

Chemically Degradable Vitrimers Based on Divanillin Imine Diepoxy Monomer and Aliphatic Diamines for Enhanced Carbon Fiber Composite Applications

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1. Structural characterization of the monomer

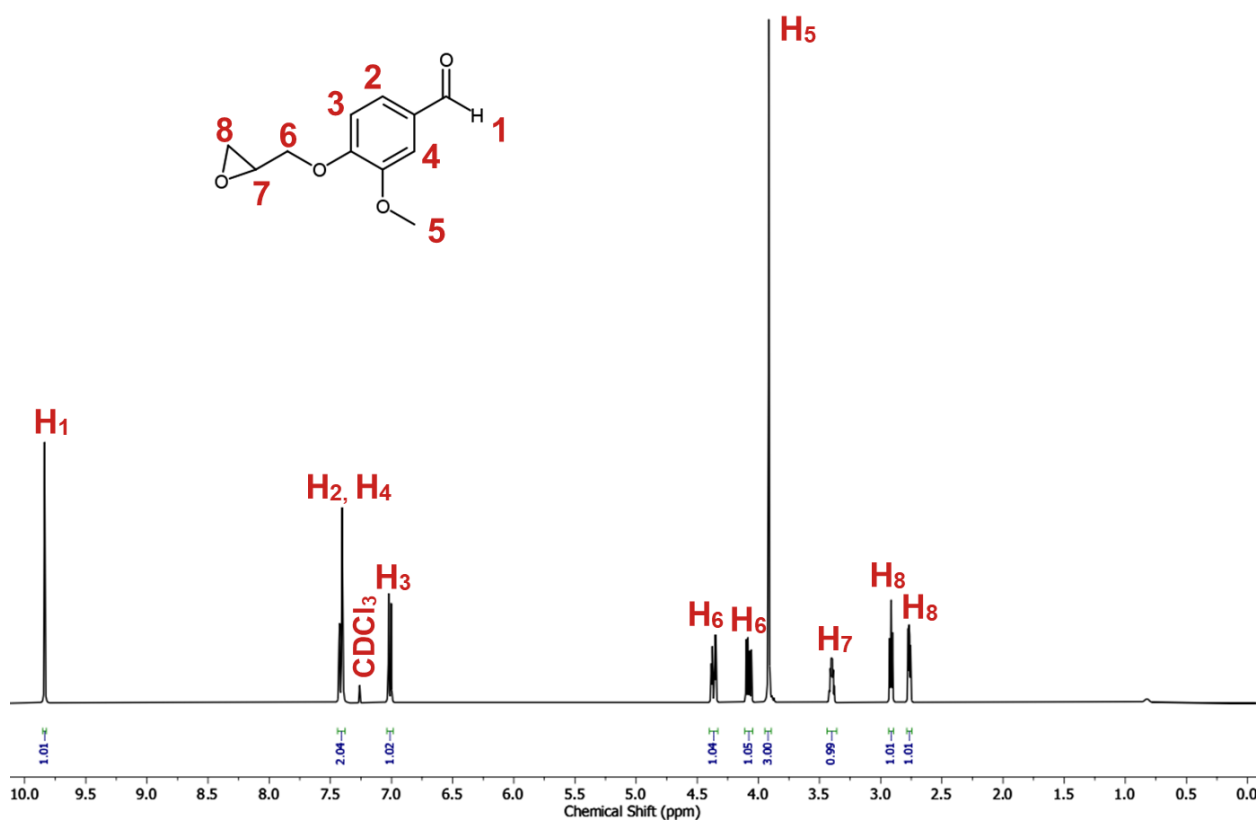


Figure S1. ^1H NMR spectrum of EVan in CDCl_3 .

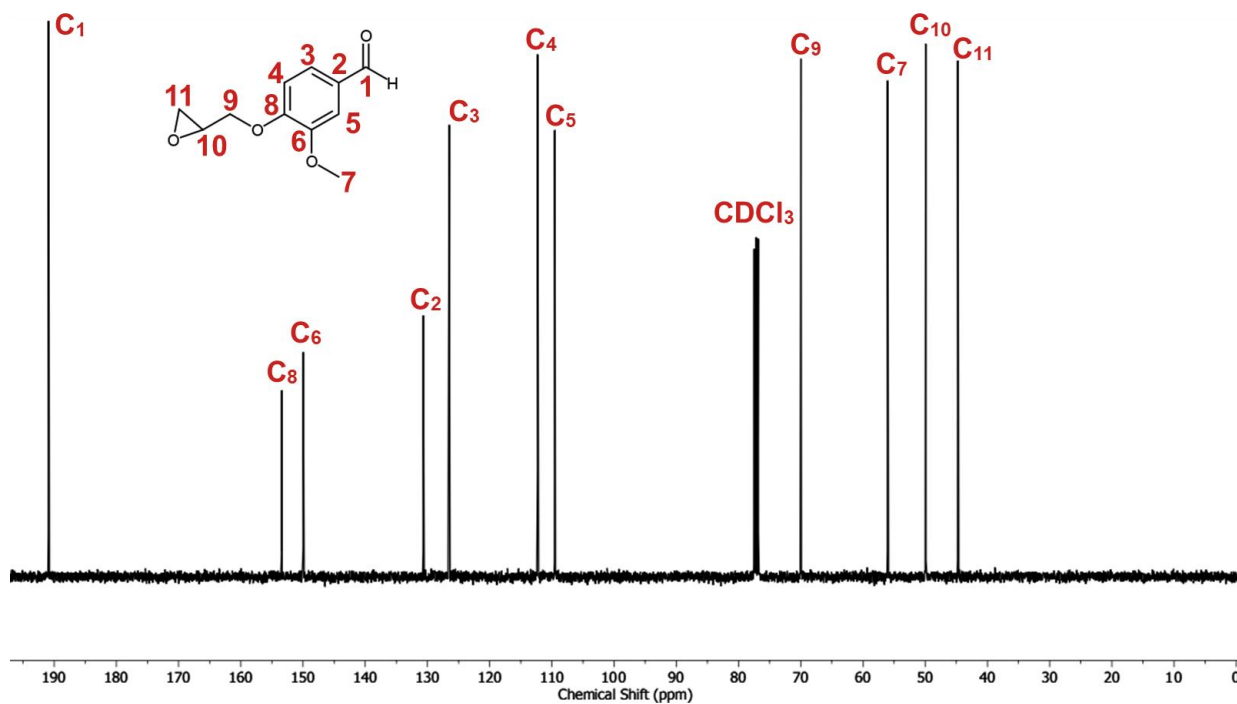


Figure S2. ^{13}C NMR spectrum of EVan in CDCl_3 .

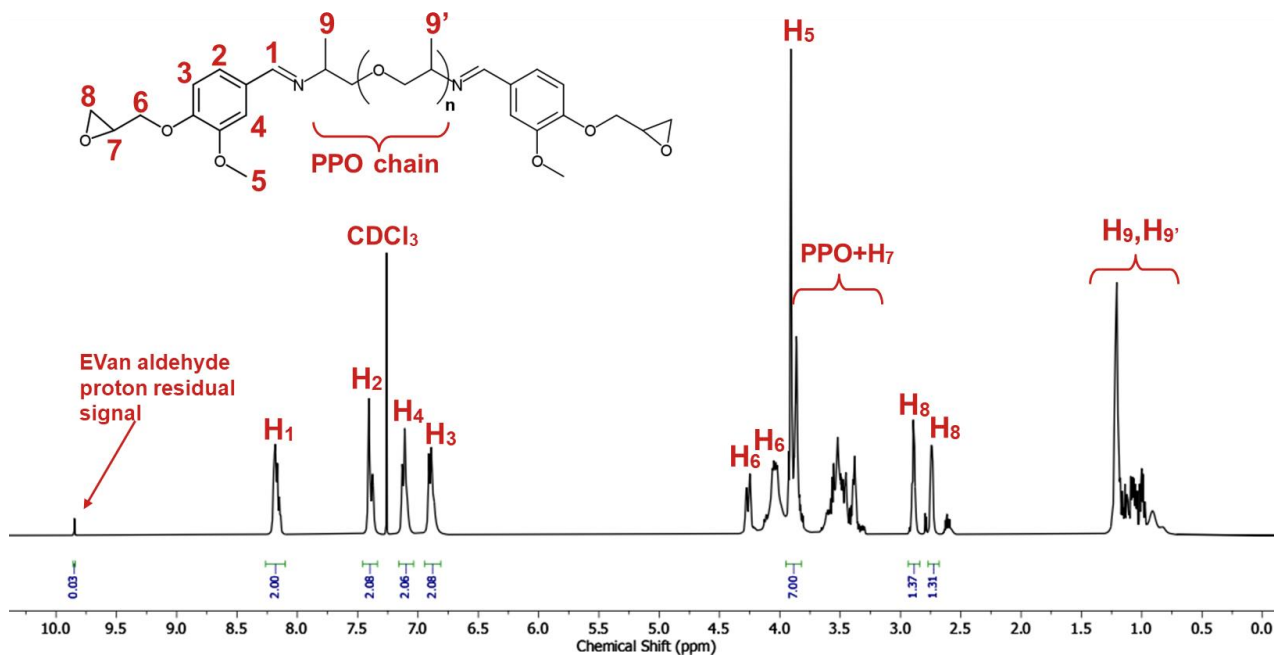


Figure S3. ^1H NMR spectrum of DIDE-PPO in CDCl_3 .

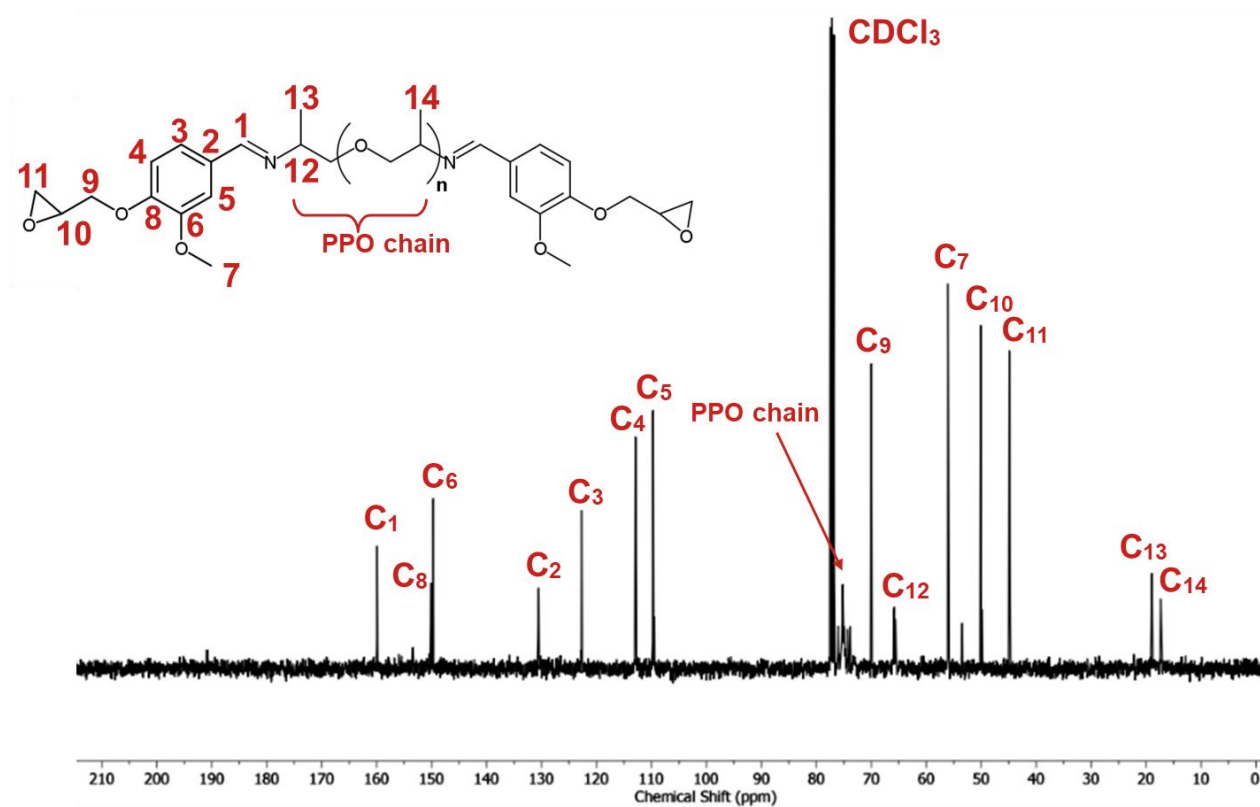


Figure S4. ^{13}C NMR spectrum of DIDE-PPO in CDCl_3 .

2. Study of the curing process by ^1H NMR spectroscopy

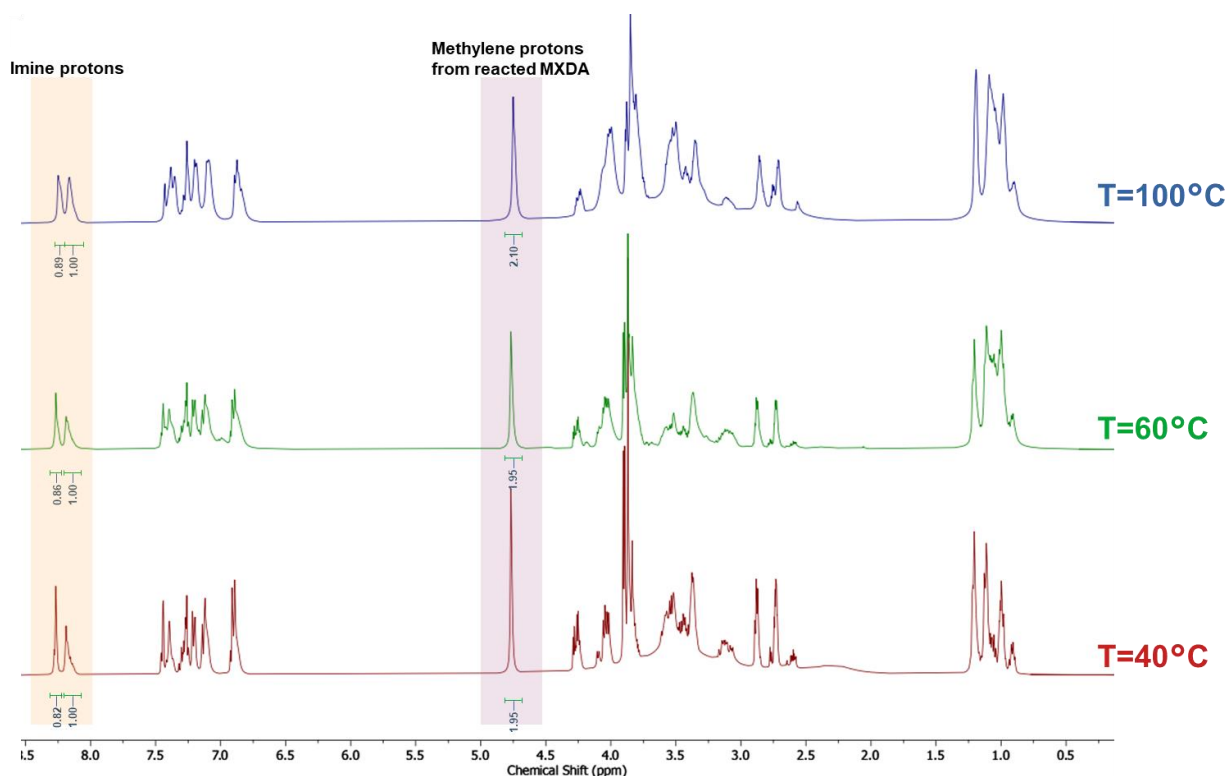


Figure S5. ^1H NMR spectra in CDCl_3 of the curing mixture at 40, 60 and 100 $^\circ\text{C}$ collected after 10 minutes of equilibration at each temperature.

Table S1. Integrals of the main peaks of the curing mixture at different temperatures.

Temperature ($^\circ\text{C}$)	$I_{8.27 \text{ ppm}}^{\text{a}}$	$I_{8.18 \text{ ppm}}^{\text{b}}$	$I_{4.77 \text{ ppm}}^{\text{c}}$	$I_{8.18 \text{ ppm}}/I_{8.27 \text{ ppm}}^{\text{d}}$
40	1.00	0.82	1.95	0.82
60	1.00	0.86	1.95	0.86
100	1.00	0.89	2.10	0.89

^a Normalized integral of the original imine proton signal.

^b Normalized integral of the MXDA-derived imine proton signal.

^c Normalized integral of the methylene protons signal relative to the reacted MXDA hardener.

^d Ratio between the integrals of the two imine signals.

3. Thermogravimetric analysis of the prepared materials

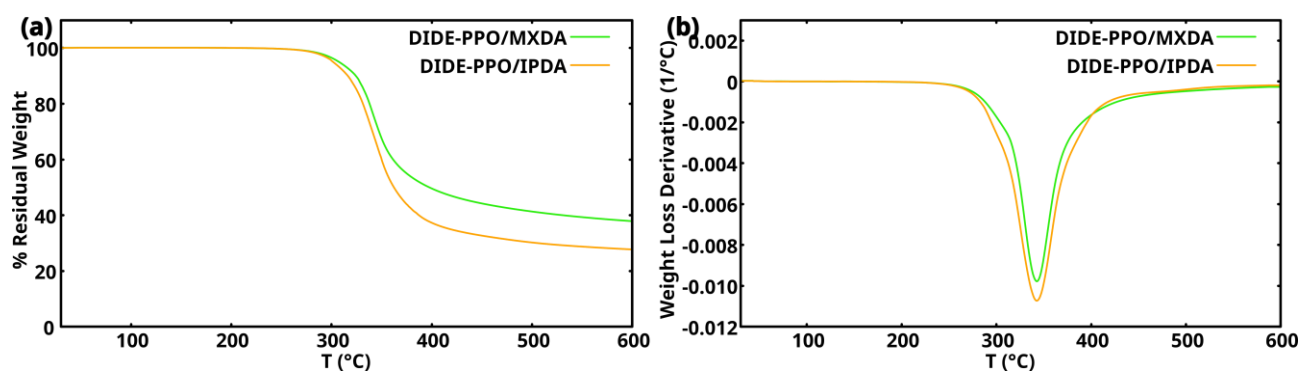


Figure S6. (a) Thermogravimetric curves in N₂ atmosphere and (b) their derivatives for the materials prepared.

4. Storage Moduli as a function of the angular frequency for the prepared materials

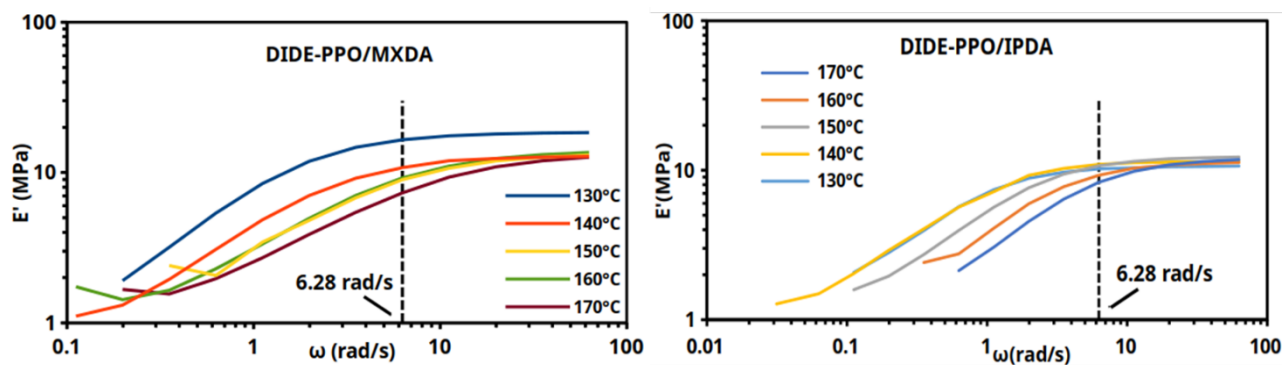


Figure S7. Storage modulus of DIDE-PPO/MXDA and DIDE-PPO/IPDA as a function of angular frequency at different temperatures.

5. Stress relaxation profiles and Kohlrausch-Williams-Watts (KKW) model analysis

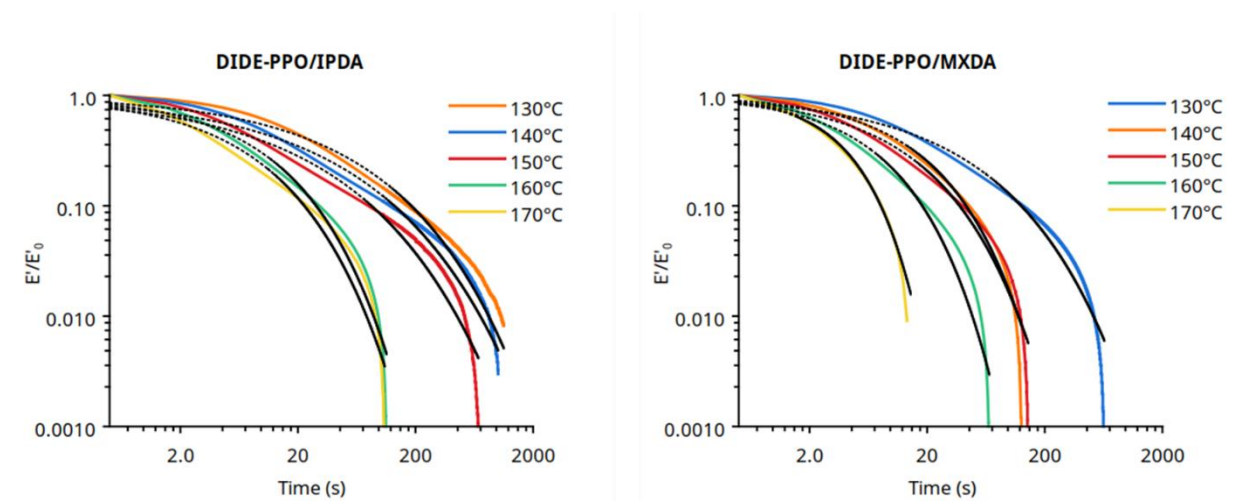


Figure S8. Tension mode stress relaxation profiles of the prepared materials (colored lines) fit by a stretched exponential decay function (dashed black lines).

Table S2. Fitting parameters for the stretched exponential decay function for the prepared materials.

Sample	T (°C)	β^a	τ^b (s)	R ²
DIDE-PPO/IPDA	130	0.46	29.64	0.976
	140	0.41	17.39	0.962
	150	0.41	11.27	0.958
	160	0.46	7.36	0.976
	170	0.57	5.22	0.955
DIDE-PPO/MXDA	130	0.49	22.95	0.981
	140	0.68	12.49	0.991
	150	0.60	9.38	0.980
	160	0.71	5.55	0.976
	170	1.00	3.42	0.983

^a Stretched exponential parameter.

^b Characteristic relaxation time.

6. Frequency sweeps experiments for DIDE-PPO/MXDA

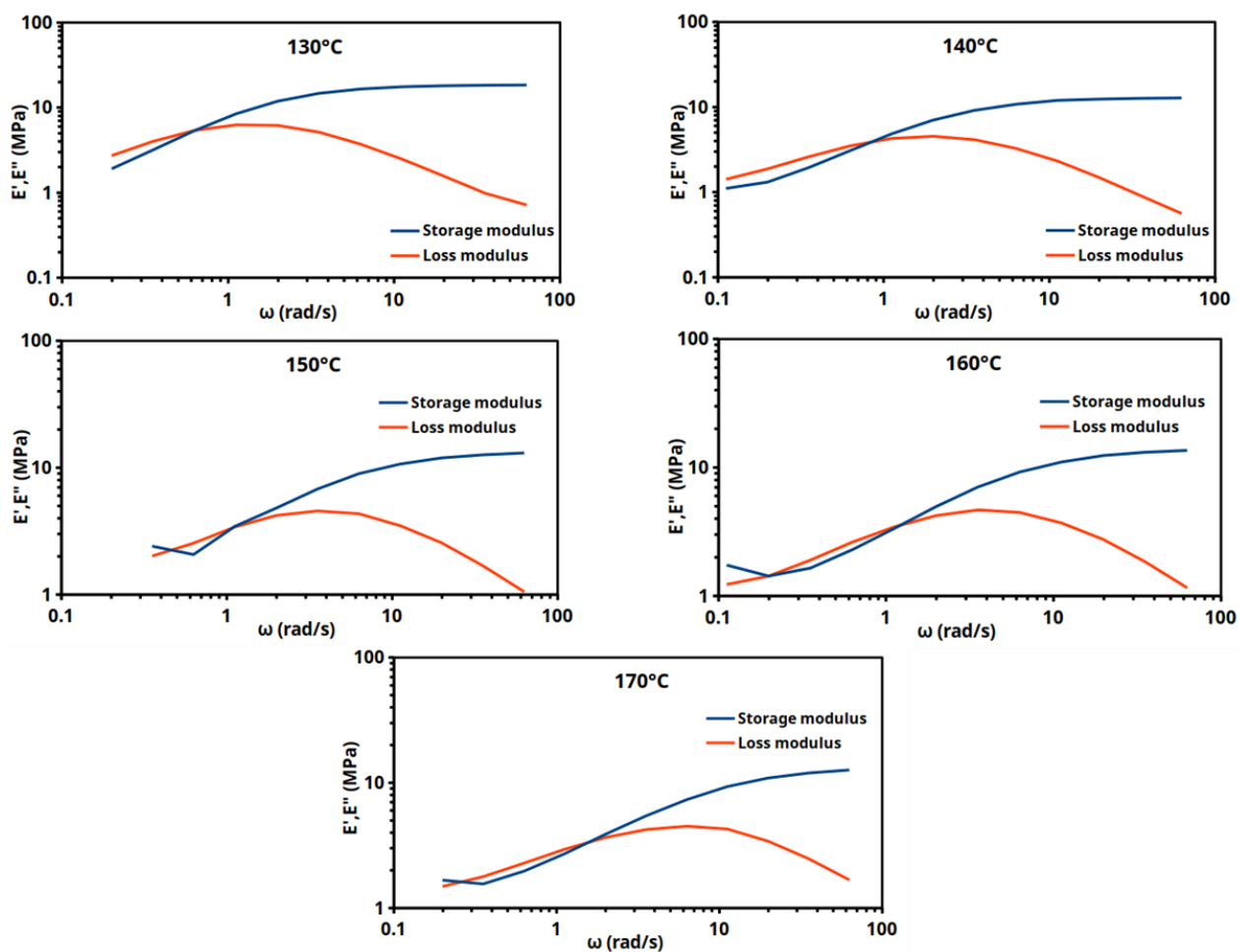


Figure S9. Storage and Loss Moduli of DIDE-PPO/MXDA as a function of the angular frequency.

7. Frequency sweeps experiments for DIDE-PPO/IPDA

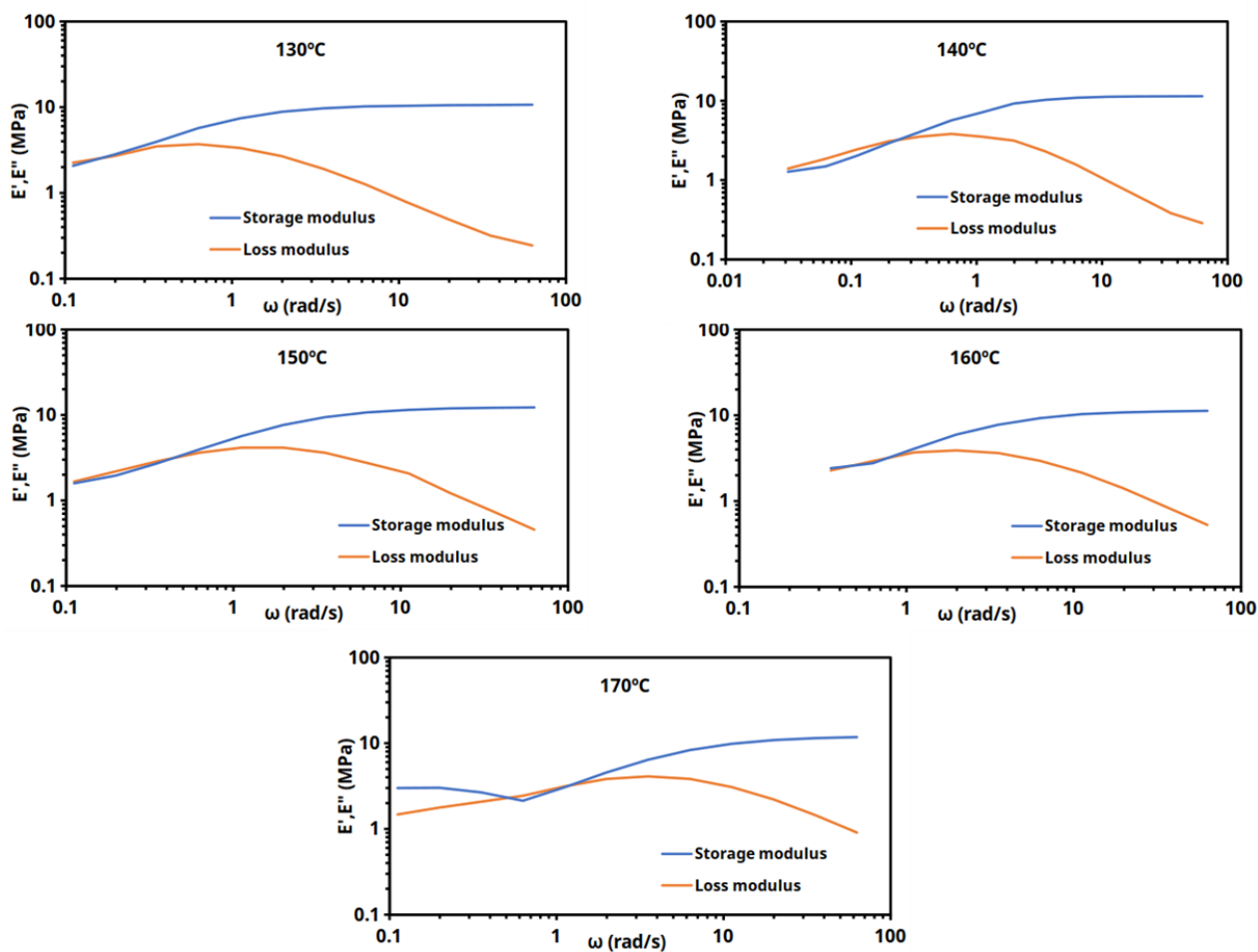


Figure S10. Storage and Loss Moduli of DIDE-PPO/IPDA as a function of the angular frequency.

8. Carbon fibers reinforced composites

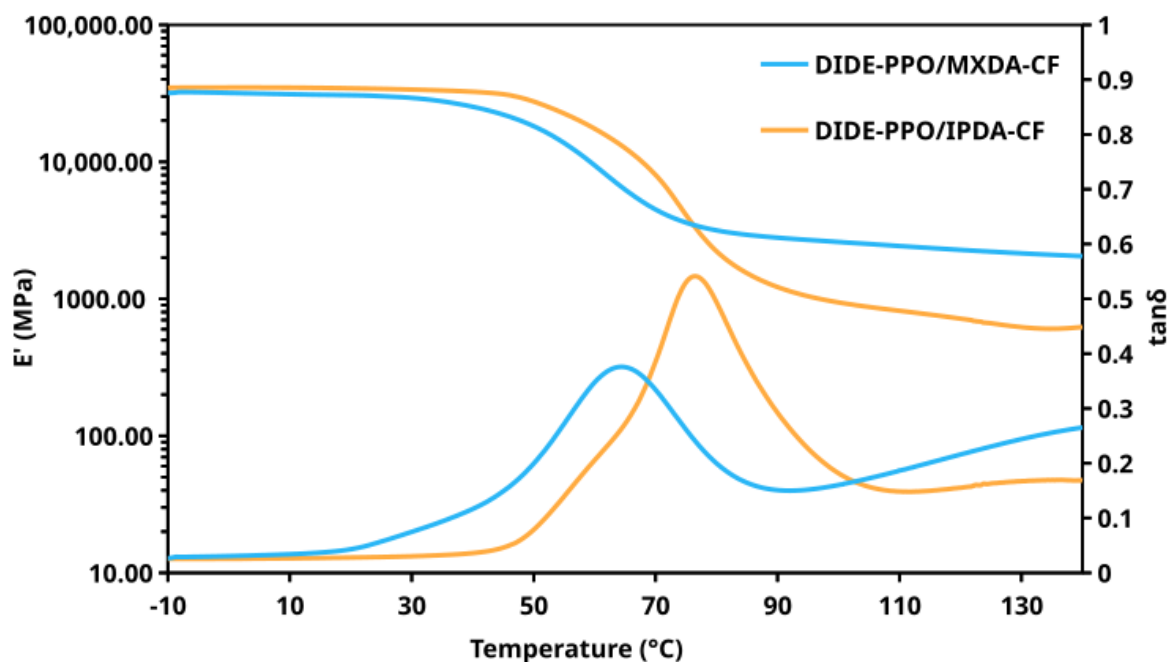


Figure S11. Evolution of storage modulus and $\tan \delta$ with temperature for the prepared composite materials.

Table S3. Main thermomechanical properties of the prepared composite materials.

Sample	E'_{Glassy}^a (MPa)	E'_{Rubbery}^b (MPa)	$T_{\tan \delta}^c$
DIDE-PPO/IPDA-CF	34422	708	76
DIDE-PPO/MXDA-CF	31146	2427	64

Table S4. Creep rate and strain recovery for the prepared carbon fiber reinforced composites.

Sample	$d\varepsilon/dt^a$	% Residual deformation ^b
DIDE-PPO/IPDA	2.39×10^{-3}	0.61
DIDE-PPO/MXDA	2.13×10^{-3}	0.37
DIDE-PPO/IPDA-CF	4.24×10^{-4}	0.13
DIDE-PPO/MXDA-CF	4.12×10^{-4}	0.02

^a Determined from the slope of the steady-state region of the creep curve.

^b Determined after 30 min of releasing the stress.