

Leveraging Lignin's Ortho-Methoxy Groups for Intramolecular Hydrogen Bonding to Reduce Dielectric Constant of Epoxy Resins

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1.1 Structural characterizations of diphenols

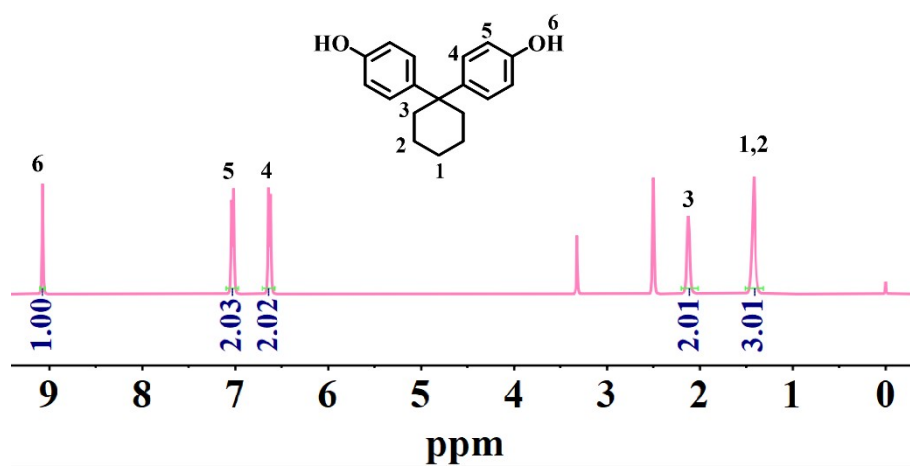


Fig. S1. ¹H NMR spectrum of PCD.

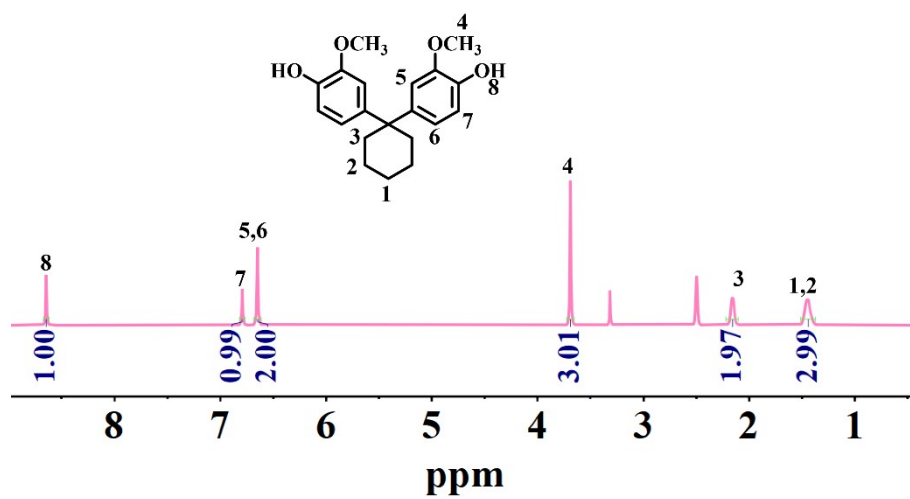


Fig. S2. ¹H NMR spectrum of GCD.

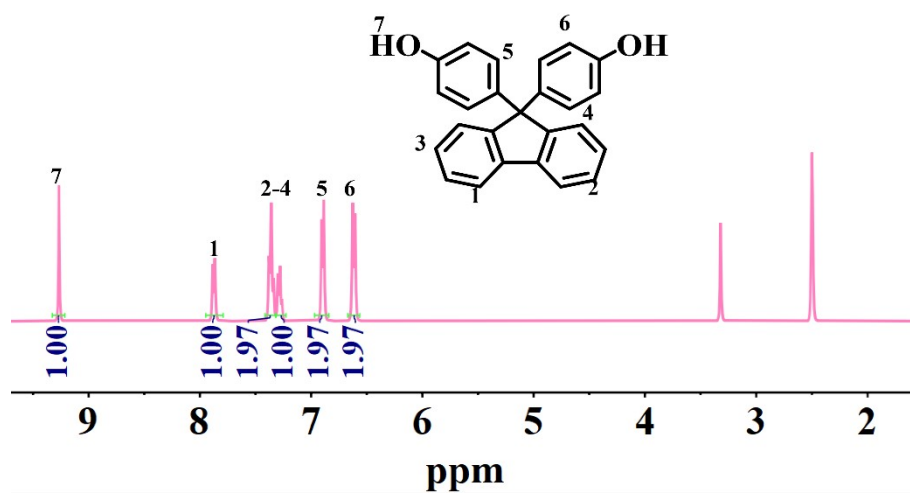


Fig. S3. ^1H NMR spectrum of PFD.

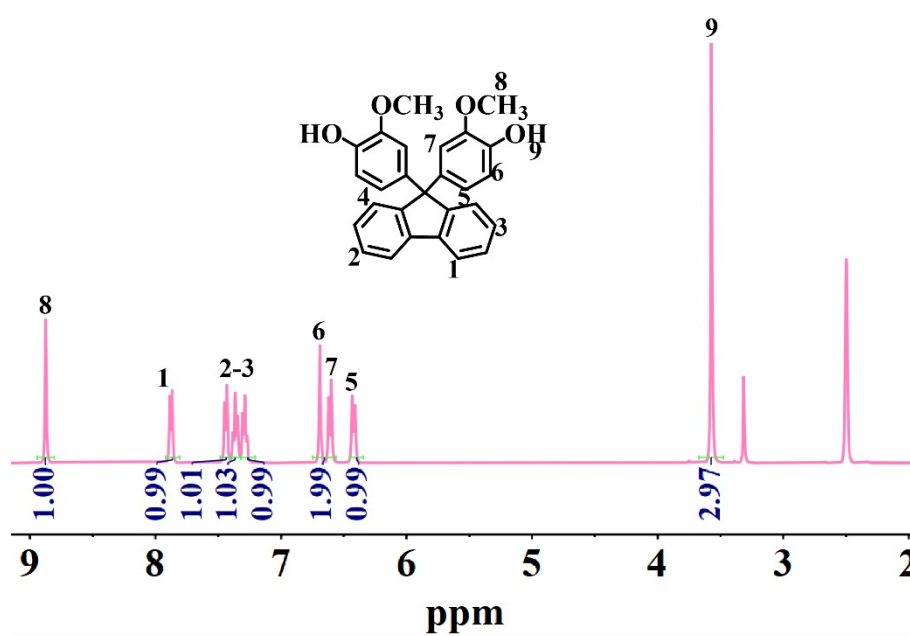


Fig. S4. ^1H NMR spectrum of GFD.

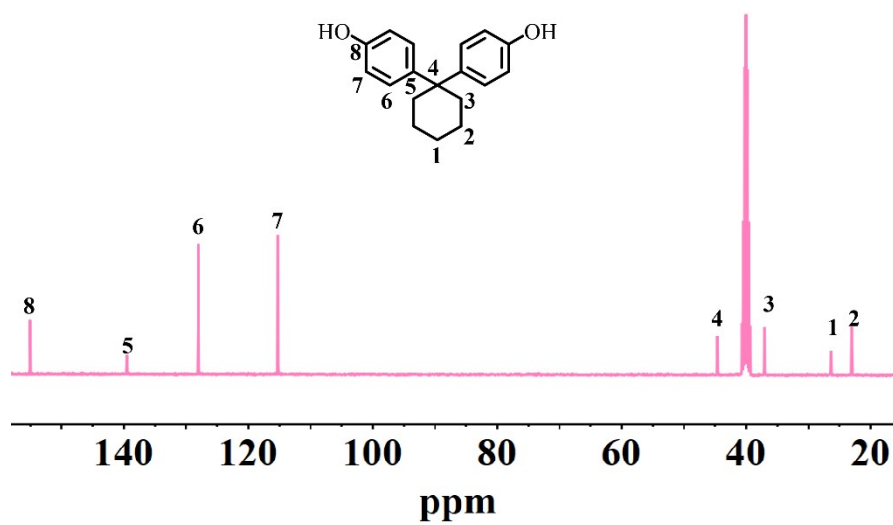


Fig. S5. ^{13}C NMR spectrum of PCD.

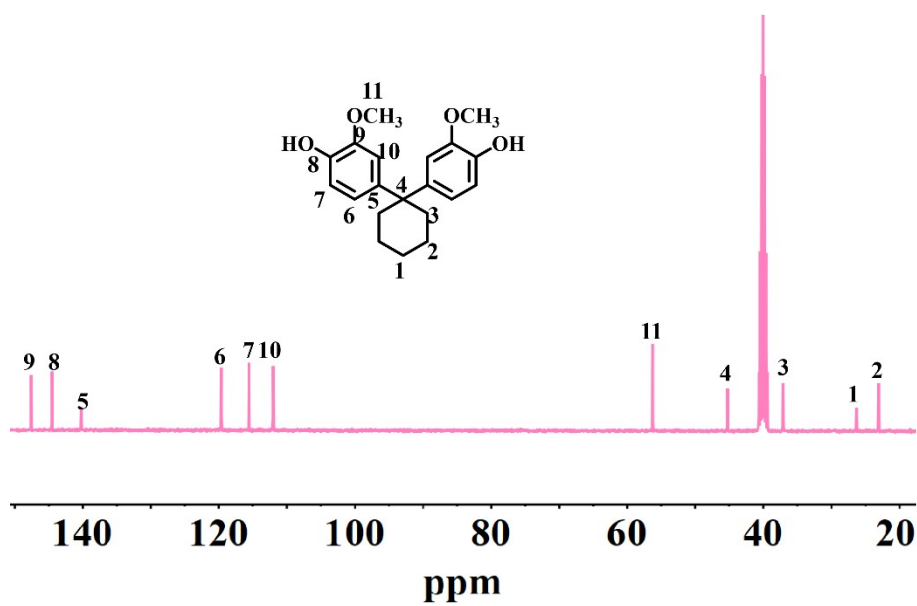


Fig. S6. ^{13}C NMR spectrum of GCD.

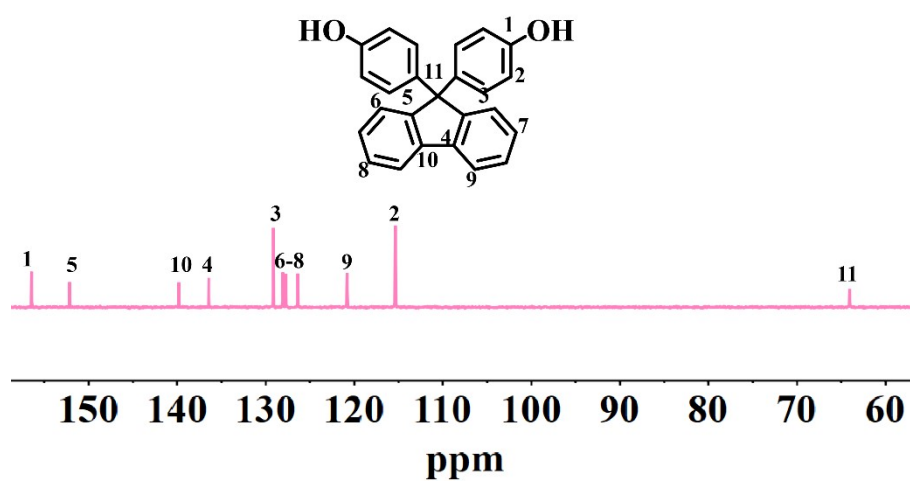


Fig. S7. ^{13}C NMR spectrum of PFD.

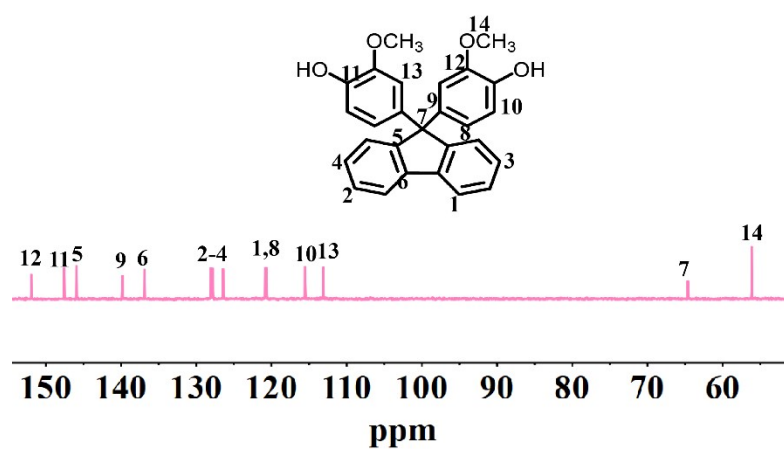


Fig. S8. ^{13}C NMR spectrum of GFD.

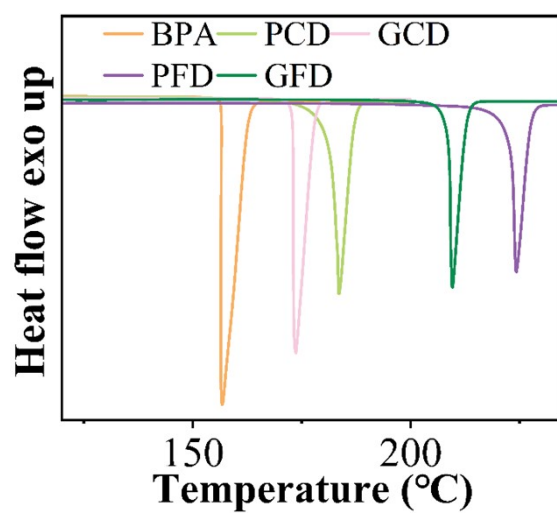


Fig. S9. Melting points of diphenols.

1.2 Structural characterizations of epoxy resins

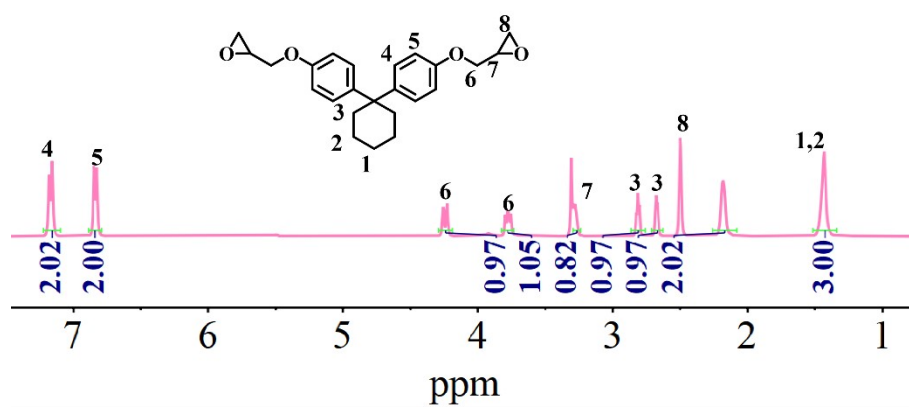


Fig. S10. ¹H NMR spectrum of PCE.

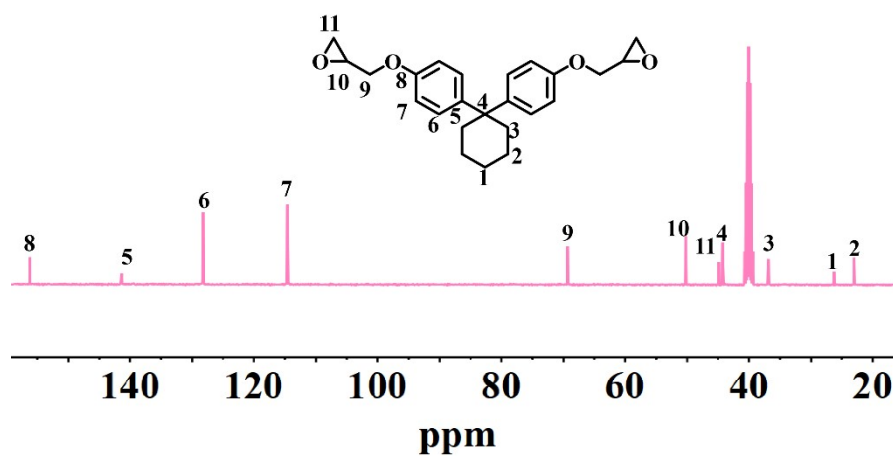


Fig. S11. ^{13}C NMR spectrum of PCE.

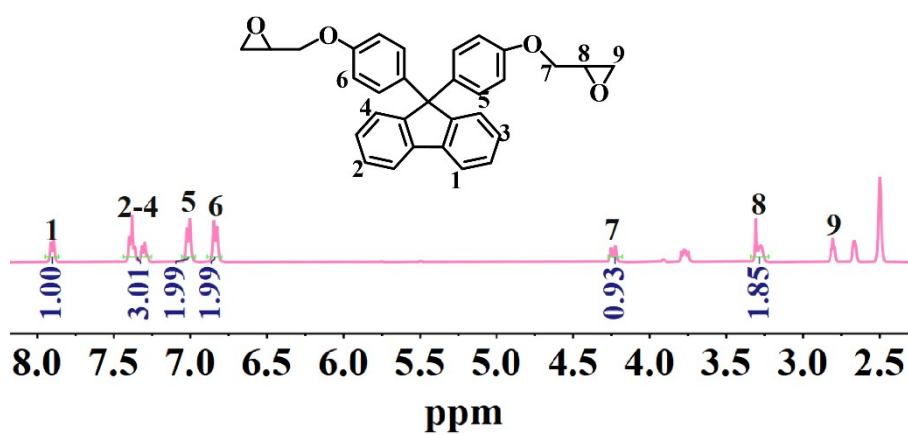


Fig. S12. ^1H NMR spectrum of PFE.

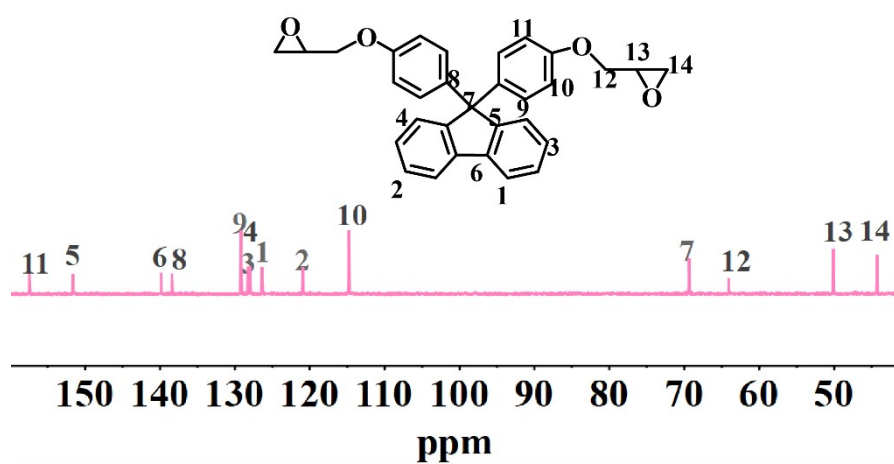


Fig. S13. ¹³C NMR spectrum of PFE.

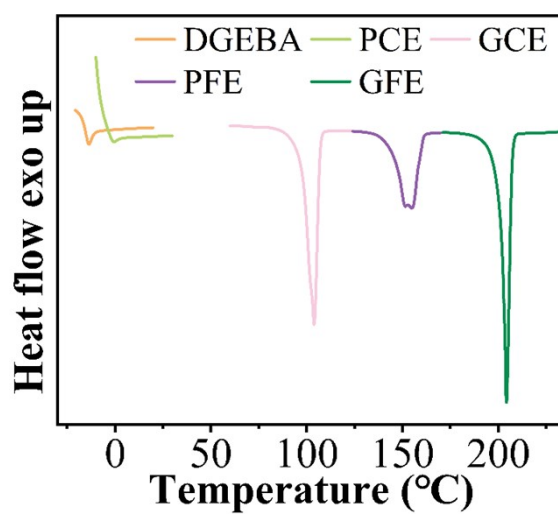


Fig. S14. Melting points of epoxy resins.

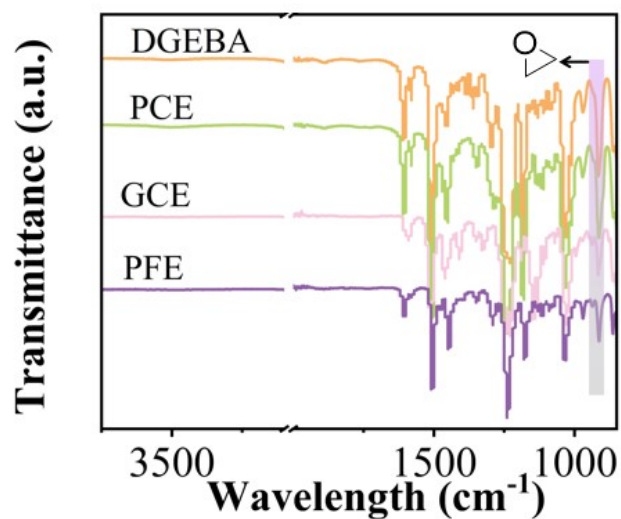


Fig. S15. FTIR spectra of epoxy resins.

1.3 Structural characterizations of the cured epoxy resins

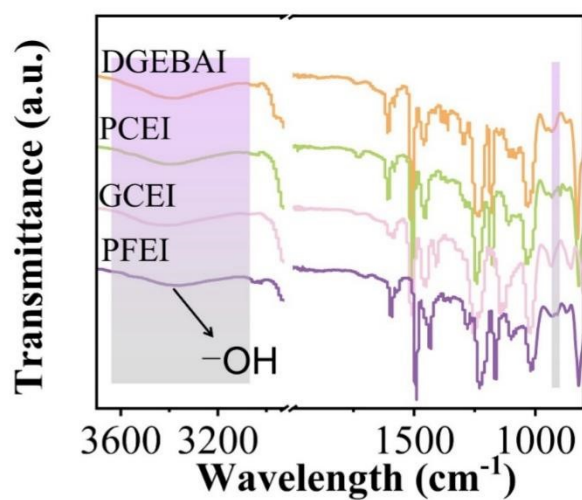


Fig. S16. FTIR spectra of the cured epoxy resins.

Table S1. FTIR analysis of epoxy monomers and cured epoxy resins.

	DGEBA	DGEBAI	PCE	PCEI	PFE	PFEI	GFE	GFEI	GCE	GCEI
Epoxy Group	√	—	√	—	√	—	√	—	√	—
Hydroxyl Group	—	√	—	√	—	√	—	√	—	√

√: existence

—: inexistence

1.4 Swelling Photos' images of epoxy thermosets

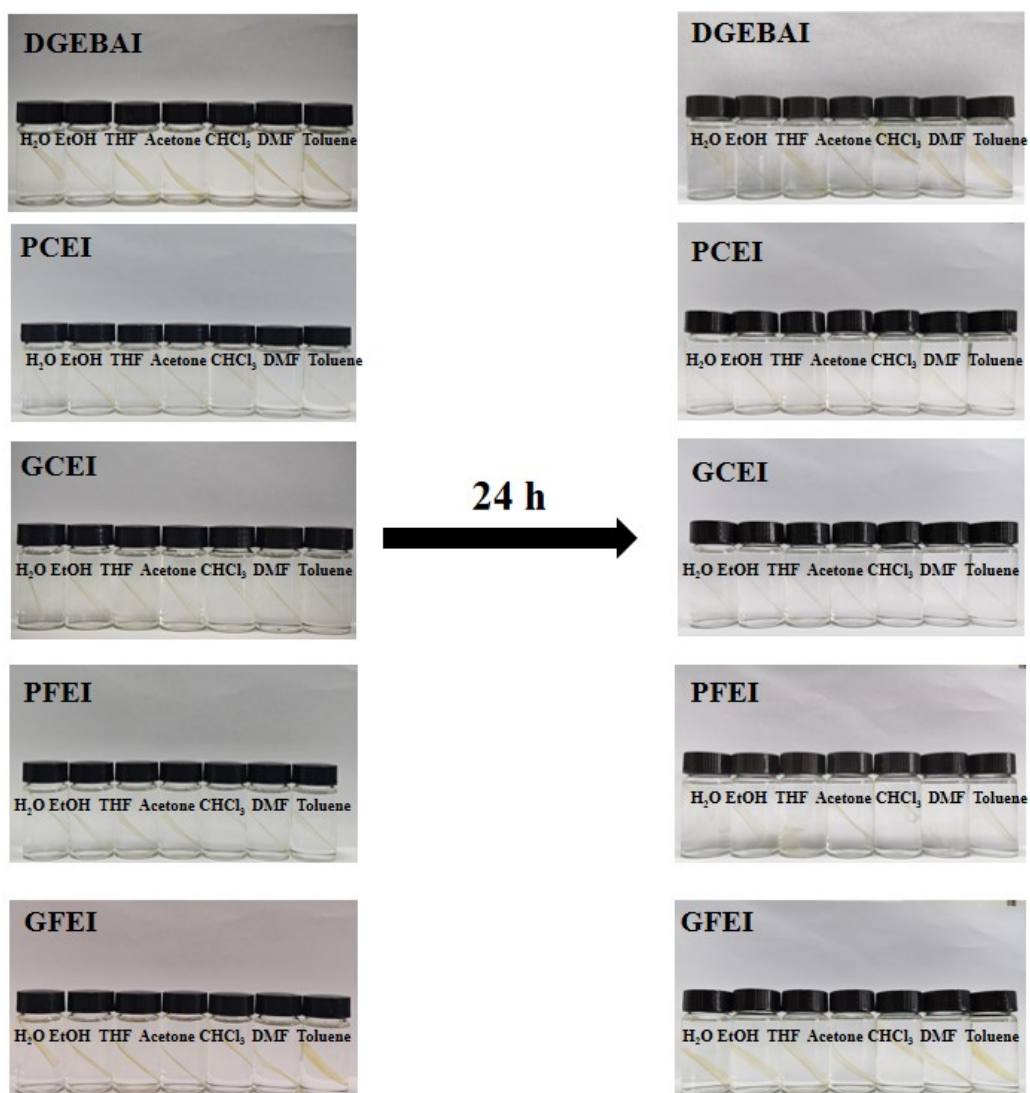


Fig. S17. Swelling photos of epoxy thermosets in different solvents.

1.5 Non-isothermal DSC curves of epoxy thermosets after hygrothermal aging

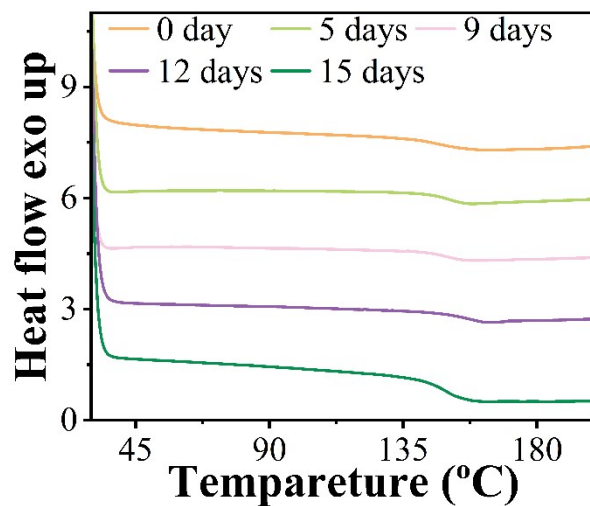


Fig. S18. Non-isothermal DSC curves of epoxy thermosets (DGEBAI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

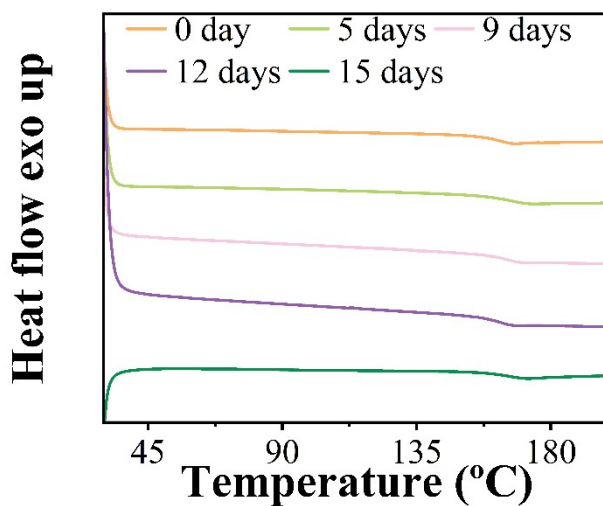


Fig. S19. Non-isothermal DSC curves of epoxy thermosets (PCEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

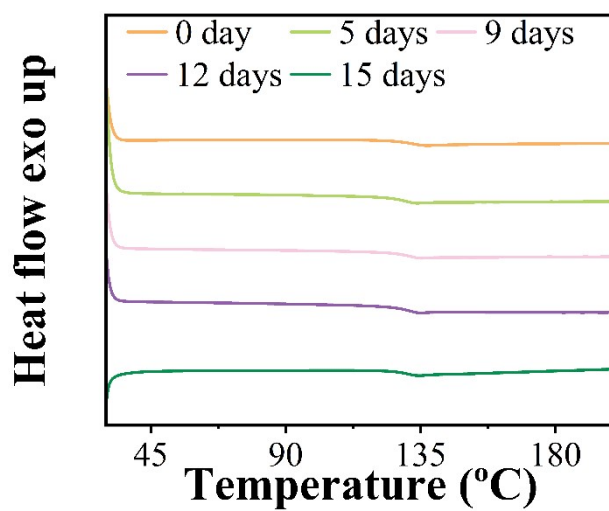


Fig. S20. Non-isothermal DSC curves of epoxy thermosets (GCEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

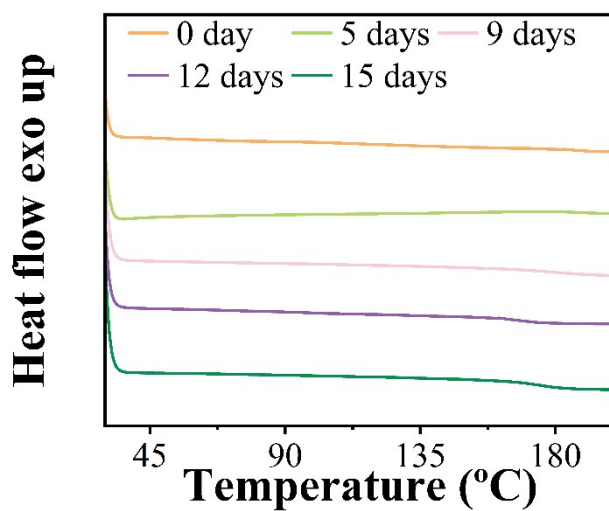


Fig. S21. Non-isothermal DSC curves of epoxy thermosets (PFEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

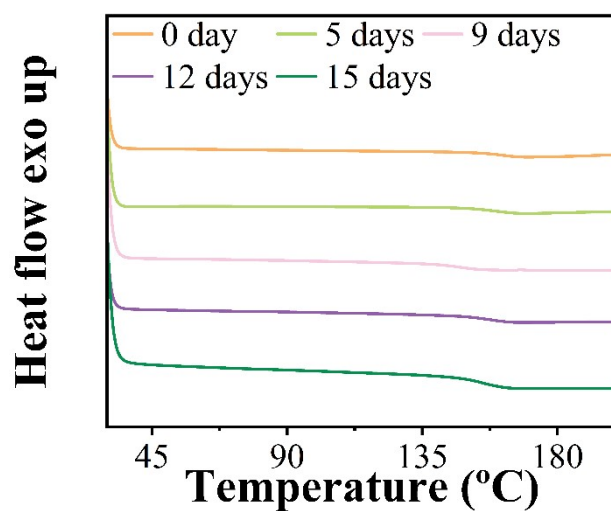


Fig. S22. Non-isothermal DSC curves of epoxy thermosets (GFEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

1.6 Thermal and mechanical properties of epoxy thermosets

Table S2. Thermal and mechanical properties of epoxy thermosets.

Samples	T_g (°C)	T_g+60 (K)	$T_g+60(K)$ Storage Modulus (MPa)	v_e (mol m ⁻³)
DGEBAI	154	487	12.47	1027
PCEI	160	493	12.52	1018
GCEI	128	461	8.96	779
PFEI	161	494	11.6	1072
GFEI	173	506	10.43	826

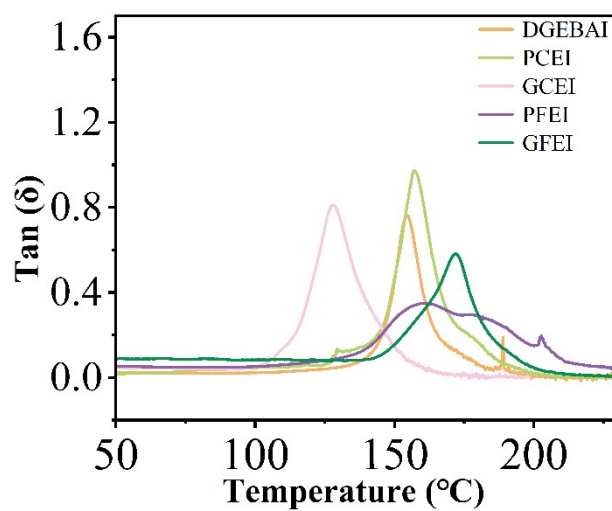


Fig. S 23 Tan δ as a function of temperatures through DMA.

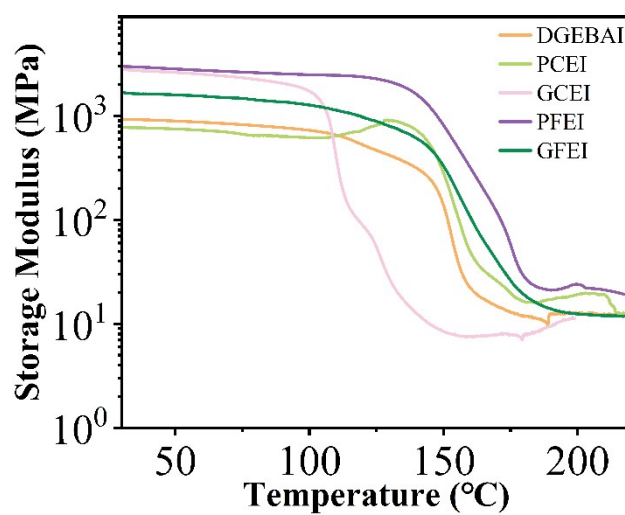


Fig. S24. Storage modulus as a function of temperatures through DMA.

1.7 Tensile stress-strain curves of epoxy thermosets after hygrothermal aging

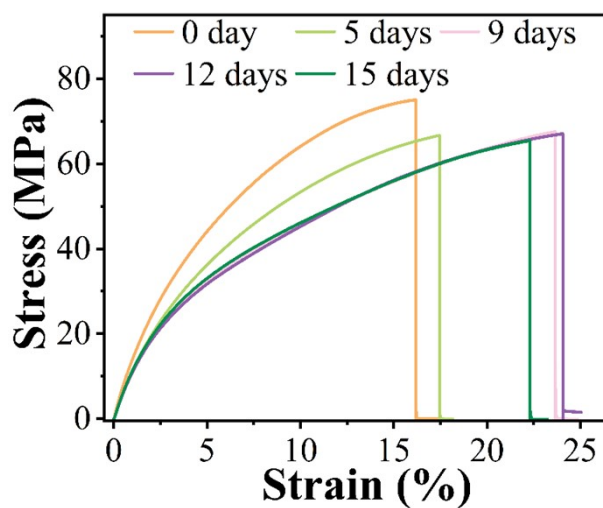


Fig. S25. Tensile stress-strain curves of epoxy thermosets (DGEBAI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

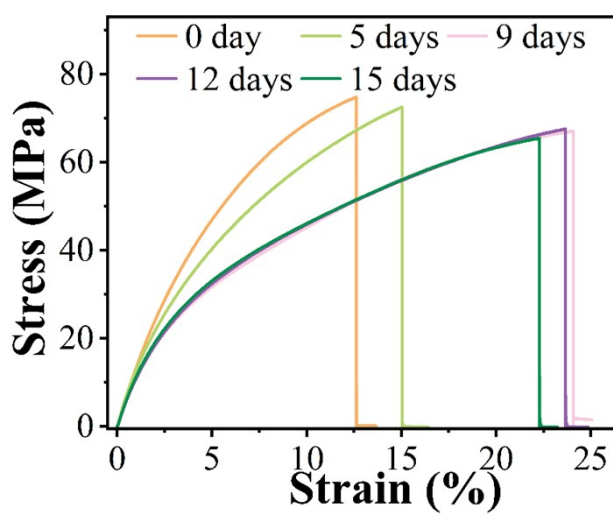


Fig. S26. Tensile stress-strain curves of epoxy thermosets (PCEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

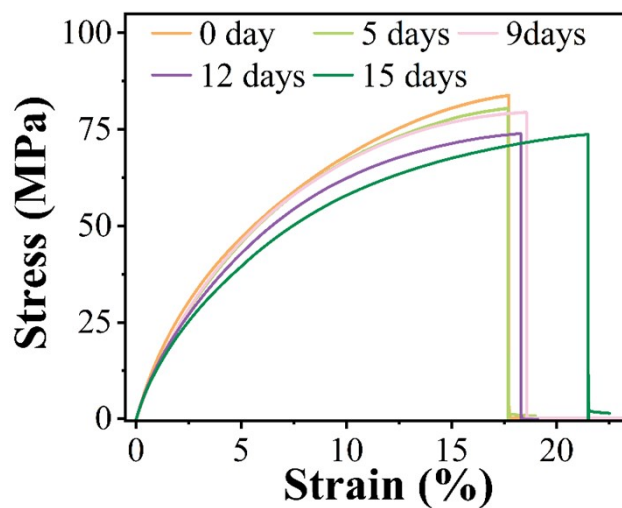


Fig. S27. Tensile stress-strain curves of epoxy thermosets (GCEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

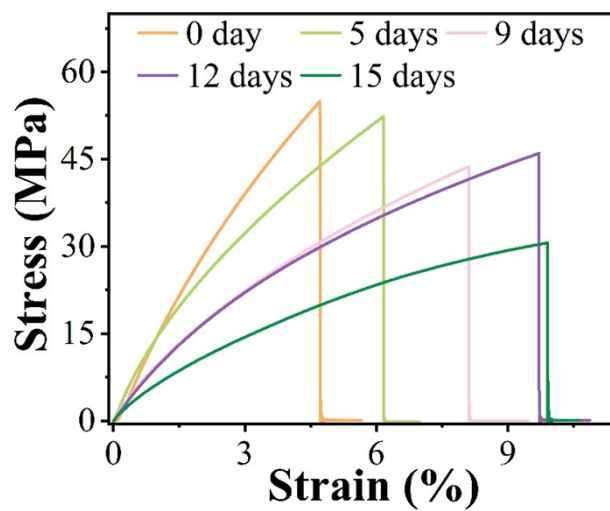


Fig. S28. Tensile stress-strain curves of epoxy thermosets (PFEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

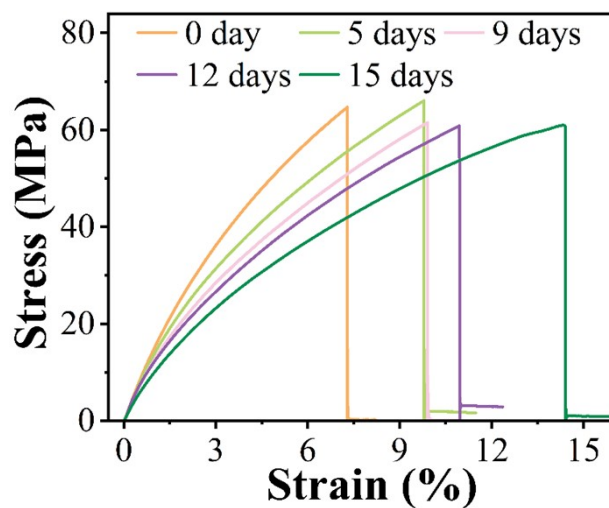


Fig. S29. Tensile stress-strain curves of epoxy thermosets (GFEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hydrothermal aging.

1.8 Moduli of epoxy thermosets after hydrothermal aging

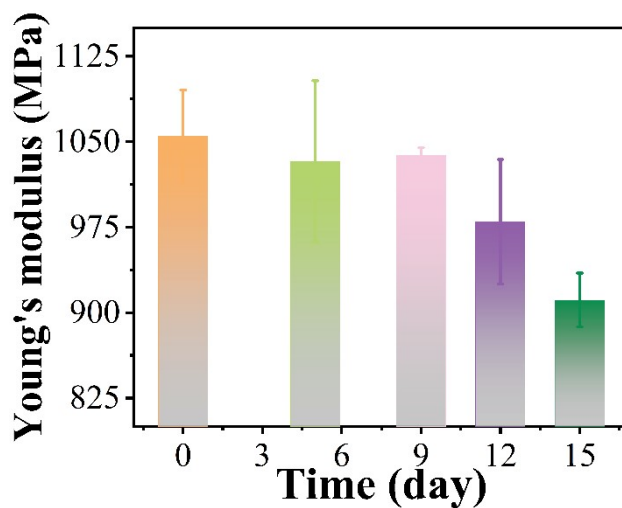


Fig. S30. Moduli of epoxy thermosets (DGEBAI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hydrothermal aging.

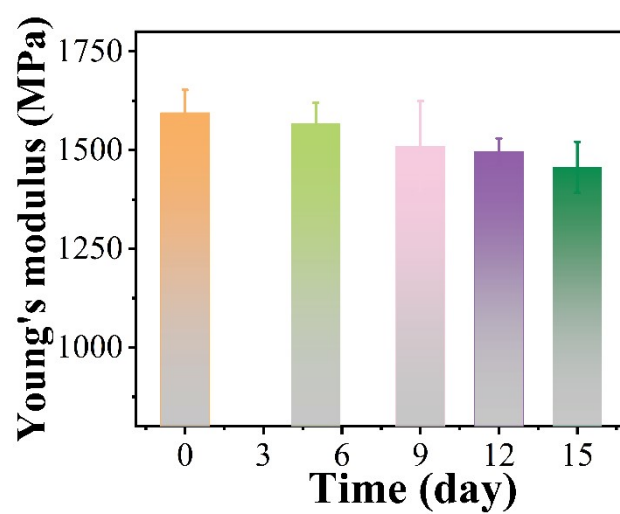


Fig. S31. Moduli of epoxy thermosets (PCEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

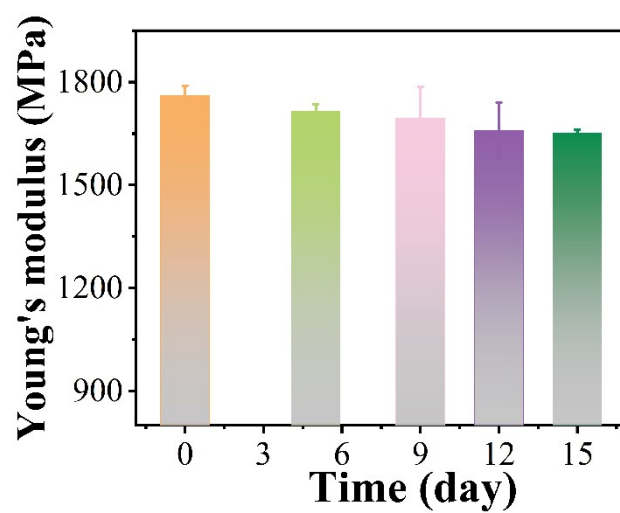


Fig. S32. Moduli of epoxy thermosets (GCEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

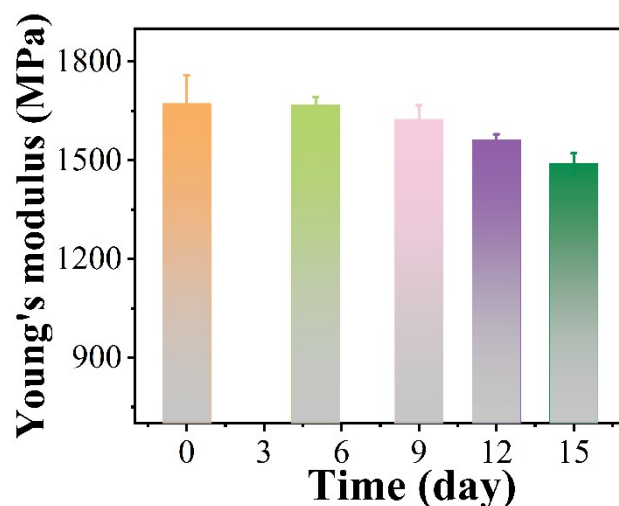


Fig. S33. Moduli of epoxy thermosets (GFEI) after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging.

Table S3. Tensile strength of epoxy thermosets after 0 day, 5 days, 9 days, 12 days, and 15days' hygrothermal aging

Time	Samples	DGEBAI	PCEI	GCEI	PFEI	GFEI
		Strength (MPa)				
0 day		75.0±9.1	74.7±5.1	83.8±6.3	54.9±4.1	64.7±8.0
5 days		67.5±5.3	67.6±3.0	80.5±7.1	50.0±6.0	61.4±2.8
9 days		67.1±6.2	67.2±2.8	79.9±4.4	45.4±3.7	61.0±5.1
12 days		65.5±4.3	65.4±5.4	74.3±8.3	44.1±6.5	60.7±6.3
15 days		65.4±5.2	65.3±4.1	73.7±5.9	30.6±4.5	60.9±3.5

Table S4. Moduli of epoxy thermosets after 0 day, 5 days, 9 days, 12 days, and 15 days' hygrothermal aging

Time	Samples	DGEBAI	PCEI	GCEI	PFEI	GFEI
		Modulus (MPa)				
0 day		1055±41	1595 ± 58	1762±27	1376 ± 89	1672 ± 85
5 days		1033±71	1566 ± 53	1715±21	1177 ± 67	1668 ± 24
9 days		1038±7	1510 ± 114	1696±90	1069 ± 35	1624 ± 42
12 days		980±55	1496 ± 34	1660±81	958 ± 57	1561 ± 17
15 days		911±24	1456 ± 65	1652±10	834 ± 42	1490 ± 31

1.9 Thermal-dependent FTIR spectra of epoxy thermosets without ortho-methoxy groups

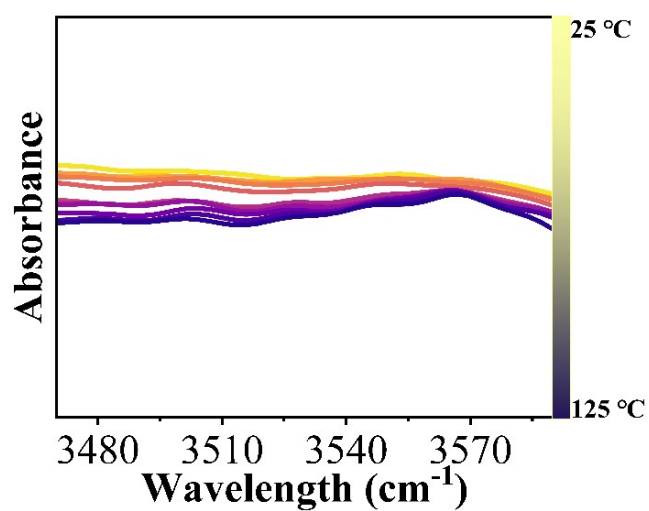
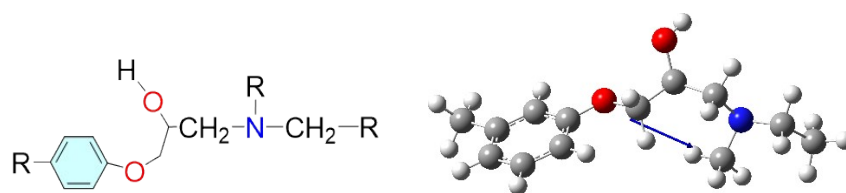


Fig. S34. Thermal-dependent FTIR spectra of epoxy thermosets without ortho-methoxy groups.

2.0 The detail for calculating EPI₁ and EPI₂.

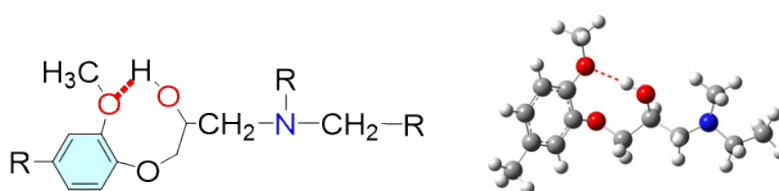


EPI₁

Generated by Multiwfn (<http://sobereva.com/multiwfn>)

C	4.38621680	-0.06414603	0.55404495
C	3.11481468	0.47673270	0.72169119
C	2.04536536	0.07626260	-0.09234201
C	2.24790427	-0.88035108	-1.09069842
C	3.52640912	-1.42313675	-1.25671990
C	4.58574273	-1.02683892	-0.44964733
O	0.84887964	0.67581259	0.17445433
C	-0.27138431	0.31592118	-0.61826200
C	-1.46038085	1.14621202	-0.16568714
O	-1.22489588	2.46380896	-0.66096140
C	-2.79782975	0.58155274	-0.69394352
N	-3.56608603	-0.14555901	0.31459571
C	-4.91877532	-0.42639024	-0.17411607
C	-2.88476362	-1.35641851	0.75484325
H	2.92234788	1.22333770	1.48640988
H	1.43760866	-1.20399765	-1.73259549
H	3.68629717	-2.16763589	-2.03216585
H	5.57245014	-1.45978255	-0.59240803
H	-0.08071323	0.52560919	-1.67993274
H	-0.48315231	-0.75559740	-0.51118771

H	-1.49316999	1.14856070	0.93297026
H	-1.81888489	3.06489011	-0.18647380
H	-3.39969982	1.43698873	-1.01763789
H	-2.61873743	-0.03146420	-1.60289350
H	-5.30756604	0.50807822	-0.59551581
H	-4.90378329	-1.16223169	-1.00382403
H	-3.47526759	-1.86084704	1.52277204
H	-1.91972272	-1.10738838	1.20395619
H	-2.70818860	-2.07467704	-0.07133216
C	5.53492602	0.37965123	1.42751556
H	5.20031912	1.06682995	2.21038514
H	6.01897669	-0.47612477	1.91319490
H	6.30526051	0.89301646	0.83853066
C	-5.87332924	-0.90117043	0.92020008
H	-6.89540470	-0.94686473	0.52888928
H	-5.62262127	-1.89982675	1.29138229
H	-5.85511152	-0.20621532	1.76609104



EPI₂

Generated by Multiwfn (<http://sobereva.com/multiwfn>)

C	3.93475223	-1.33137829	-0.32512060
C	4.36899220	-0.06601670	-0.71595423
C	2.70982992	-1.42677697	0.35274634

C	3.60947203	1.07973639	-0.45351090
H	5.31628730	0.03912189	-1.23779077
C	1.94221927	-0.30111427	0.62307275
H	2.34326427	-2.39286007	0.68991709
C	2.39014506	0.97017115	0.20976209
H	3.97379143	2.04593879	-0.78219150
O	0.76359685	-0.37189642	1.31927278
O	1.55290660	2.01884369	0.49572828
C	-0.32392131	-1.09006436	0.71315953
C	1.92133692	3.30640574	0.02162811
H	-0.24279282	1.36374988	0.10580203
C	-1.46801743	-0.10592025	0.46444278
H	-0.63341339	-1.88602325	1.40066011
H	0.00470686	-1.53748604	-0.23194104
H	2.02423427	3.31469617	-1.07063113
H	2.86009902	3.64123852	0.47946222
H	1.11266318	3.97565898	0.31824207
O	-1.00760825	0.96250527	-0.35005526
C	-2.63659390	-0.81058390	-0.22612577
H	-1.82011394	0.26878978	1.43805570
N	-3.92712034	-0.15563230	-0.02639666
H	-2.71954376	-1.81942879	0.19767273
H	-2.38004879	-0.92925937	-1.29740799
C	-4.02024618	1.10431499	-0.76134831
C	-5.02844384	-1.06038441	-0.35742274
H	-4.05959865	0.94789632	-1.85807143

H	-4.91971996	1.64824951	-0.46045480
H	-4.84184308	-2.00782267	0.16298592
H	-5.04948259	-1.29423001	-1.44235733
C	4.75096656	-2.56915237	-0.60717614
H	5.01771835	-3.09357827	0.31862090
H	4.19751952	-3.28093650	-1.23219260
H	5.68023269	-2.32049328	-1.12879256
H	-3.14749838	1.71892101	-0.54286297
C	-6.39442718	-0.52963760	0.07468515
H	-6.70564948	0.34142485	-0.51011934
H	-7.15713755	-1.30456136	-0.05979731
H	-6.37137988	-0.24279521	1.13137385