*Electronic Supplementary Information for*

# High biomass content, anti-flammable and degradable epoxy thermosets by curing a tyramine-derived epoxy monomer with a furan-derived diamine for non-destructively recyclable carbon fiber composite application

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## Characterization

The proton and carbon nuclear magnetic resonance (NMR) spectra were performed using an AVANCE III 400 NMR spectrometer (Bruker, Germany). The samples were dissolved in deuterated dimethyl sulfoxide prior to measurements.

The flight mass spectrometer was determined by a matrix-assisted laser desorption time-of-flight mass spectrometer (Bruck Company) and the test mode was ESI mode.

Dynamic mechanical analysis (DMA) was conducted using a TA instrument (DMA Q800, America) at a heating rate of 5°C $min^{-1}$ (from 25 to 200°C). The dimension of the cured samples for measurement was 55 mm× 10 mm× 3.0 mm.

The tensile property was measured by an electromechanical universal testing

machine (MTS criterion 43, USA) at a crosshead speed of 2 mm/min. The average value from at least 5 repetitive measurements was reported.

Charpy impact test of the non-notch samples was carried out on a ZBC1400-A pendulum impact instrument (MTS, China). According to GB/T1043.1-2008, the impact energy was 4J, and the sample size was 80 mm × 10 mm × 4 mm.

Thermal decomposition behavior of the samples was evaluated using a Q5000 Thermogravimetric analyzer (TGA) (TA Instruments, USA) in $N_2$ and air. The sample (approximately 5.0 mg) was heated from 50 to 800 °C with a ramp rate of 20 °C/min.

The anti-flammability of the samples was studied using a UL-94 vertical burning chamber following ASTM D3801–10, and the dimension of the samples was 125 mm× 13 mm × 3 mm.

Limiting oxygen index (LOI) measurement was carried out on an HC-2 LOI apparatus (Jiangning, China) following ASTM D2863-97, and the dimension of the samples was 100 mm × 6.5 mm × 3 mm.

Cone calorimeter measurement was performed on a TESTECH cone calorimeter (Suzhou, China) under 50 kW/m$^2$ following ISO5660-1, and the dimension of the samples was 100 mm × 100 mm × 3 mm.

The microstructure of the char residues and carbon fibers were studied using an LabRAM-HR Confocal Raman Microprobe with a 532 nm argon-ion laser.

The elemental composition of the char residues was obtained by an ESCALAB 250Xi X-ray photoelectron spectrometer (XPS) (Thermo Fisher Scientific, USA).

The morphology of the char residues and carbon fibers were observed using

SU8200 field emission scanning electron microscopy (SEM) (Hitachi, Japan) at an accelerating voltage of 3kV.

**Table S1.** DMA data of DGEBA/DDM and TVEP/DFDA

| Sample | $T_g$ (℃) | E' (30 ℃) (MPa) | G' (30 ℃) (MPa) | E' ($T_g$+30 ℃) (MPa) | $\upsilon_e$ (mol/m$^3$) |
|---|---|---|---|---|---|
| DGEBA/DDM | 158.4 | 2100.1 | 22.6 | 17.0 | $1.47 \times 10^3$ |
| TVEP/DFDA | 162.7 | 2776.2 | 55.5 | 26.2 | $2.25 \times 10^3$ |

**Table S2.** TGA data of DGEBA/DDM and TVEP/DFDA in $N_2$

| Sample | $T_{5\%}$ (℃) | $T_{max}$ (℃) | V (wt.%/℃) | Char residue (%) at 800℃ |
|---|---|---|---|---|
| DGEBA/DDM | 375.3 | 385.3 | 1.80 | 14.2 |
| TVEP/DFDA | 275.4 | 307.0 | 0.64 | 30.6 |

**Table S3.** TGA data of DGEBA/DDM and TVEP/DFDA in air

| Sample | $T_{5\%}$ (℃) | $T_{max1}$ (℃) | $V_1$ (wt.%/℃) | $T_{max2}$ (℃) | $V_2$ (wt.%/℃) | Char residue (%) at 800℃ |
|---|---|---|---|---|---|---|
| DGEBA/DDM | 373.3 | 383.8 | 1.82 | 570.0 | 0.43 | 2.5 |
| TVEP/DFDA | 277.0 | 303.3 | 0.55 | 546.4 | 0.44 | 2.0 |

**Table S4.** The UL-94 and LOI data of DGEBA/DDM and TVEP/DFDA

| Sample | LOI (%) | UL-94 | | |
|---|---|---|---|---|
| | | $t_1+t_2$ | Dripping | Rating |
| DGEBA/DDM | 23.5 | BC | Yes | No |
| TVEP/DFDA | 28.5 | 6+2 | No | V-0 |

**Table S5.** The UL-94 and LOI data of CF-DGEBA/DDM and CF-TVEP/DFDA

| Sample | LOI (%) | UL-94 | | |
|---|---|---|---|---|
| | | $t_1+t_2$ | Dripping | Rating |
| CF-DGEBA/DDM | 33 | BC | No | No |
| CF-TVEP/DFDA | 45 | 1+1 | No | V-0 |

**Table S6.** Comparison of several carbon fiber reinforced epoxy composites

| Sample | Epoxy monomer | Curing agent | LOI (%) | UL-94 Rating |
|---|---|---|---|---|
| CF-DGEBA/DDM | DGEBA | DDM | 33 | No |
| CF-TVEP/DFDA | TVEP | DFDA | 45 | V-0 |
| RTM6-CF[1] | RTM6 | | 33.2 | HB |
| AH-16-t-58 aliphatic composit[2] | Eporezit AH-16 | Eporezit T-58 | 29 | HB |
| D.E.R.330-D.E.H.24 aromatic composite[2] | D.E.R.330 | D.E.H.24 | 33 | HB |
| EP+CF ref[3] | Ipox MR-3012 | Ipox MH-3111 | 33 | HB |
| CFR0[4] | RTM6 | | 31 | NR |

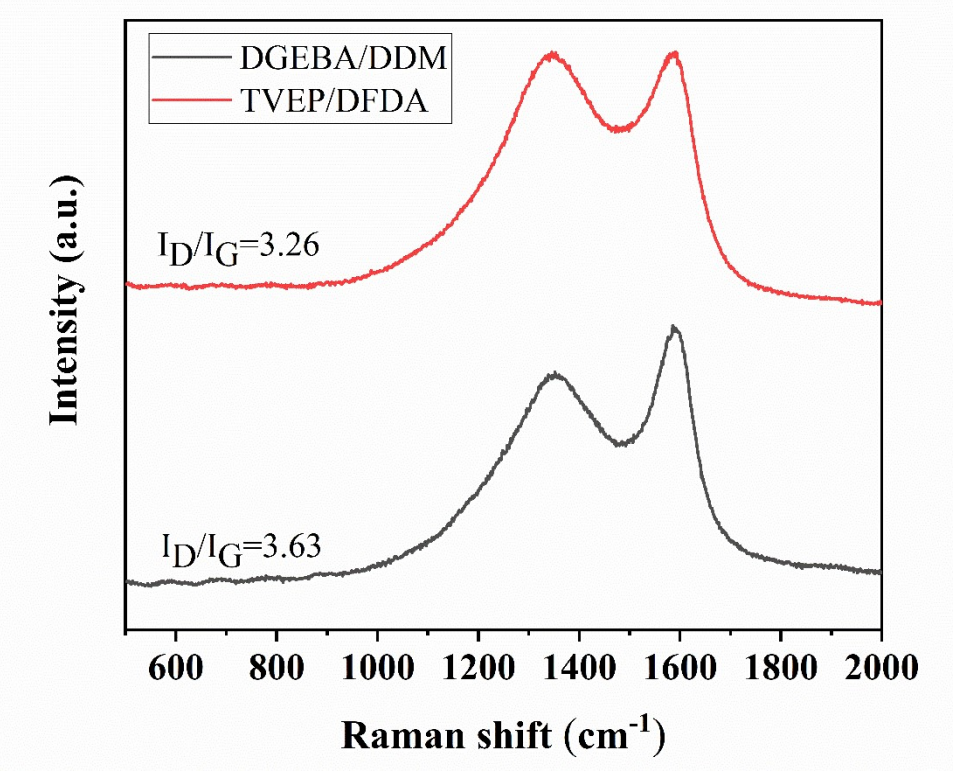


**Figure S1.** Raman spectra of the residual chars of DGEBA/DDM and TVEP/DFDA.

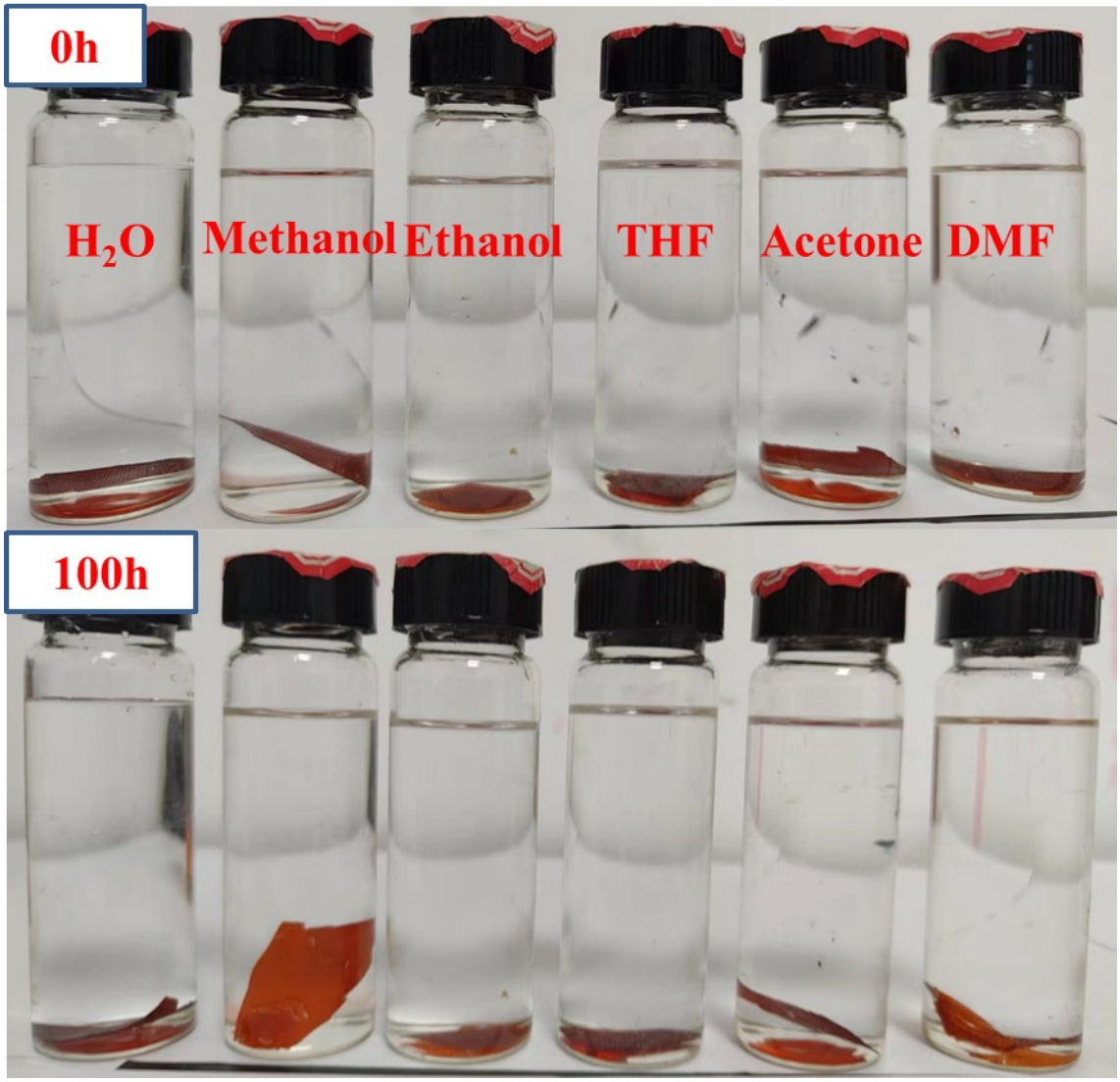


**Figure S2.** The digital photographs of TVEP/DFDA in water, methanol, ethanol, THF, acetone and DMF.

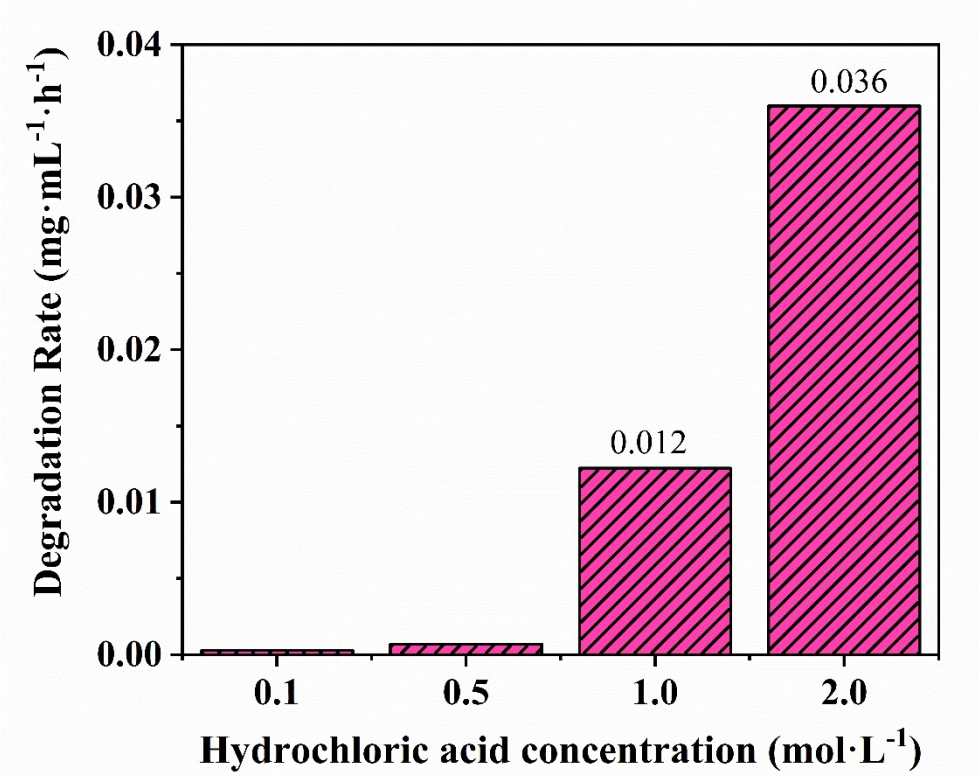


**Figure S3.** The degradation rate of TVEP/DFDA in mixed solutions of tetrahydrofuran with different hydrochloric acid concentrations.

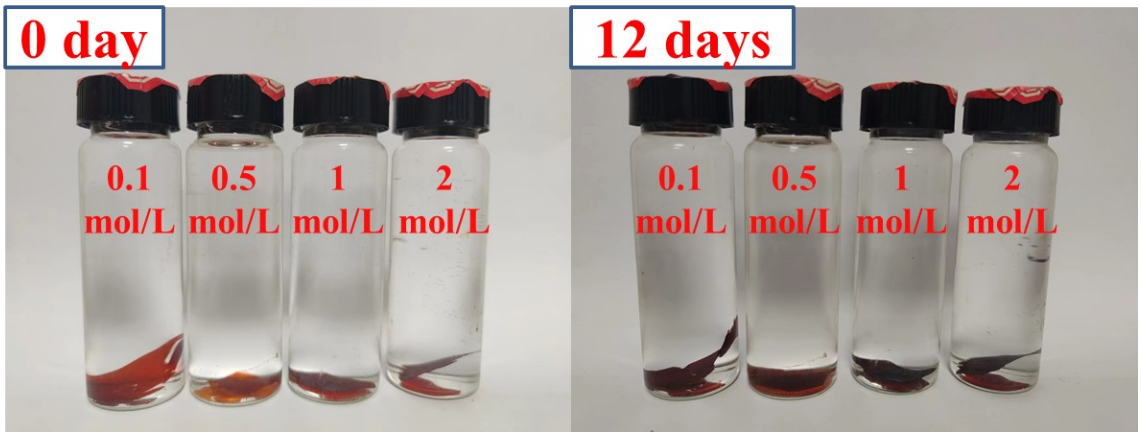


**Figure S4.** The digital photographs of TVEP/DFDA in aqueous hydrochloric acid solution at different concentrations.

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