Supporting Information

Construction of Brick/Mortar-Like Graphene/Thermoset Composites with Highly Anisotropic Thermal Conductivity and Strong Electromagnetic Interference Shielding Performance

Zhenhao Yao, Jianan Song, Yin Lu, Rui Yang, Yuan Zhang, and Kan Zhang**

Institute of Polymer Materials, School of Materials Science and Engineering, Jiangsu University, Zhenjiang 200237, China.

*: To whom correspondence should be addressed:

Jianan Song: songjianan@ujs.edu.cn; Kan Zhang: zhangkan@ujs.edu.cn

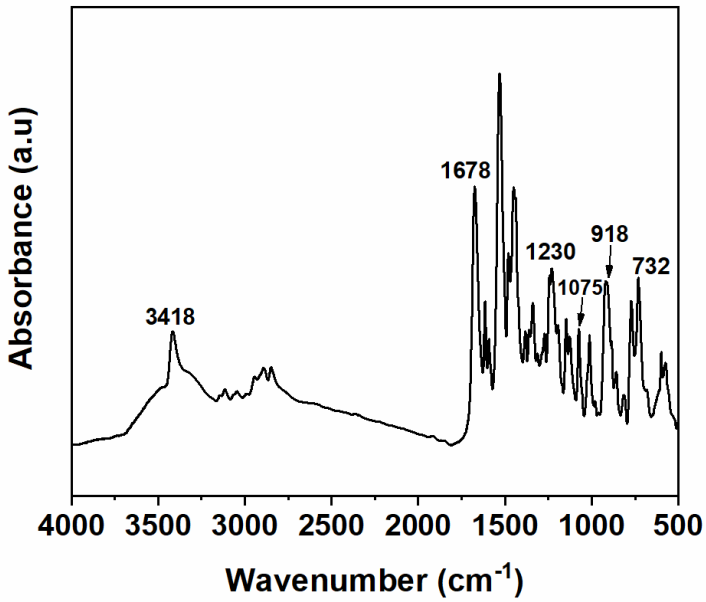


Figure S1. FTIR spectrum of BZ monomer.

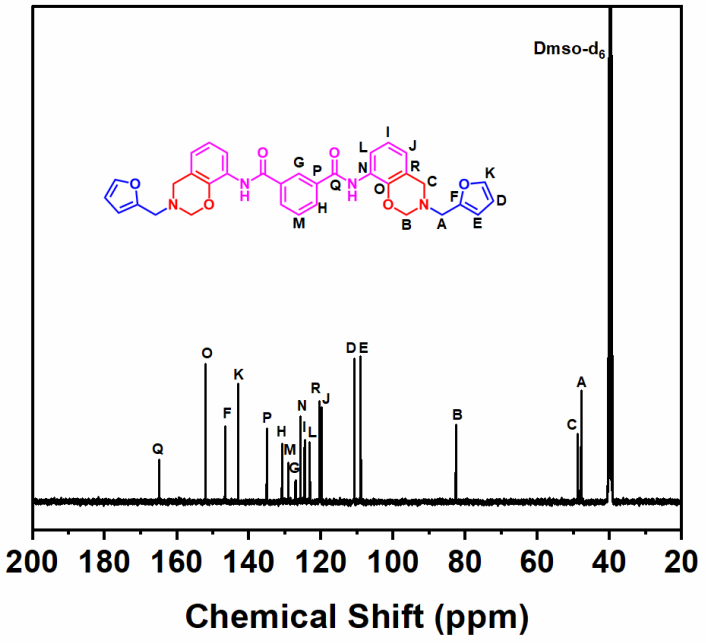


Figure S2. ^{13}C NMR spectrum of BZ monomer.

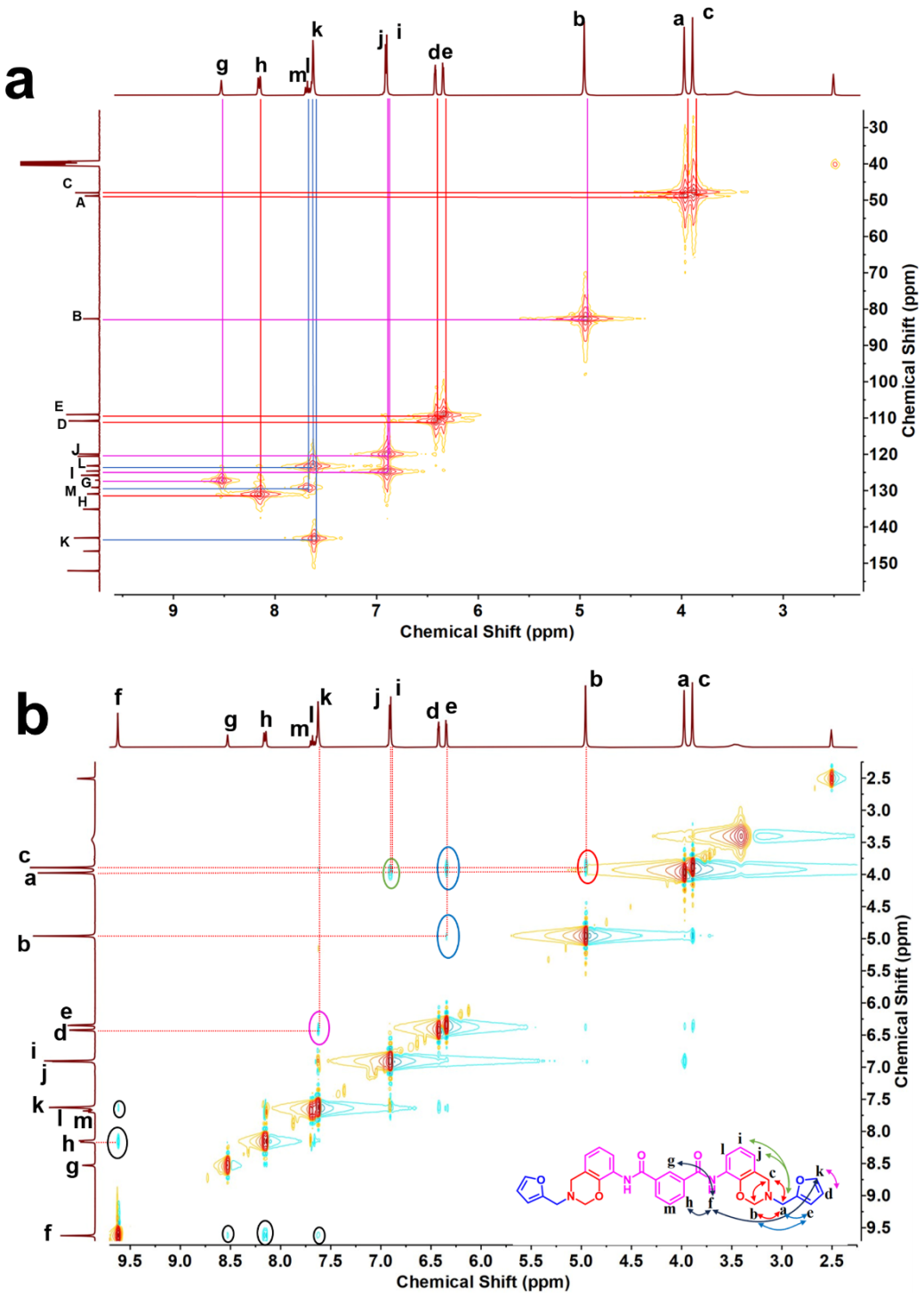


Figure S3. 2D ^{1}H-^{13}C HMQC (a) and ^{1}H-^{1}H NOESY (b) spectra of BZ monomer.

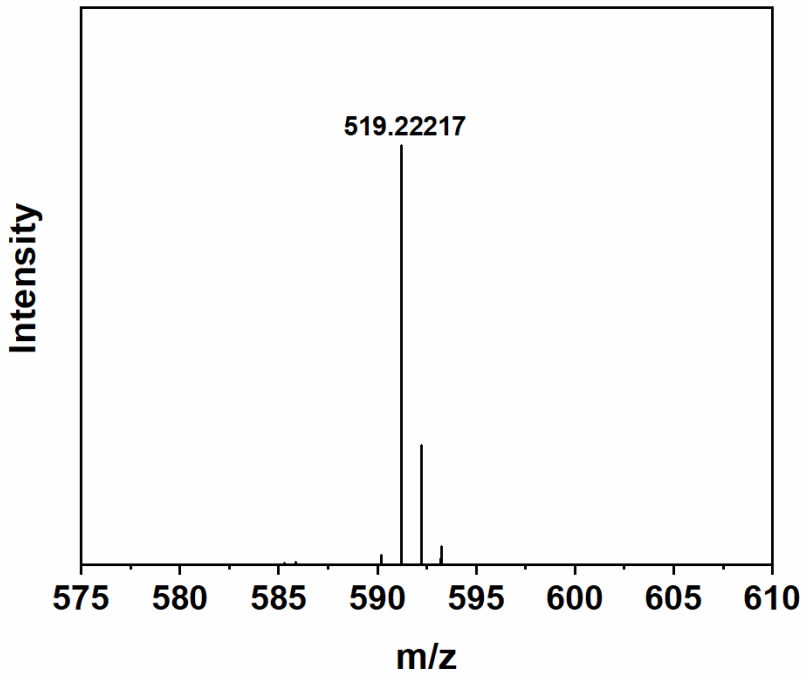
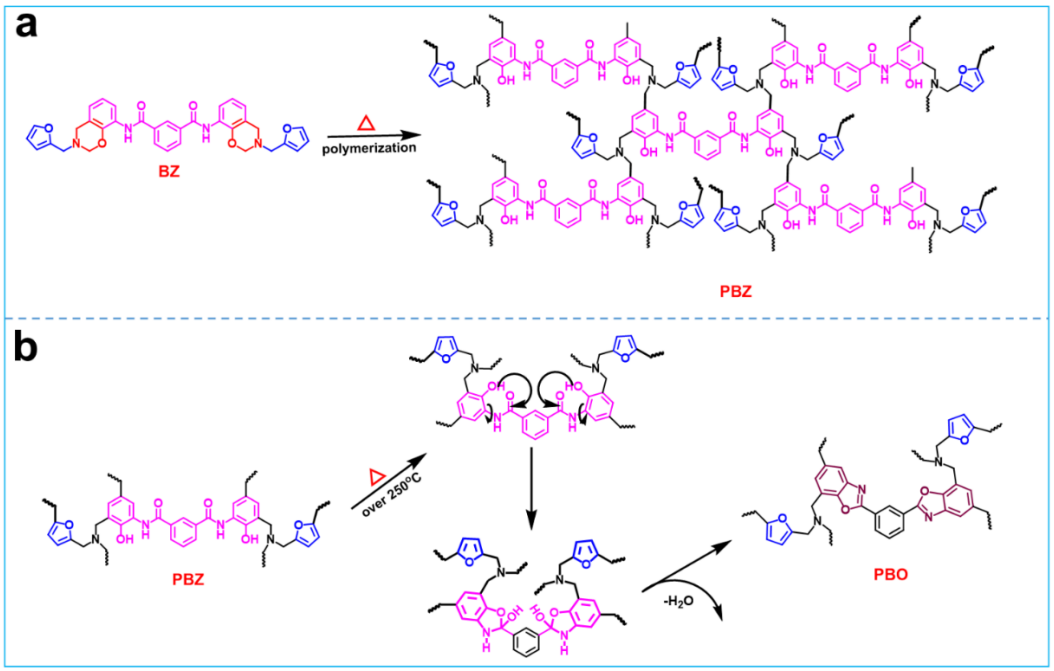


Figure S4. HR-MS spectrum of BZ monomer.



Scheme S1. Proposed polymerization mechanisms of PBZ(a), PBO(b)

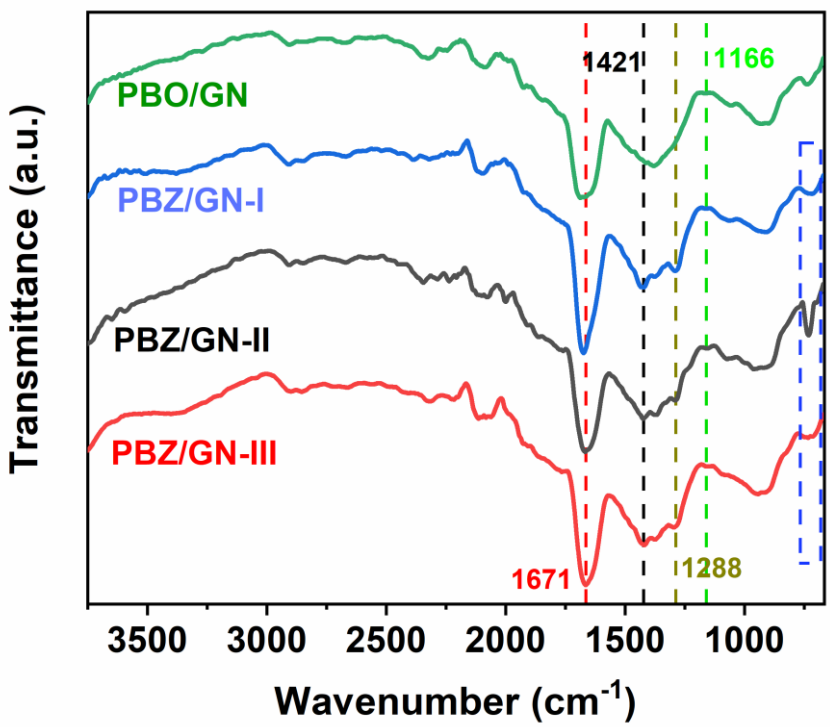


Figure S5. FTIR spectra of composite sheets.

Table S1. The polymeric matrix contents in graphene composite sheets based on TGA results.

Sample	PBZ (wt%)	PBO (wt%)	GN (wt%)
PBZ/GN-I	53.2	-	46.8
PBZ/GN-II	60.6	-	39.4
PBZ/GN-III	66.7	-	33.3
PBO/GN	-	51.1	48.9

Table S2. XRD analysis results.

Sample	002 peek/°	FWHM/°	d/Å
PBZ/GN-I	25.590	1.166	3.48
PBZ/GN-II	25.461	1.028	3.49
PBZ/GN-III	25.270	1.042	3.52
PBO/GN	25.920	1.225	3.43
GN	26.550	0.361	3.35
PBZ	20.850	0.959	4.26

Table S3. Raman analysis results.

	D	G
PBZ/GN-I	1344	1582
PBZ/GN-II	1344	1583
PBZ/GN-III	1345	1582
PBO/GN	1344	1577
GN	1349	1583

Table S4. TGA analysis data.

	GN	PBZ	PBO	PBZ/GN-I	PBZ/GN-II	PBZ/GN-III	PBO/GN
T_{d5} (°C)	514	315	406	354	344	349	365
T_{d10} (°C)	-	381	475	397	386	388	421
Yc (%)	93.35	61.01	66.35	76.14	73.73	71.75	79.55

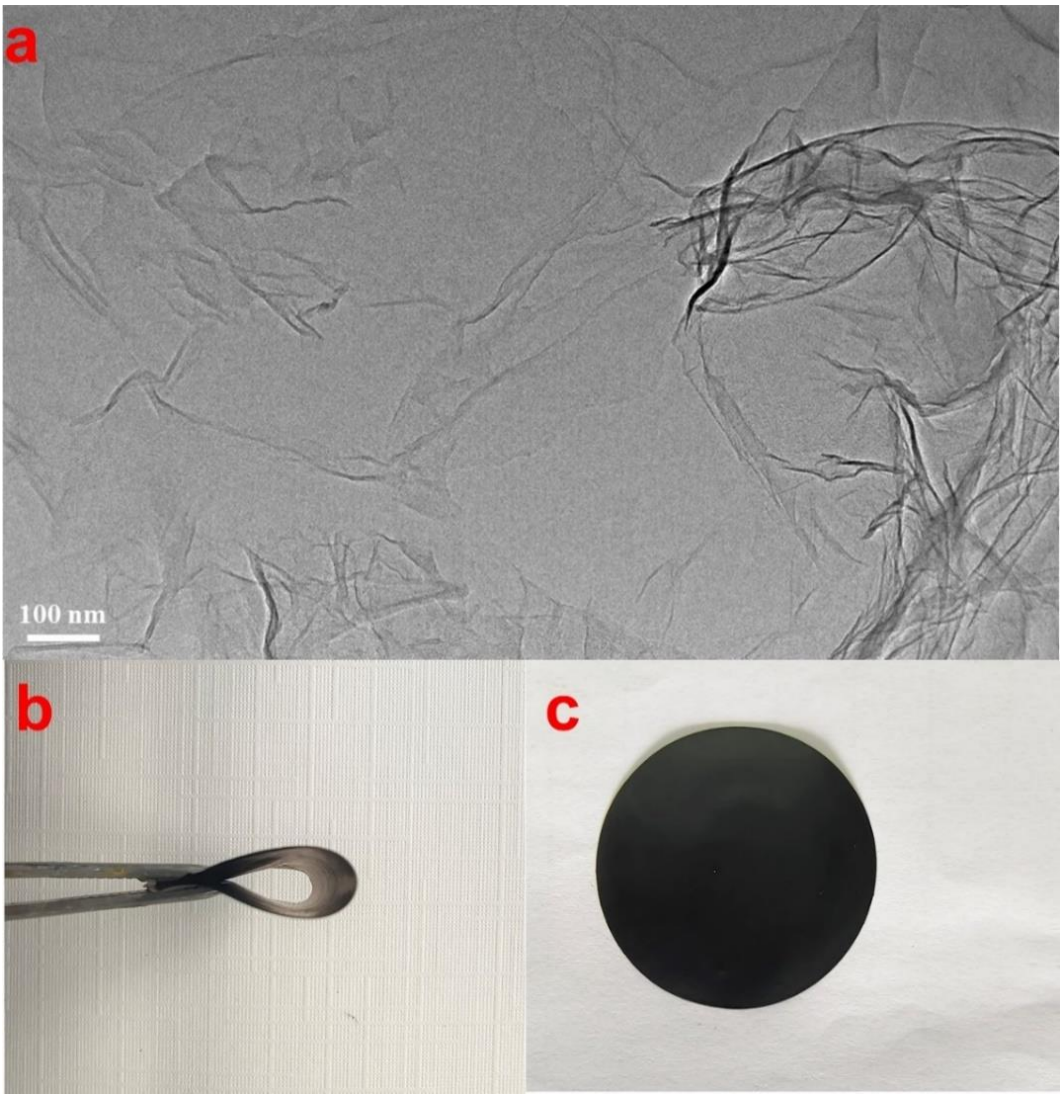


Figure S6. (a) TEM of graphene. (b and c) Digital photographs of PBO/GN sheet.

Table S5. Horizontal and vertical thermal conductivities of graphene composite sheets.

	λ_x/(Wm^{-1}K^{-1})	λ_z/ (Wm^{-1}K^{-1})	λ_x/λ_z
PBZ/GN-III	16.5	0.41	40.24
PBZ/GN-II	31.5	0.5	63
PBZ/GN-I	39.2	0.61	64.3
PBO/GN	47.8	0.71	67.32

Table S6. Interfacial thermal resistance and the factor of TCE of graphene composite sheets.

	PBZ/GN-III	PBZ/GN-II	PBZ/GN-I	PBO/GN
R_{BD}(m^2K/W)	1.01×10^{-6}	5.22×10^{-7}	4.19×10^{-7}	3.02×10^{-7}
The factor of TCE (%)	156.8	255.3	268.0	289.7

Table S7. Average EMI SE and EMI SE coefficient per micron thickness between 8.2 and 12.4 GHz for graphene composite sheets.

	Thickness(μm)	Average EMI SE between 8.2 and 12.4 GHz (dB)	SE/t (dB/μm)
PBO/GN	40.6	48.4	1.19
PBZ/GN-I	42.4	44.1	1.04
PBZ/GN-II	48.3	34.0	0.70
PBZ/GN-III	56.7	23.7	0.42

Table S8. Comparison of EMI shielding property of some reported materials.

Composition	EMI SE (dB)	Thickness (μm)	SE/t (dB/μm)	Ref.
GNP/PDMS	86	2000	0.043	[S1]
GNP/POM	44.7	150	0.298	[S2]
GNS/Cellulose	27.4	33	0.830	[S3]
rGO /PS	45.1	2500	0.018	[S4]
PBZ/GN/Ni	32.9	1000	0.033	[S5]
PVDF/GN/CNT	28.5	2000	0.014	[S6]
GN/Ni paper	85	250	0.340	[S7]
MXene/PVA	40	75	0.533	[S8]
GNPS/glass fiber reinforced ploymers	60	240	0.259	[S9]
GN/PBO	48.4	40.6	1.19	This work

Reference

S1. J. Li, X. Zhao, W. Wu, X. Ji, Y. Lu and L. Zhang, *Chem. Eng. J.*, 2021, **415**, 129054.

S2. J. Li, Y. Wang, T.-N. Yue, Y.-N. Gao, Y.-D. Shi, J.-B. Shen, H. Wu and M. Wang, *Compos. Sci. Technol.*, 2021, **206**, 108681.

S3. L. Li, Z. Ma, P. Xu, B. Zhou, Q. Li, J. Ma, C. He, Y. Feng and C. Liu, *Compos. Part A Appl. Sci. Manuf.*, 2020, **139**, 106134.

S4. D.-X. Yan, H. Pang, B. Li, R. Vajtai, L. Xu, P.-G. Ren, J.-H. Wang and Z.-M. Li, *Adv. Funct. Mater.*, 2015, **25**, 559-566.

S5. S. Zhang, Y. Wang, Q. Ran, Q. Fu and Y. Gu, *React. Funct. Polym.*, 2019, **143**, 104324.

S6. X. Ma, B. Shen, L. Zhang, Y. Liu, W. Zhai and W. Zheng, *Compos. Sci. Technol.*, 2018, **158**, 86-93.

S7. J. Li, L. Huang, Y. Yuan, Y. Li and X. He, *Mater. Sci. Eng. B.*, 2021, **263**, 114893.

S8. Y. Zhang, K. Ruan and J. Gu, *Small,* 2021, **17**, 2101951.

S9. I.U. Din, K. Naresh, R. Umer, K.A. Khan, L.T. Drzal, M. Haq and W.J. Cantwell, *Compos. Part A Appl. Sci. Manuf.*, 2020, **134**, 105901.