analyses were performed on a Vario Macro cube elementar analyzer (Elementar Corp., Germany). Differential scanning calorimetry (DSC) tests were carried out on a DSC-2 (Mettler -Toledo Corp., Switzerland) with heating rate of 10 °C/min, N_2 flow rate of 45 mL/min, and a temperature range of 30 to 300 °C. Dynamic mechanical analysis (DMA) was performed using a DMA-1 analyzer (Mettler-Toledo Corp., Switzerland) with a sample size of 40 mm × 10 mm × 1.5 mm, heating rate of 3 °C/min, and temperature range of 30 to 250 °C. Thermo gravimetric analysis (TGA) was performed on a TGA/DSC 2 (Mettler Toledo Corp., Switzerland) with heating rate of 10 °C/min, N_2 flow rate of 50 mL/min, and temperature range of 30 to 800 °C. Stress-strain experiments were performed on a universal testing machine (5943, INSTRON, USA) with a tensile rate of 5 mm/min. The TGA-GC/MS system consists of four components: thermal gravimetry (TGA, Mettler-Toledo corp., Switzer-land), sample storage (SRA IST16, Mettler-Toledo corp., Switzer-land), gas chromatography (GC, 7890GC, Agilent Technologies Inc., USA), and mass spectrometry (MS, 5975MSD, Agilent Technologies Inc., USA). Volatile components were collected from the TGA unit,

temporarily stored in the sample storage instrument and sequentially injected into the GC-MS unit. The GC separation was carried out using an Agilent 19091s-433 capillary column (30 m, 0.25 mm i.d., 0.25 μm thickness). The injector temperature was set at 240 °C in the split mode with the split ratio of 5: 1 with a helium gas flow of 1.2 mL/min. MS transfer line and its oven temperatures were set at 260 °C, respectively. The decomposition products were identified using mass spectrum library attached in the GC/MS system. The fracture surface morphology and carbon fiber surface before and after degradation were tested by scanning electron microscopy (SEM) (3000 V, Vltra55, ZEISS, Germany), and the surface of the samples was gold-plated before testing. Raman spectra (Raman spectra) were obtained on inVia-Reflex (Renishaw, UK) using a laser with a wavelength of 532 nm and an energy of 12 mw to test the chemical structure indicated by the carbon fibers in the range of 500 to 4000 cm^{-1}.

2. Complementary information of all compounds

EGE: Yield = 75%, ^{1}H NMR (DMSO-d6), δ = 8.78, 6.84, 6.69, 3.76, 3.08, 2.70 and 2.56; ^{13}C NMR (DMSO-d6), δ = 147.94, 145.59, 128.94, 121.49, 115.84, 113.61, 56.11, 52.67, 46.54 and 38.24; Partially good solvents: ethanol, ethyl acetate, dichloromethane, acetone etc.

DEE: Yield = 83%, ^{1}H NMR (DMSO-d6), δ = 6.96, 6.67, 5.03, 3.73, 2.68, 2.24, 1.95 and 1.47; ^{13}C NMR (DMSO-d6), δ = 173.21, 155.45, 147.93, 145.45, 139.43, 129.62, 128.63, 121.56, 116.63, 115.79, 113.49, 55.97, 52.69, 46.52, 44.18, 38.21, 33.13, 31.09 and 27.66; elemental analyses, C 69.53, H 6.43, O 24.04 for $C_{27}H_{30}O_{7}$;

found C 69.28, H 6.61; Partially good solvents: ethanol, ethyl acetate, acetone, etc.

DEE-EP: Yield = 81%, ^{1}H NMR (DMSO-d6), δ =7.07, 6.88, 5.01, 4.29, 3.96, 3.78, 3.31, 2.83, 2.76, 2.71, 2.32, 2.09 and 1.53; ^{13}C NMR (DMSO-d6), δ =173.21, 156.65, 147.39 145.31, 138.77, 128.38, 126.88, 121.67, 115.14, 114.45, 111.56, 69.36, 55.82, 52.54, 50.21, 47.21, 44.19, 38.20, 34.23, 32.18 and 27.51; elemental analyses, C 68.14, H 6.62, O 25.24 for $C_{36}H_{42}O_{10}$; found C 67.78, H 6.73; EEW, experimental value 197.8, theoretical value 212.5; viscosity: 10.3 Pa·s at 50 °C; Partially good solvents: dichloromethane, ethyl acetate, acetone, ethanol, etc.

3. Equations for calculating Ea of the polymerization

The apparent activation energies of the polymerization reactions were calculated according to the Kissinger (eq S1) and modified Ozawa (eq S2) methods.

$$-\ln\left(\frac{\beta}{T_P^2}\right)=\frac{E_a}{RT_P}-\ln\left(\frac{AR}{T_P}\right) \quad (S1)$$

$$\ln\beta=-1.052\times\frac{E_a}{RT_P}+c \quad (S2)$$

where β is the heating rate, T_p is the peak exothermic temperature, and R is the gas constant.

4. Equation for calculating cross-linking density

According to the theory of entropic elasticity of rubber, the cross-linking density can be calculated from eq S3.

$$\nu_e=\frac{E'}{3RT} \quad (S3)$$

where υ_e is the crosslink density, E' is the energy storage modulus at T_g+30k, R is the gas constant, and T is the absolute temperature at T_g+30k.

5. Supplementary Figures and Tables

Table S1. Curing process of epoxy resin/hardener mixture

Mixtures (epoxy/hardener)	Mole ratio (epoxy/hardener)	Curing promoter	Heating process
DEE-EP/SA	1/1.5		140℃（2h），160℃（2h），180℃（2h）
DEE-EP/DDS	1/0.75		180℃（2h），200℃（2h），220℃（2h）
DEE-EP/EMI	1/0.3		90℃（1h），120℃（1h），150℃（2h）
DGEBA/SA	1/1	BDMA*	140℃（1h），160℃（1h），180℃（1h）
DGEBA/DDS	1/0.5		180℃（2h），200℃（2h），220℃（2h）
DGEBA/EMI	1/0.2		100℃（1h），130℃（1h），150℃（1h）

* BDMA dosage: 0.5 wt% of DGEBA.

Table S2. Degradation conditions of cured resin or composite

	Organic solvent	Organic solvent to water ratio	Acid concentration (mol/L)	Acid type	Temperature (℃)
1	THF	8/2	1	HCl	25

2	DMF	8/2	1	HCl	25
3	DMSO	8/2	1	HCl	25
4	Ethanol	8/2	1	HCl	25
5	THF	8/2	1	HCl	25
6	THF	6/4	1	HCl	25
7	THF	4/6	1	HCl	25
8	THF	2/8	1	HCl	25
9	THF	8/2	2	HCl	25

Table S2 (continued). Degradation conditions of cured resin or composite

	Organic solvent	Organic solvent to water ratio	Acid concentration (mol/L)	Acid type	Temperature (℃)
10	THF	8/2	1	HCl	25
11	THF	8/2	0.5	HCl	25
12	THF	8/2	0.05	HCl	25
13	THF	8/2	1	H_2SO_4	25
14	THF	8/2	1	HCl	25
15	THF	8/2	1	H_3PO_4	25
16	THF	8/2	1	CH_3COOH	25
17	THF	8/2	1	HCl	25
18	THF	8/2	1	HCl	35
19	THF	8/2	1	HCl	45
20	THF	8/2	1	HCl	55

Table S3. Important DSC factors of the epoxide mixtures and their activation energy (E_a) values.

Mixture	T_p(℃)				E_a(kJ/mol)	
	5℃/min	10℃/min	15℃/min	20℃/min	Kissinger	Ozawa
DEE-EP/SA	165	181	188	193	75.5	83.9
DGEBA/SA	269	286	300	307	79.1	89.1
DEE-EP/DDS	176	193	198	203	79.5	81.4
DGEBA/DDS	197	214	222	228	83.8	85.1
DEE-EP/EMI	96	109	116	119	64.4	84.9
DGEBA/EMI	113	120	128	132	67.4	87.2

Table S4. Main thermal and mechanical properties of the cured epoxy.

Cured resin	T_g(℃)	E'(25℃) (MPa)	$v_e \times 10^3$ (mol/m^3)	T_{d10} (℃)	CY (%)	Tensile strength (MPa)	Elongation at break(%)
DEE-EP/SA	85	1765	1.53	354	22	57.1 ± 2.3	5.4 ± 0.3
DGEBA/SA	77	2478	1.38	372	12	50.4 ± 2.1	5.3 ± 0.5
DEE-EP/DDS	181	2943	1.45	305	23	67.9 ± 3.4	5.5 ± 0.4
DGEBA/DDS	168	2633	1.18	394	12	59.4 ± 4.2	4.3 ± 0.2
DEE-EP/EMI	134	2254	1.48	318	25	56.2 ± 4.5	4.0 ± 0.1
DGEBA/EMI	121	2530	1.36	371	6	45.5 ± 2.7	3.7 ± 0.2

Table S5. Dissolution rate of DEE-EP/SA resin in different solvents

Sample	Swelling ration (%)

	THF	DMF	Ethanol	DMSO	H_2O
DEE-EP/SA	22.3	15.5	1.8	11.7	0.4

Table S6. Mechanical properties of composites and carbon fibers before and after recycling

Sample	Stress (MPa)	Strain (%)
DEE-EP/SA/CF	713.7 ± 12.8	3.3 ± 0.3
DEE-EP/SA/RCF	655.6 ± 9.8	3.0 ± 0.2
Ref 56	145.4 ± 17.1	19.2 ± 0.9
Ref 57	622.0 ± 17.0	4.0 ± 0.2
Ref 58	381.0 ± 66.0	4.2 ± 0.6
Virgin CF	3892.1±16.5	2.1±0.3
Recycled CF	3606.6±10.1	1.9±0.2

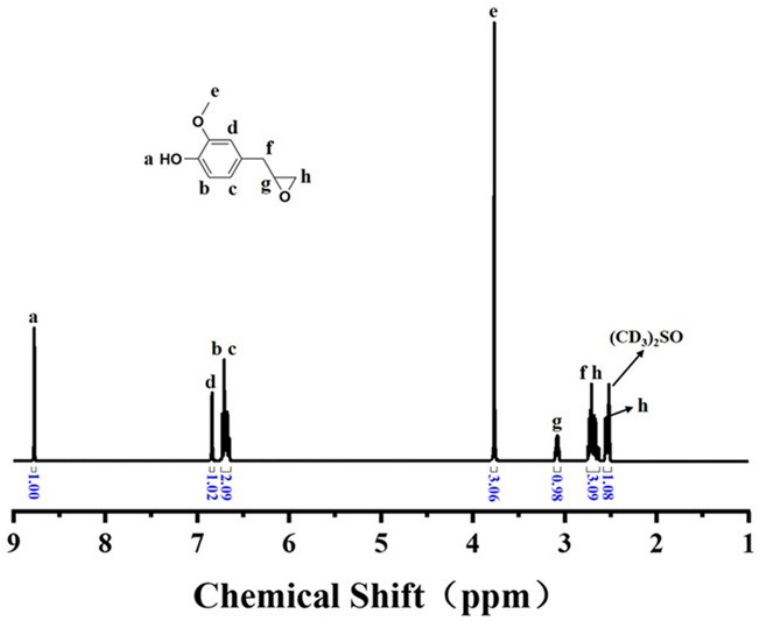

Figure S1. The ^{1}H NMR spectrum of EGE.

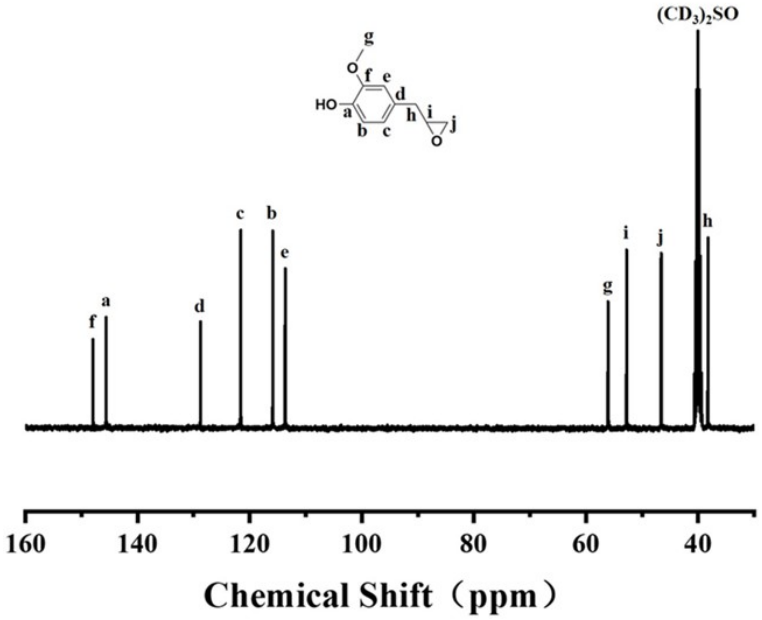

Figure S2. The ^{13}C NMR spectrum of EGE.

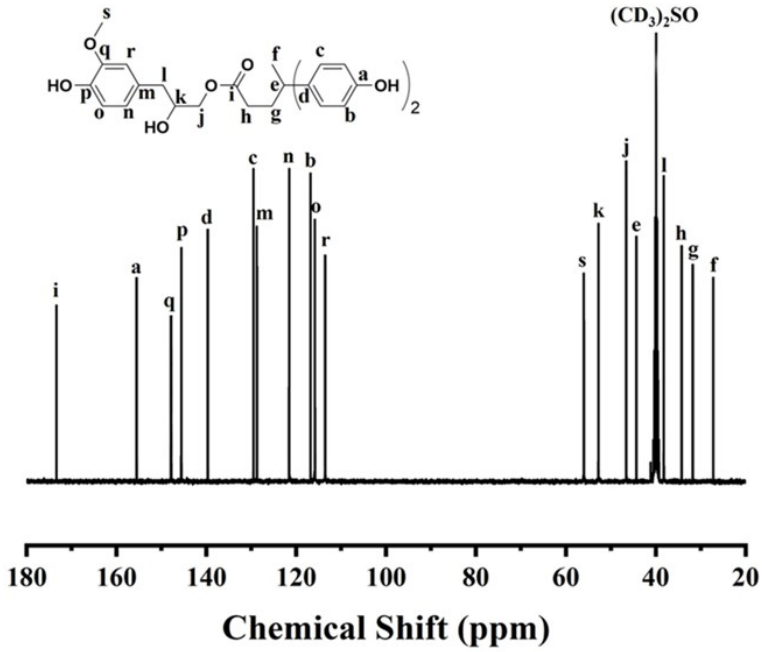

Figure S3. The ^{13}C NMR spectrum of DEE.

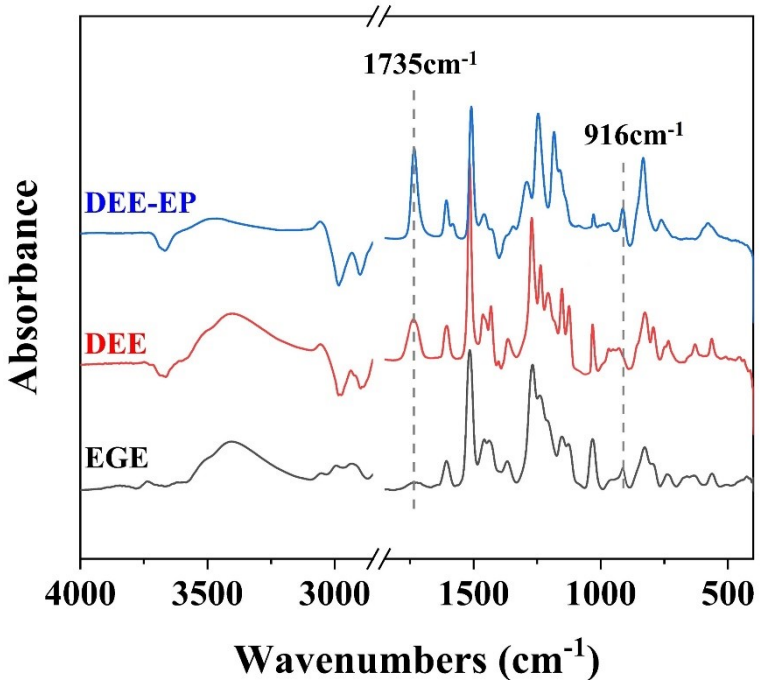

Figure S4. The FTIR spectra of EGE, DEE and DEE-EP.

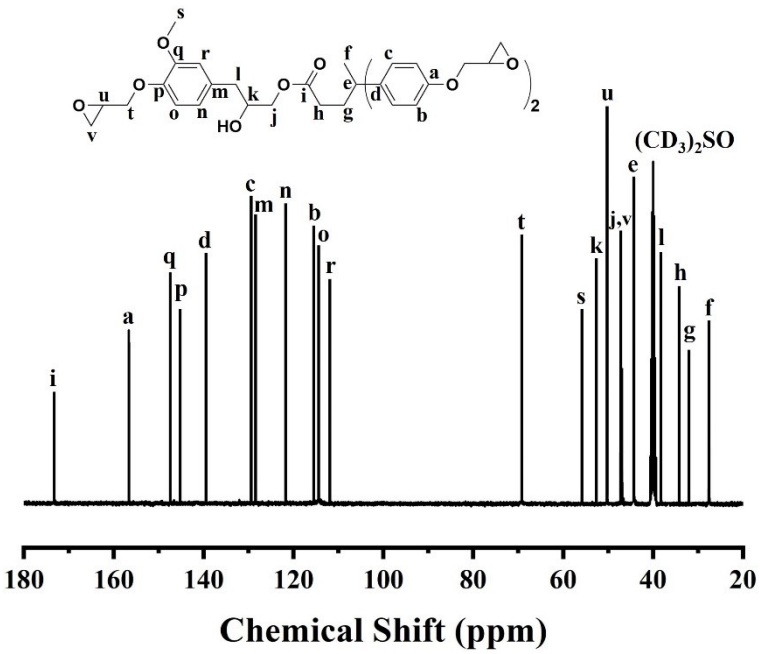

Figure S5. The ^{13}C NMR spectrum of DEE-EP.

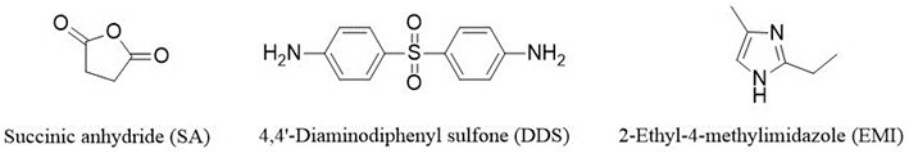

Figure S6. The hardeners used in this work.

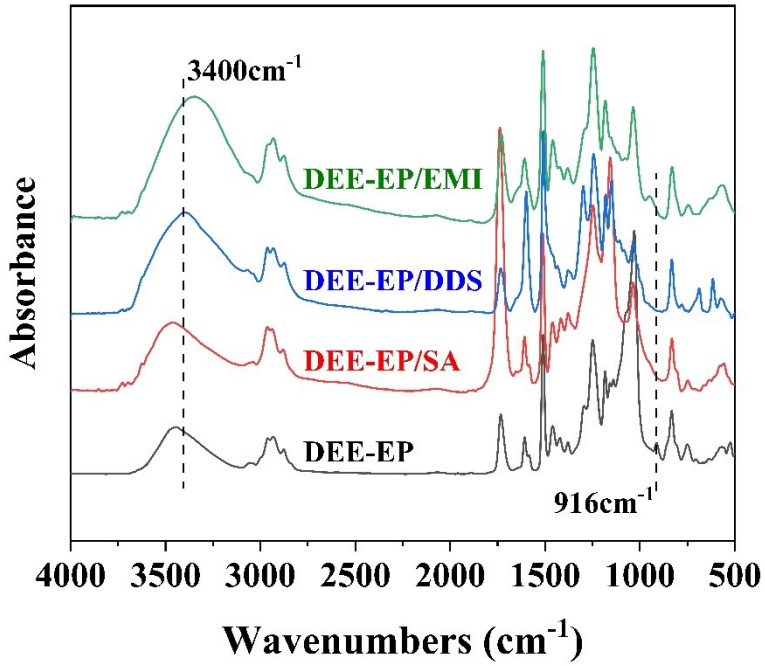

Figure S7. FTIR spectra of DEE-EP before and after curing.

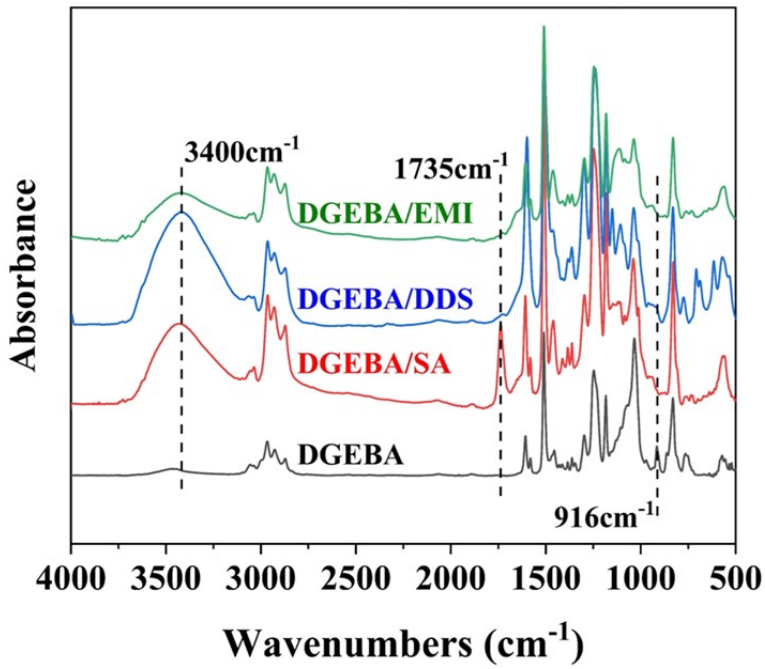

Figure S8. FTIR spectra of DGEBA before and after curing.

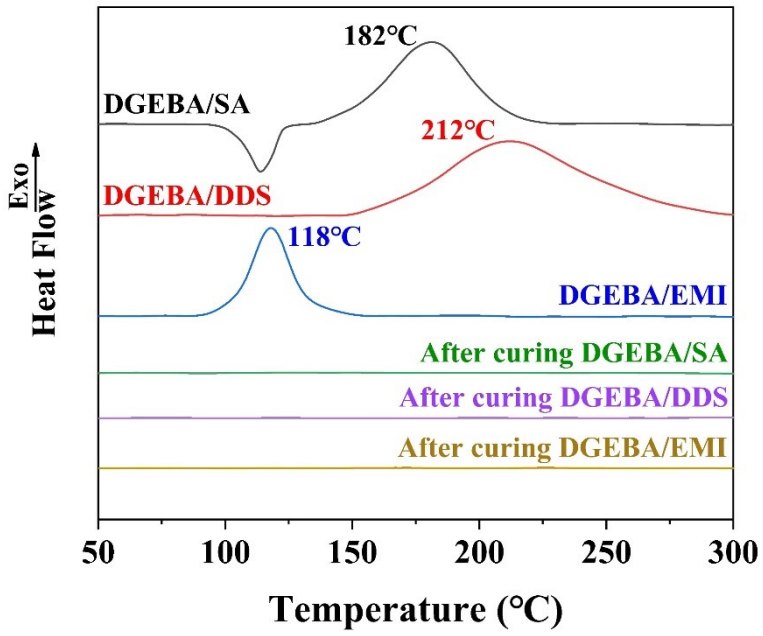

Figure S9. DSC spectra of DGEBA with different hardeners at heating rate of 10°C/min.

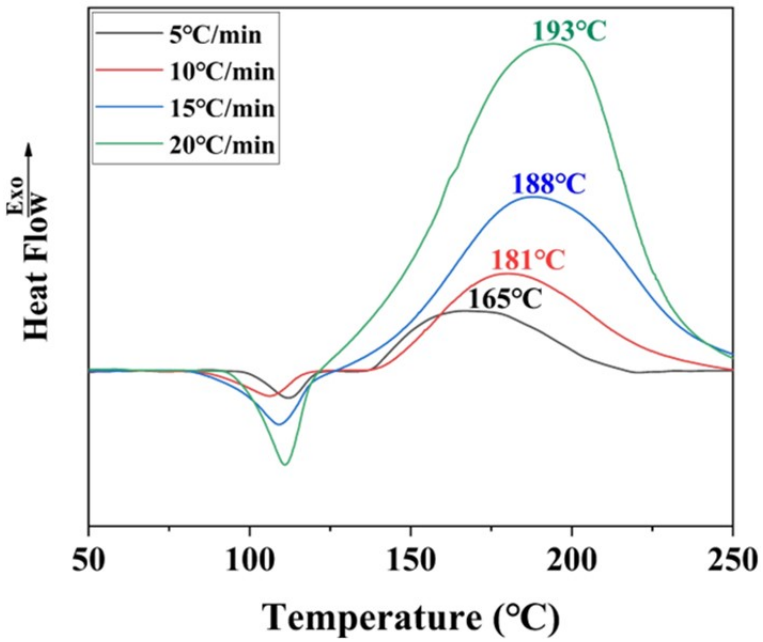

Figure S10. DSC thermograms of DEE-EP/SA mixture at different heating rates.

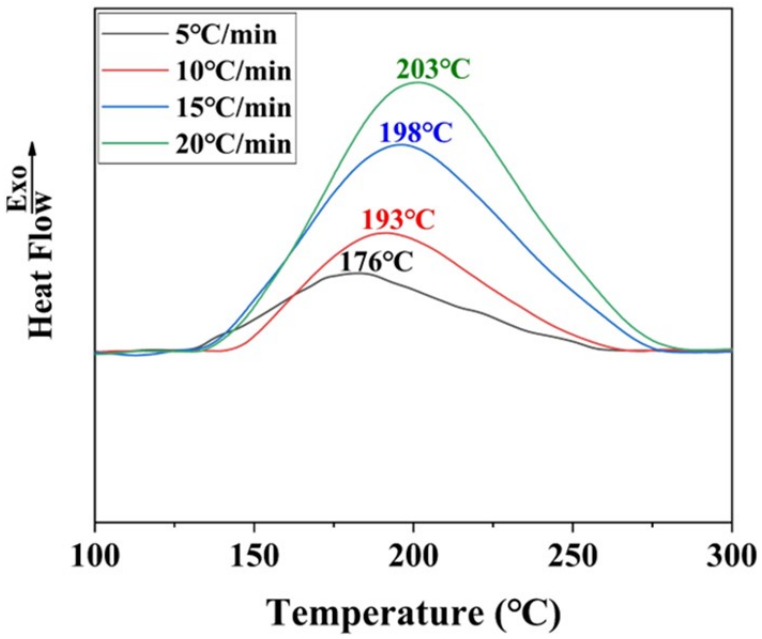

Figure S11. DSC thermograms of DEE-EP/DDS mixture at different heating rates.

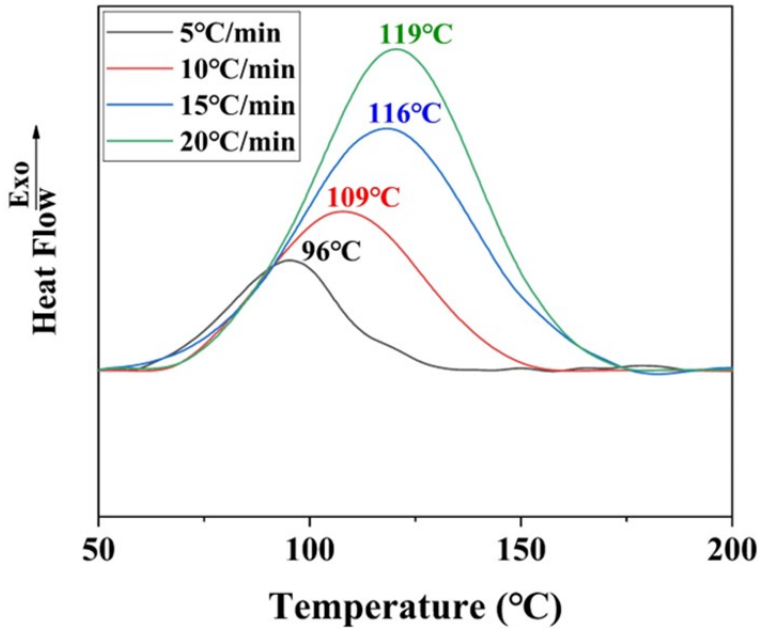

Figure S12. DSC thermograms of DEE-EP/EMI mixture at different heating rates.

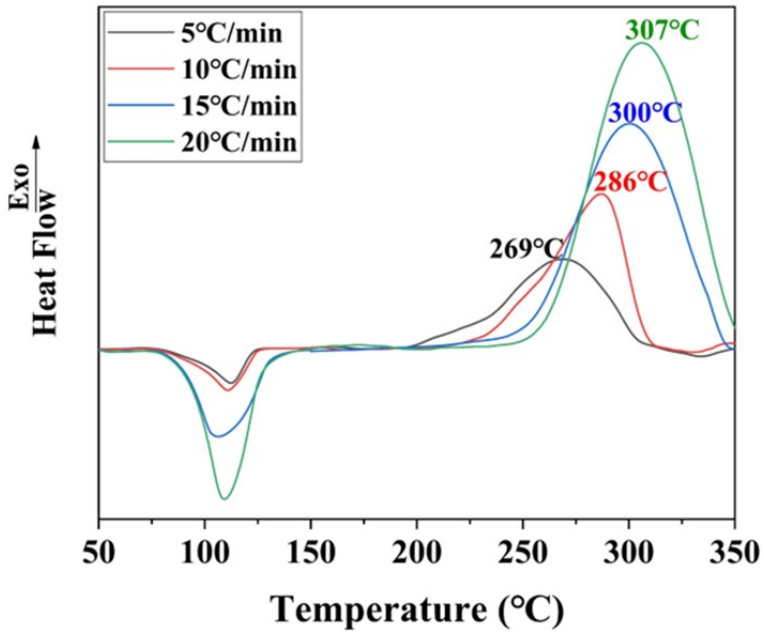

Figure S13. DSC thermograms of DGEBA/SA mixture at different heating rates.

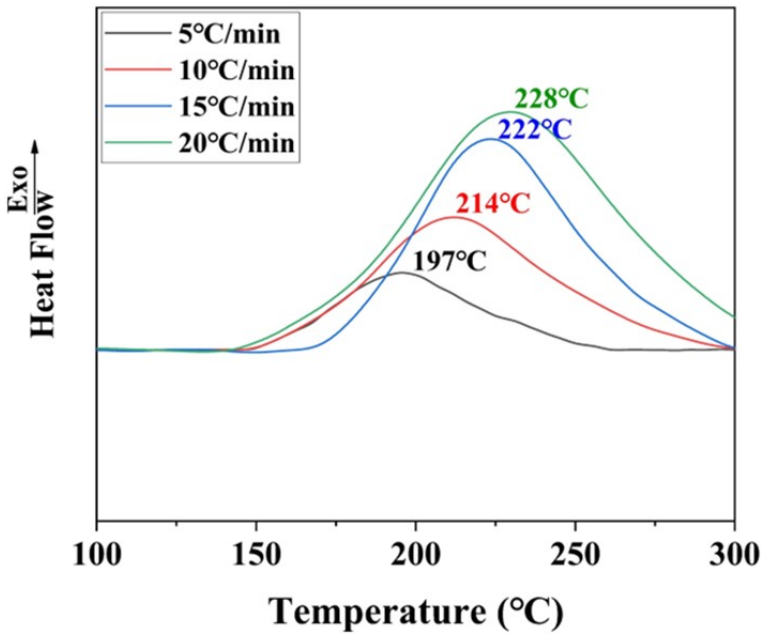

Figure S14. DSC thermograms of DGEBA/DDS mixture at different heating rates.

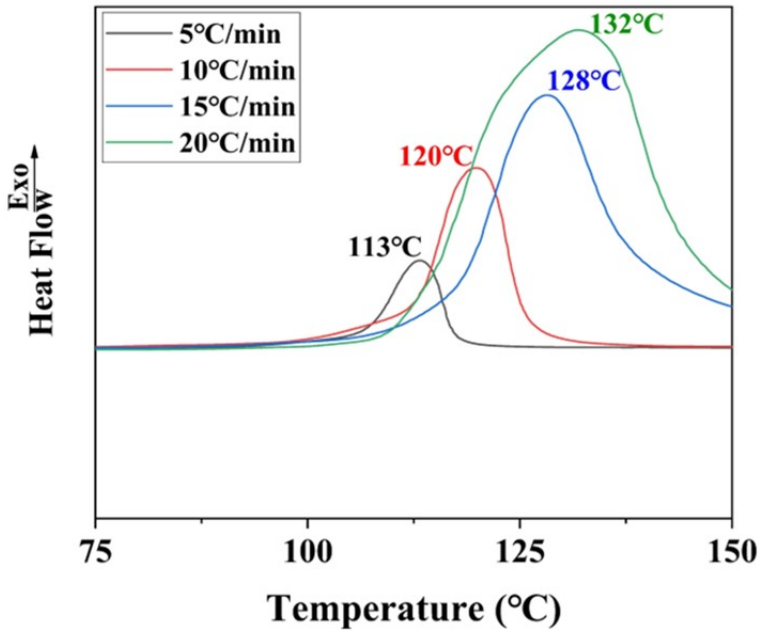

Figure S15. DSC thermograms of DGEBA/EMI mixture at different heating rates.

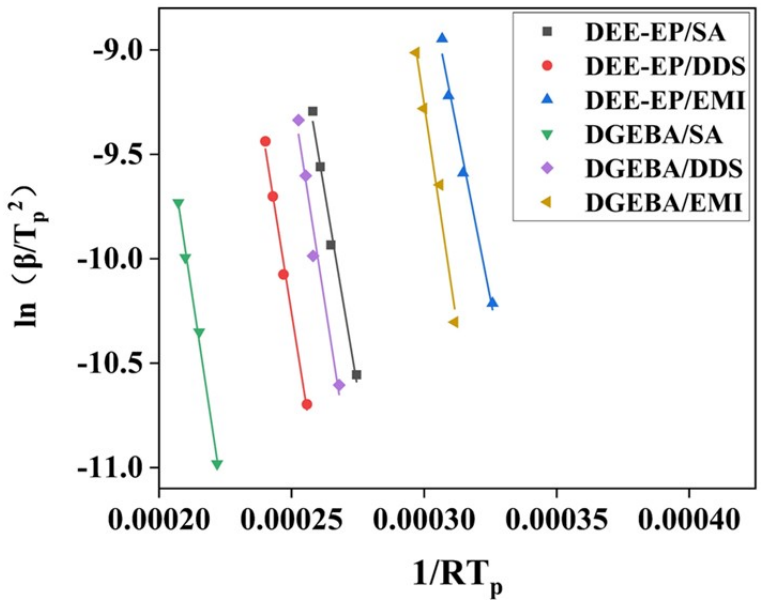

Figure S16. Plots of E_a values calculated using Kissing-Akahira-Sunose method.

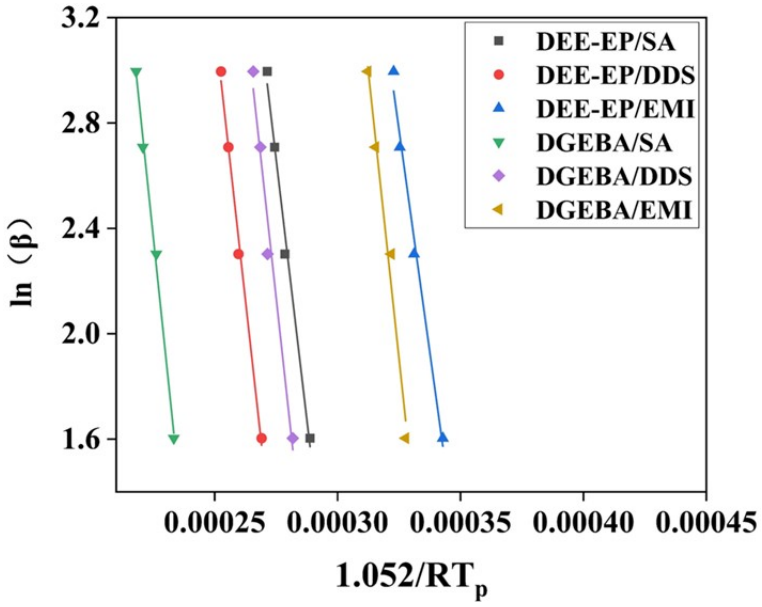

Figure S17. Plots of Ea values calculated using Flynn-Wall-Ozawa method.

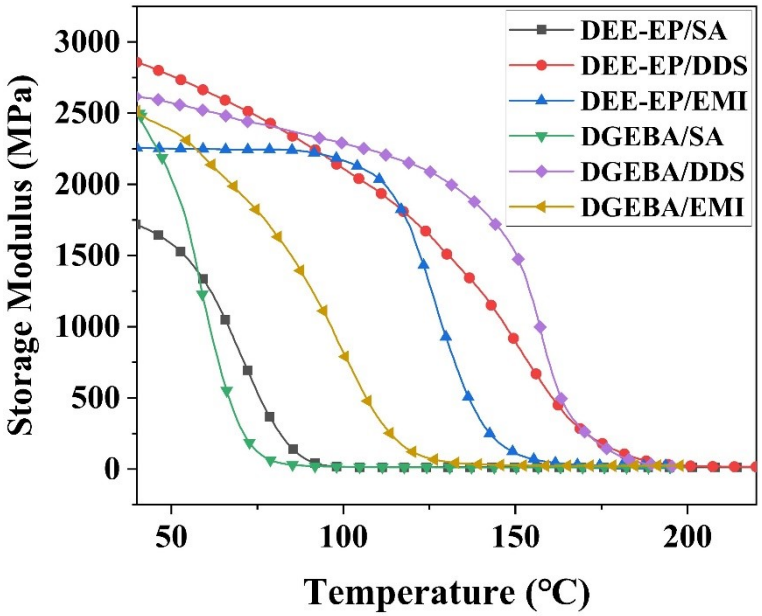

Figure S18. Storage modulus E′ of the cured resins.

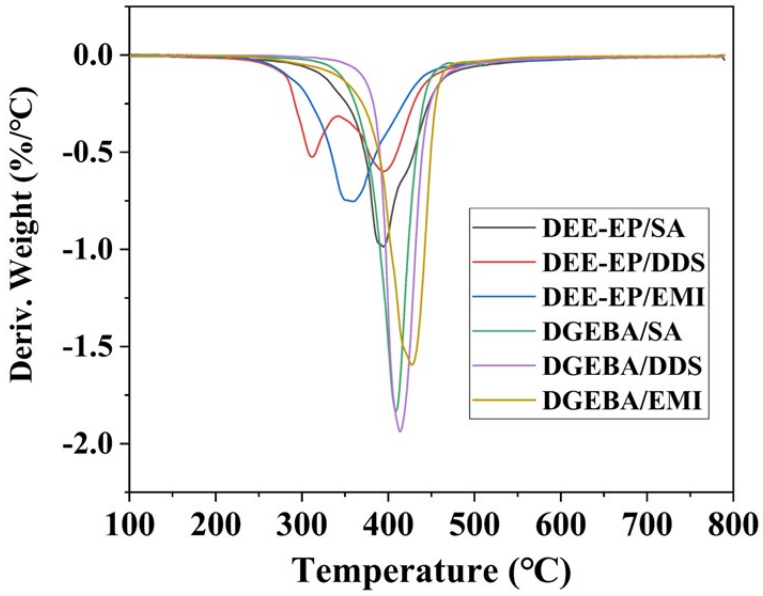

Figure S19. DTG curves of the cured resins.

a

Temperature(°C)	Thermal degradation products
320	(27.5%) (72.5%)
360	(9.1%) (70.7%) (4.5%) (9.4%) (6.3%)
400	(1.7%) (77.3%) (2.6%) (6%) (9.7%) (2.7%)
440	(2.1%) (62.2%) (1%) (1%) (19.1%) (2.4%) (2.3%) (3.9%) (6%)
480	(3.2%) (3.9%) (43.1%) (0.8%) (0.3%) (23.6%) (7.3%) (1.9%) (6.3%) (9.6%)

b

Temperature(°C)	Thermal degradation products
320	Not available
360	(100%)
400	(9.8%) (66.7%) (2.2%) (1.7%) (1%) (5.8%) (2.9%) (3.6%) (6.3%)
440	(0.8%) (66.8%) (1.1%) (1.3%) (1.2%) (8.2%) (1.9%) (11.2%) (7.6%)
480	(2.9%) (69.2%) (1%) (1.1%) (1.9%) (14%) (3.3%) (6.6%)

Figure S20. The thermal degradation products detected by the TGA-GC/MS coupling system from the cured (**a**) DEE-EP/SA and (**b**) DGEBA/SA resins at a heating rate of 10 °C/min.

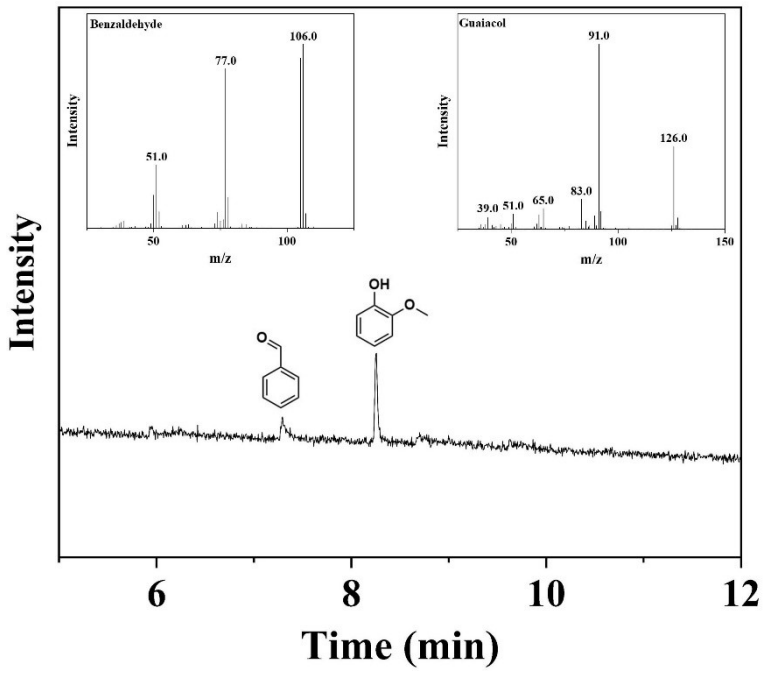

Figure S21. GC spectrum of the cured DEE-EP/SA resin decomposed at 320°C.

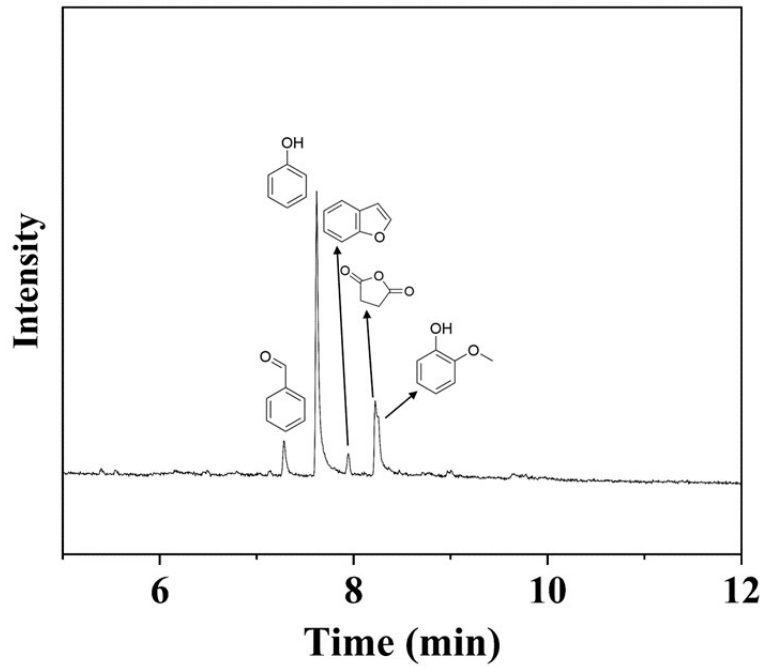

Figure S22. GC spectrum of the cured DEE-EP/SA resin decomposed at 360°C.

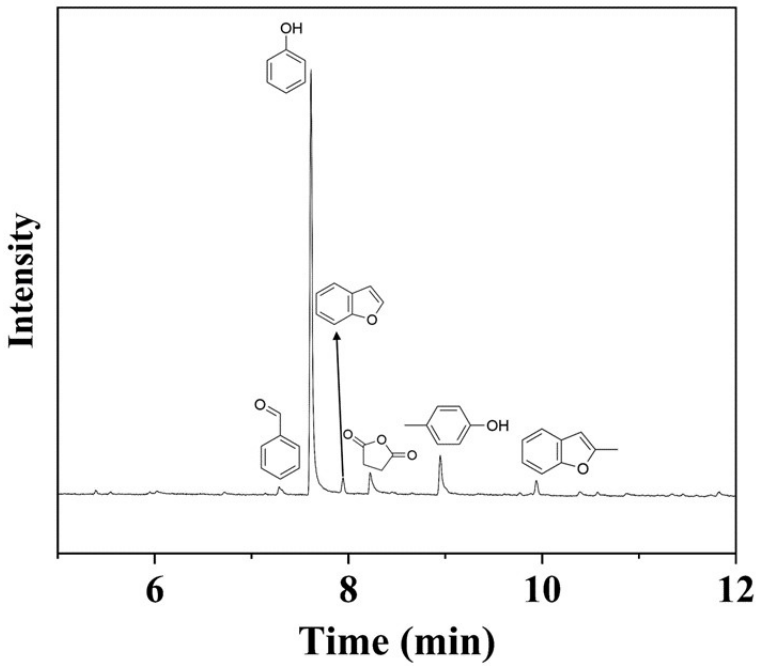

Figure S23. GC spectrum of the cured DEE-EP/SA resin decomposed at 400°C.

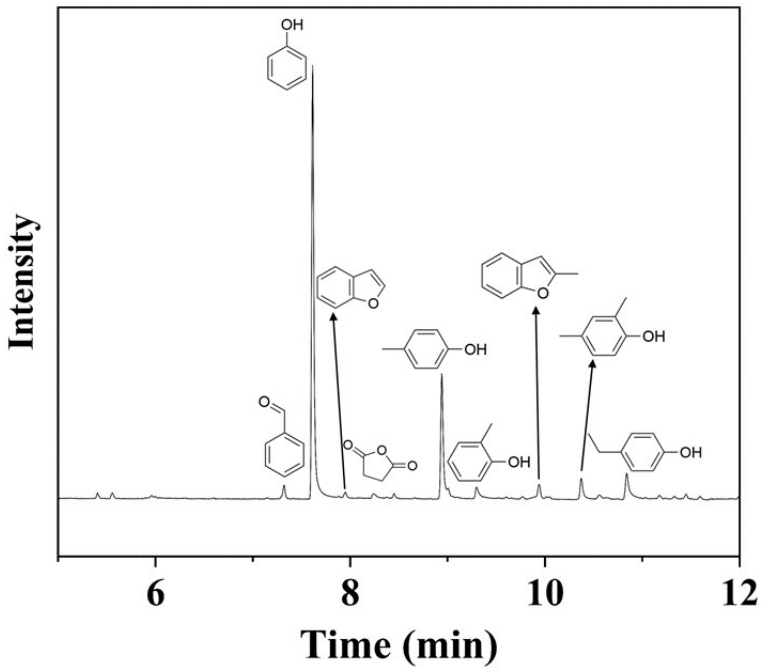

Figure S24. GC spectrum of the cured DEE-EP/SA resin decomposed at 440°C.

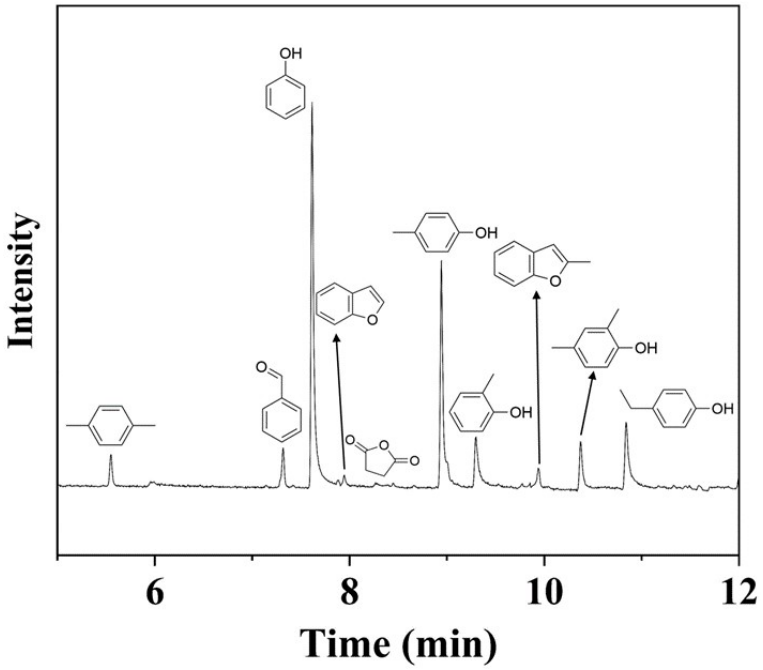

Figure S25. GC spectrum of the cured DEE-EP/SA resin decomposed at 480°C.

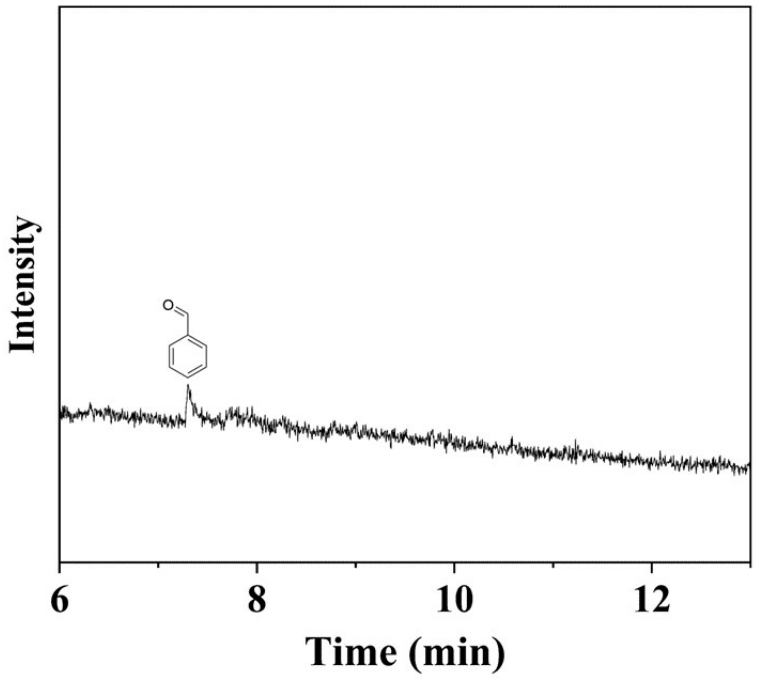

Figure S26. GC spectrum of the cured DGEBA/SA resin decomposed at 360°C.

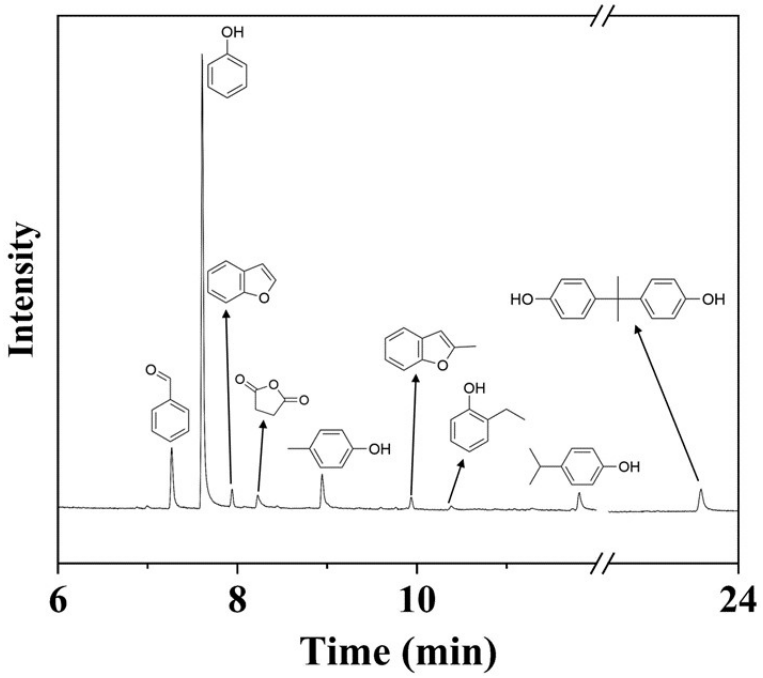

Figure S27. GC spectrum of the cured DGEBA/SA resin decomposed at 400°C.

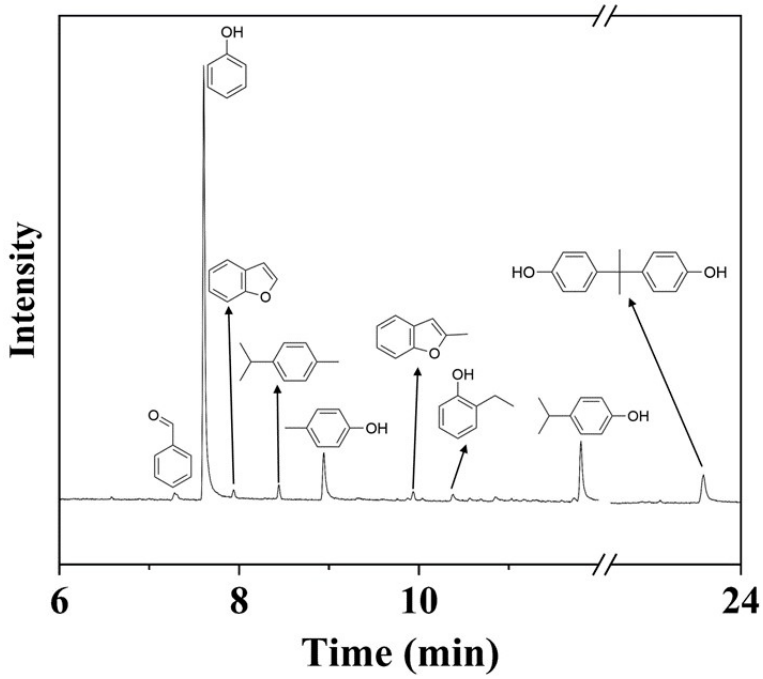

Figure S28. GC spectrum of the cured DGEBA/SA resin decomposed at 440°C.

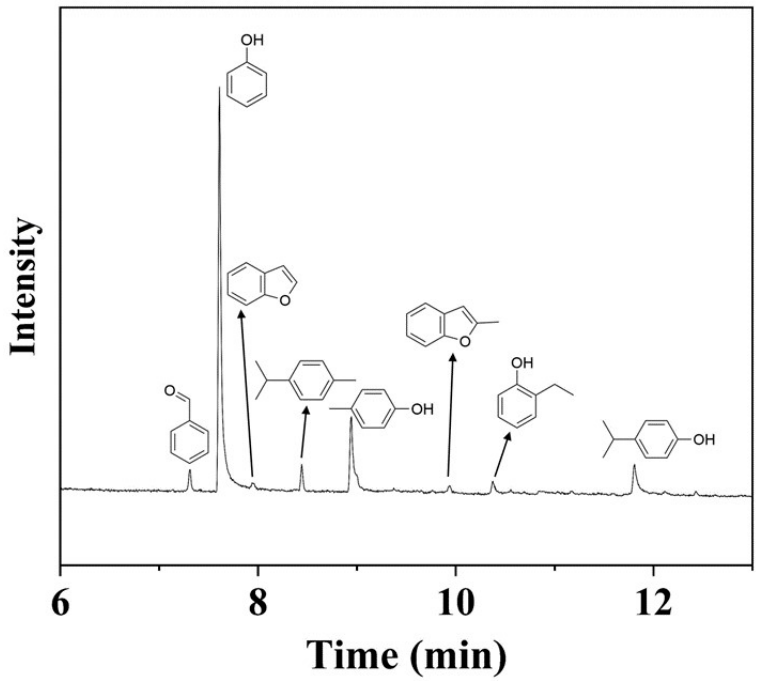

Figure S29. GC spectrum of the cured DGEBA/SA resin decomposed at 480°C.

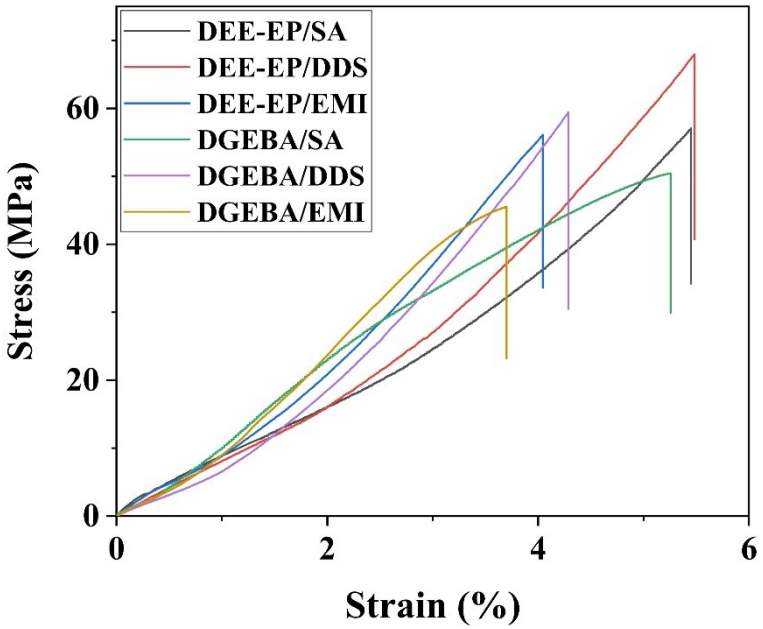

Figure S30. Stress–strain curves of the cured resins.

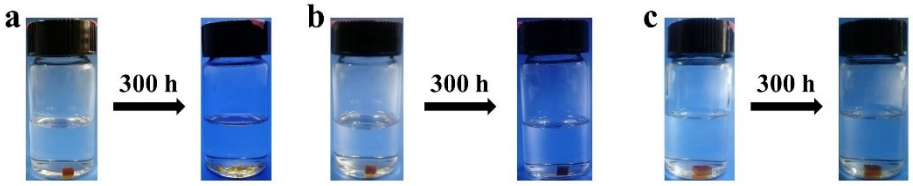

Figure S31. DGEBA/SA resin (**a**) and DEE-EP/DDS resin (**b**) and DEE-EP/EMI resin (**c**) degradation testing.

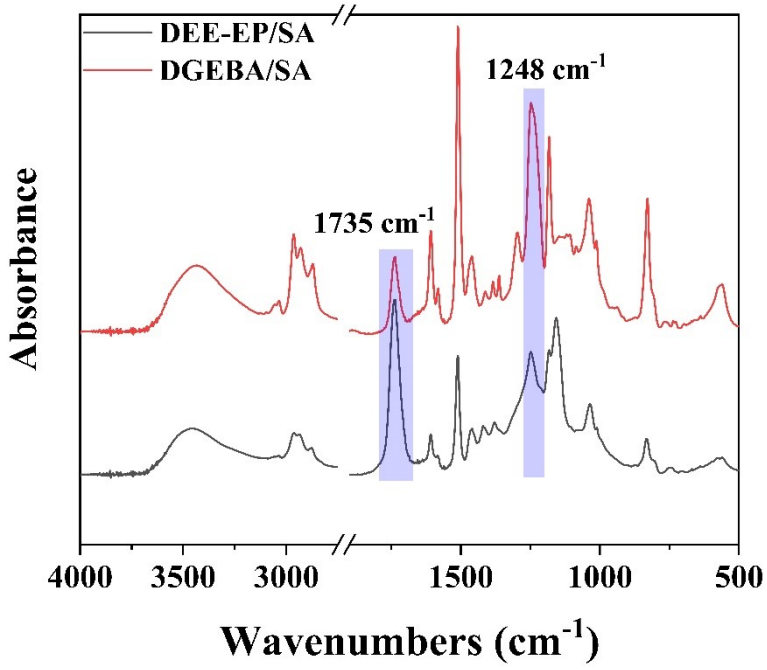

Figure S32. FTIR spectra of DEE-EP and DGEBA cured with SA.

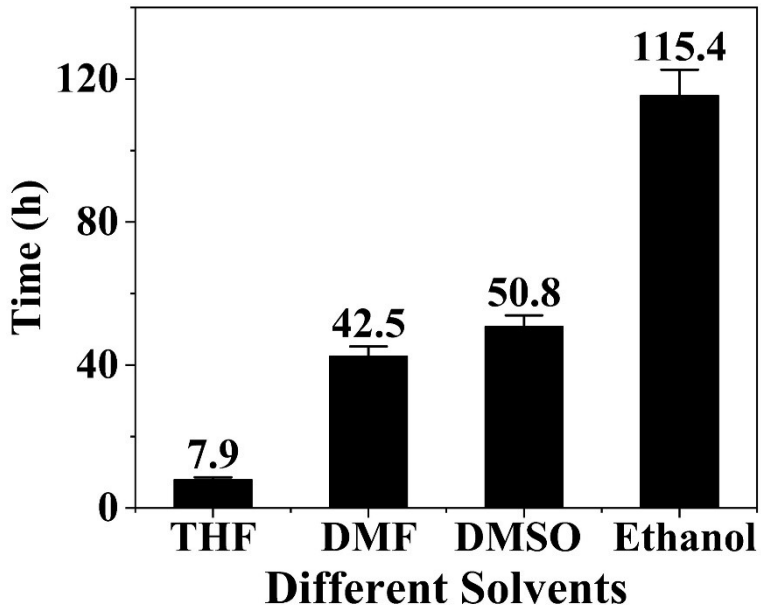

Figure S33. Degradation time of DEE-EP/SA in different organic solvents.

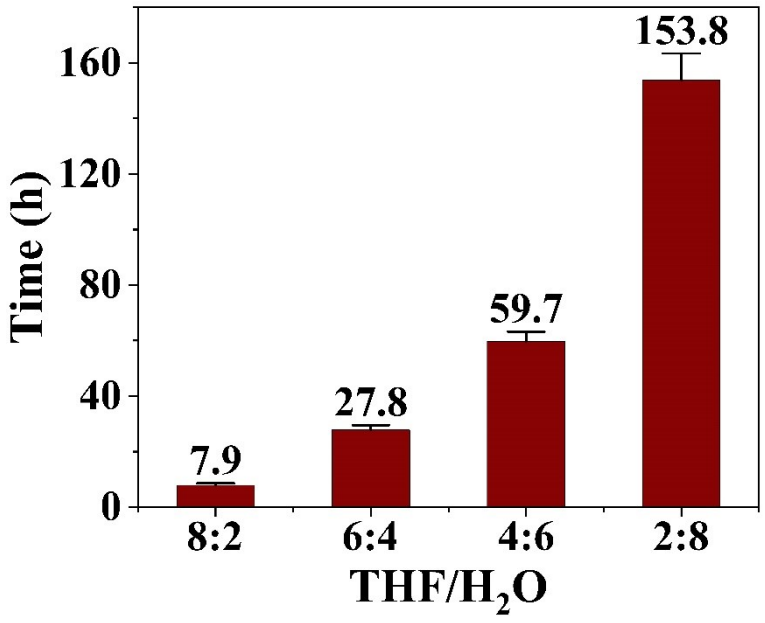

Figure S34. Degradation time of DEE-EP/SA in different ratios of organic solvents and water.

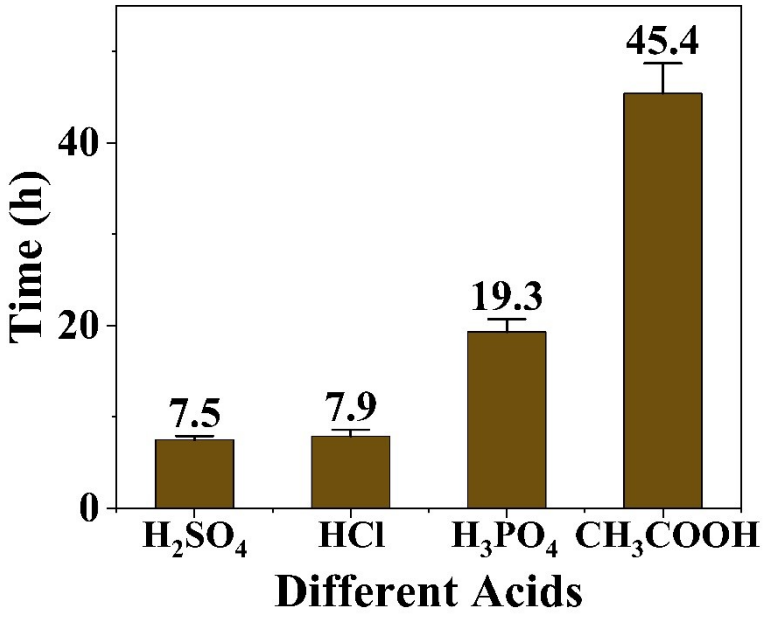

Figure S35. Degradation time of DEE-EP/SA in different types of acids.

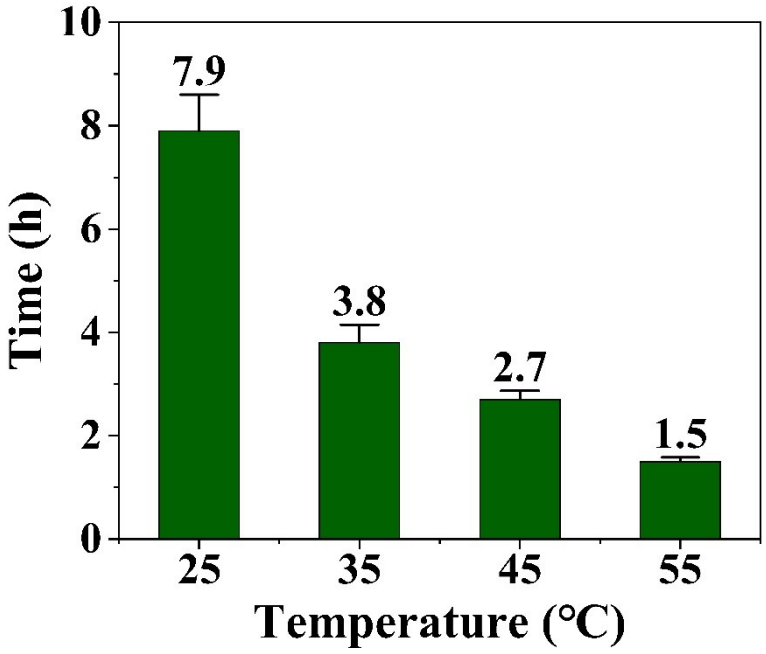

Figure S36. Degradation time of DEE-EP/SA in different temperatures.

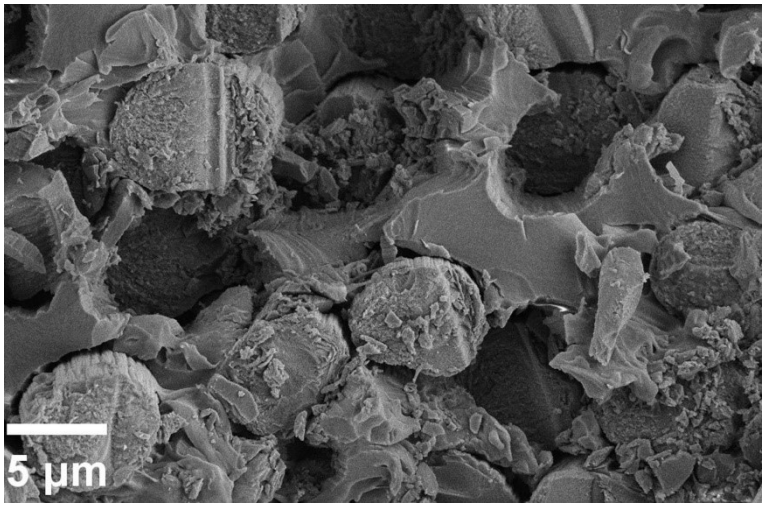

Figure S37. Fracture surface of DEE-EP/SA/CF composites.

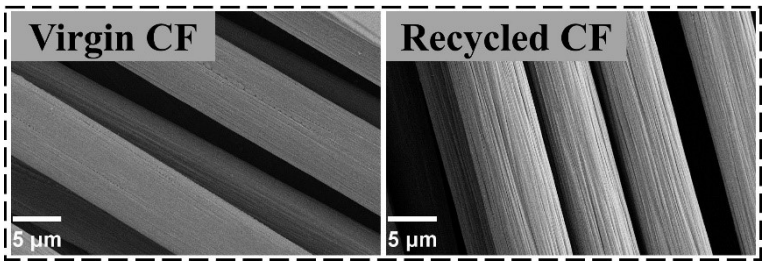

Figure S38. SEM images of original and recycled CF.

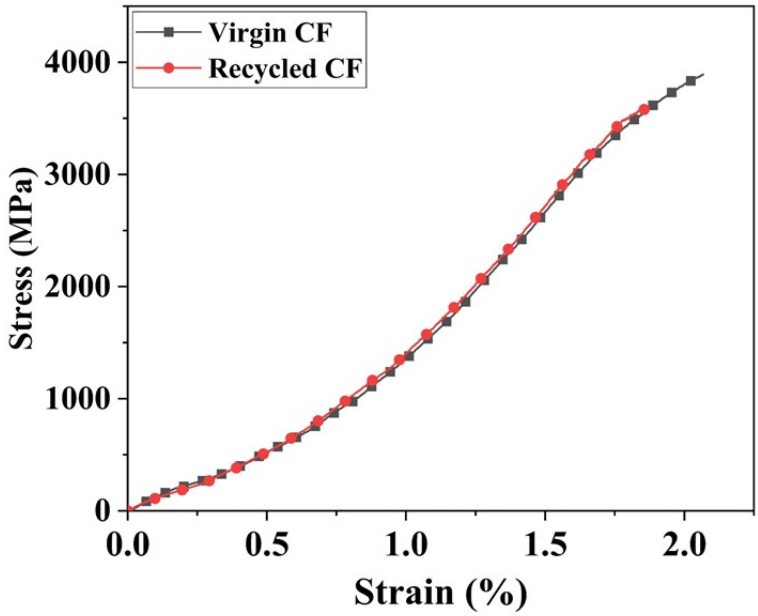

Figure S39. Stress-strain curves of the original and recovered CFs.