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Supporting Information

Design of degradable, intrinsically flame-retardant and high-performance tung-oil-based epoxy vitrimers

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1. Characterizations

1. 1 Acid value test

Accurately weigh out 0.5 g of the target compound and 50 mL anhydrous ethanol, and add them to a 250 mL conical flask. Place the flask in a microwave oscillator and employ ultrasound to facilitate the dissolution of the compound. Once fully dissolved, add 2-3 drops of phenolphthalein as an indicator and perform titration using a standardized solution of 0.1000 mol•L^{-1} KOH. The solution will transition from colorless to pale pink, maintaining this hue for at least half a minute, which signifies that the endpoint has been reached. Record the volume of KOH consumed during titration, and subsequently calculate the acid value of the target compound using the appropriate formula.

In the formula:

$$X = \frac{c \times V \times 56.1}{m}$$

X——the acid value of the target compound, mg•KOH g^{-1};

c——the concentration of the KOH standard solution, mol•L^{-1};

V——the volume of KOH standard solution consumed by the target compound, L;

m——the mass of the target compound, g;

1.2 Fourier Transform Infrared Spectroscopy (FTIR)

Fourier transform infrared spectroscopy (Nicolet IS10, Thermo Fisher, USA) was employed to analyze the distribution of functional groups in the target compound. The scanning wavenumber range extended from 4000 to 400 cm^{-1}, with transmittance as the measurement mode.

1.3 Nuclear Magnetic Resonance Spectroscopy (NMR)

The chemical structure of the target compound was elucidated using a nuclear magnetic resonance (NMR) spectrometer (Burker 400 MHz, Germany), employing deuterated chloroform ($CDCl_3$) as the solvent.

1.4 Dynamic Mechanical Analysis (DMA)

The glass transition temperature (T_g), storage modulus, loss modulus, and tan δ curves of epoxy resin were analyzed using a dynamic thermal mechanical analyzer (TA Q800, USA TA) in a double cantilever mode. The heating rate was 3 ℃·min^{-1}, the test temperature was 30-180 ℃, the frequency was 1 Hz, and the sample size of epoxy resin was 60×10×5 mm^3.

1.5 Thermogravimetric Analysis

The thermal stability of epoxy resin was tested using a thermogravimetric analyzer (TG 209F1, Netzsch, Germany). About 15 mg of sample powder was weighed, and the temperature was raised from 30 ℃ to 800 ℃ at a rate of 10 ℃·min^{-1} in nitrogen or air atmosphere.

1.6 Limiting Oxygen Index Test

The limiting oxygen index (LOI) of epoxy resin was tested using an FTT0077 oxygen index tester (UK), according to the standard of ISO 4589, with sample dimensions of 60×10×5 mm^3. The higher the LOI value, the better the flame retardancy.

1.7 Vertical Flame Propagation Test

According to the UL-94 standard, a vertical burning test is conducted, with sample dimensions of 150×10×3 mm^3. The sample is placed vertically in the test chamber, with a cotton pad placed 30 cm below the sample's lower end. The flame height is adjusted to 2-3 cm, and the single ignition time is maintained at 10 seconds. The sample's burning time and whether it drips to ignite the cotton are observed to evaluate the flammability grade of the epoxy sample. It is divided into V-0, V-1, V-2, or no grade. For more details, please refer to Table 1.

Table S1 The grade reference of UL-94

Criteria for Determination	V-0	V-1	V-2
Time for spontaneous combustion after applying flame (s)	≤10	≤30	≤30
Total self-ignition time (s) (Total burn time for	≤50	≤250	≤250

10 runs)			
Self-Ignition and incandescence time following the second application of flame (s)	≤30	≤60	≤60
The dripping substance caused ignition of the cotton.	No	No	Yes
The sample is completely burned.	No	No	No

1.8 Cone calorimeter test

The cone calorimeter (Vouch 6810 by Walch) was used to test the fire behavior of epoxy resin according to the standard ISO 5660, quantitatively evaluating the heat and smoke output of the material. The sample size was 100×100×3 mm^3, and the radiant intensity was 35 $W \cdot mm^{-1}$.

1.9 Mechanical property testing

The tensile properties of epoxy resin were tested according to the national standard GB/T1040 method using an electronic universal testing machine (CMT4000, China, Shanghai Jiehu Instrument & Meter Co., Ltd.) equipped with an electronic extensometer (25 mm) and a large deformation (rotary type). The sample size was 80×5×4 mm^3, the load was 30 kN, the tensile speed was 5 $mm \cdot min^{-1}$, and parallel tests were conducted for 5 groups. The average value was taken. Before the test, the samples were placed in a 25 ℃ test environment for 6 h.

1.10 Chemical degradation

The TO-vitrimers were degraded using an ethanol-NaOH-water system at room temperature. The mass ratio of TO-vitrimers, ethanol, NaOH, and water was 1:20:1:10.

1.11 Raman spectroscopy analysis of carbon fibers.

The Raman spectrometer (USA, Thermo Fisher, DXR2xi model) was used to observe the changes in the chemical structure of carbon fiber before and after chemical degradation. At least three laser scanning points were used, with a wavelength of 532 nm.

1.12 Scanning Electron Microscopy Analysis of Carbon Fiber

The surface morphology of carbon fibers was observed using a scanning electron microscope with an emission mode (USA, EEI Cor., Quanta 200). The accelerating voltage was 30 kV, the energy resolution was 132 eV, and the resolution was 2 nm in high vacuum mode.

2. Results and discussion

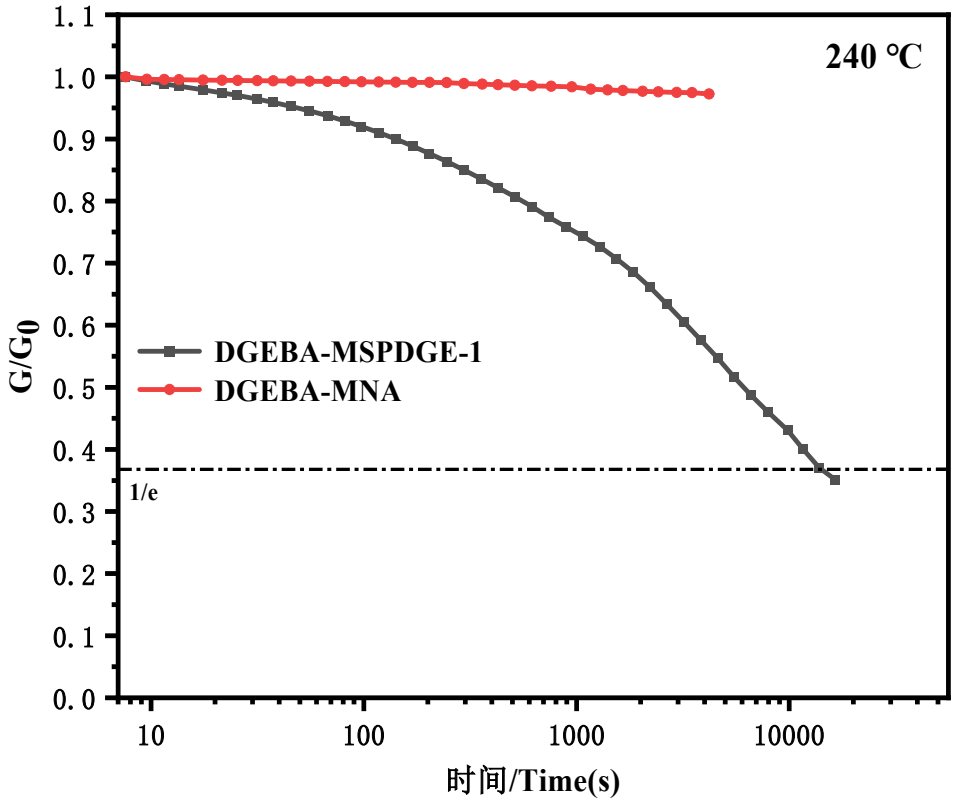

Fig. S1 Stress relaxation curves of DGEBA-MSPDGE-1 and DGEBA-MNA

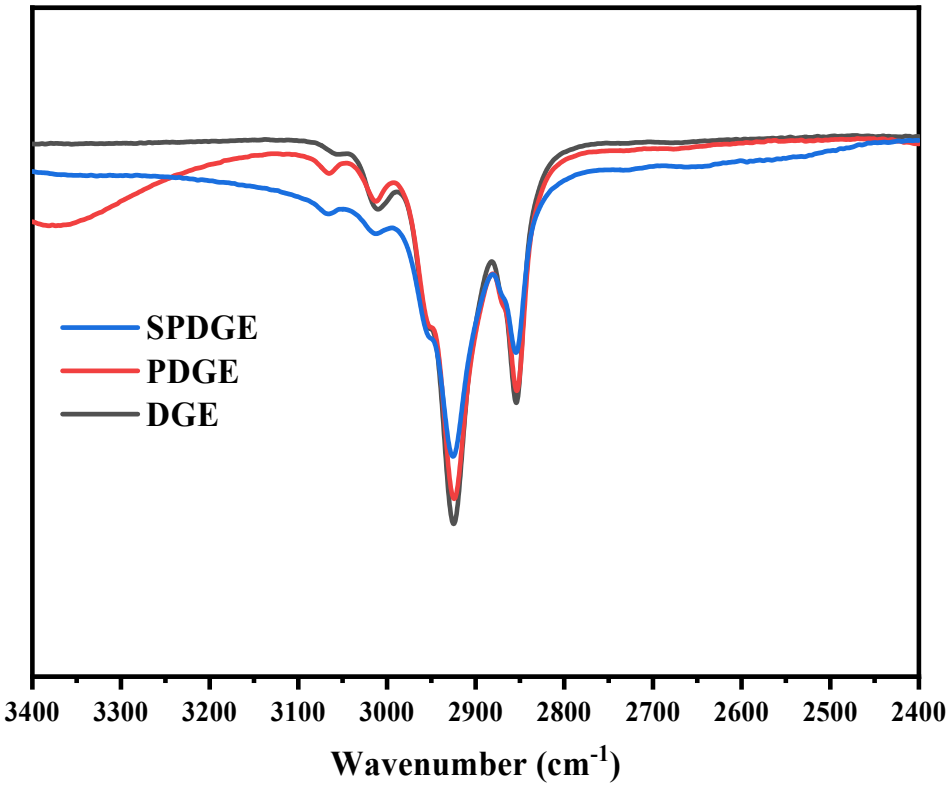

Fig. S2 FTIR spectra of DGE, PDGE and SPDGE.

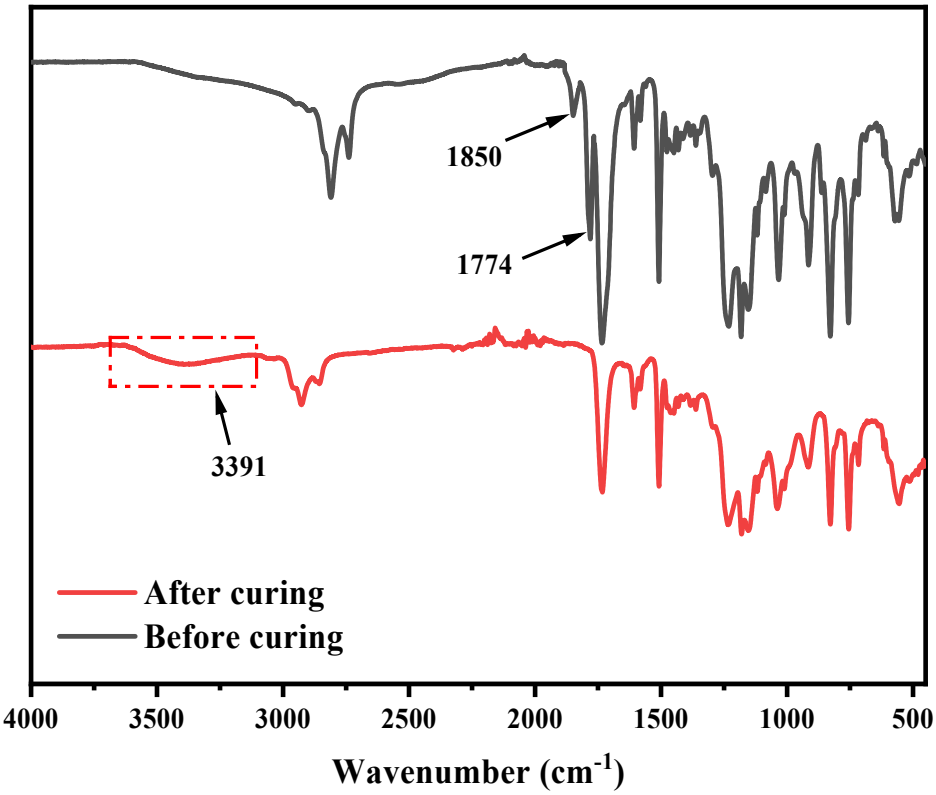

Fig. S3 FTIR spectra of DGEBA-MSPDGE-1 before and after curing

Fig. S4. The process of forming prototypical CFRCs was used in this work (50.0 wt.%).