

Electronic Supplementary Information

Enhanced Thermal Conductivity of Nanocomposites with MOF-derived Encapsulated
Magnetic Oriented Carbon Nanotube-Grafted Graphene Polyhedra

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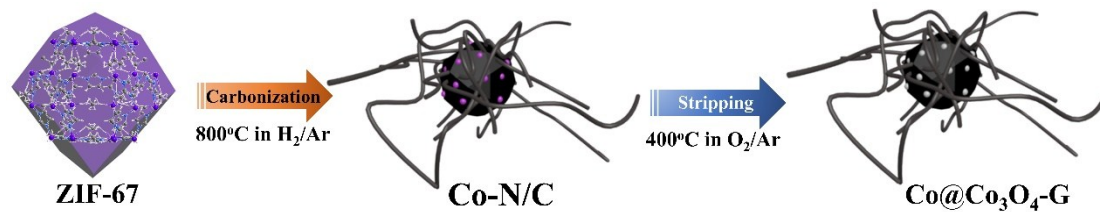


Fig. S1 Schematic illustrations of the preparation process of Co@Co₃O₄-G.

Table. S1 Physical properties of ER/Co@Co₃O₄-G nanocomposites with different Co@Co₃O₄-G volume fractions.

Vol. fraction (Vol%)	Density (g cm ⁻³)	Specific heat capacity (J g ⁻¹ K ⁻¹)	Thermal diffusivity (mm ² s ⁻¹)	Thermal conductivity (W m ⁻¹ K ⁻¹)
	1.05	1.38	0.13	0.19
0	1.02	1.34	0.12	0.17
	1.08	1.36	0.15	0.22
	1.23	1.31	0.19	0.32
1.2	1.19	1.33	0.22	0.35
	1.22	1.28	0.17	0.27
	1.24	1.32	0.34	0.56
2.6	1.26	1.31	0.36	0.60
	1.27	1.30	0.29	0.48
	1.28	1.29	0.80	1.32
4.3	1.29	1.28	0.83	1.37
	1.27	1.26	0.79	1.27
	1.32	1.22	1.04	1.67
6.5	1.35	1.23	1.03	1.72
	1.37	1.25	0.91	1.56
	1.38	1.21	1.26	2.11
8.7	1.37	1.18	1.39	2.24
	1.37	1.16	1.30	2.07

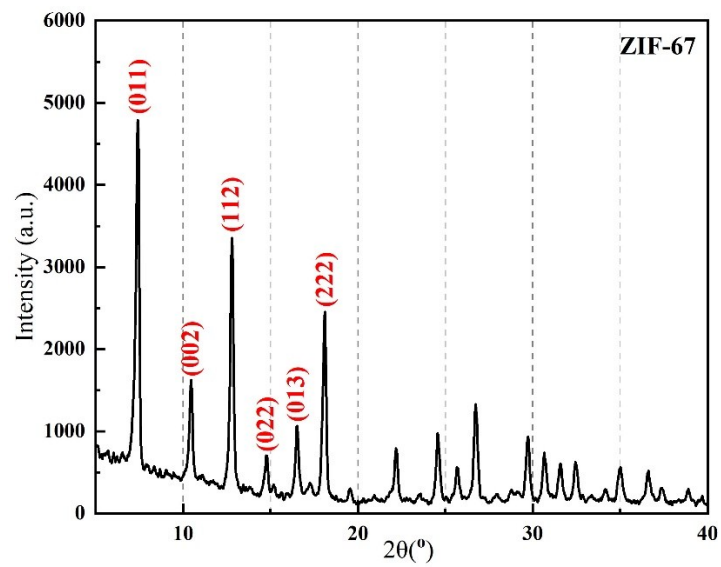


Fig. S2 The XRD of ZIF-67.

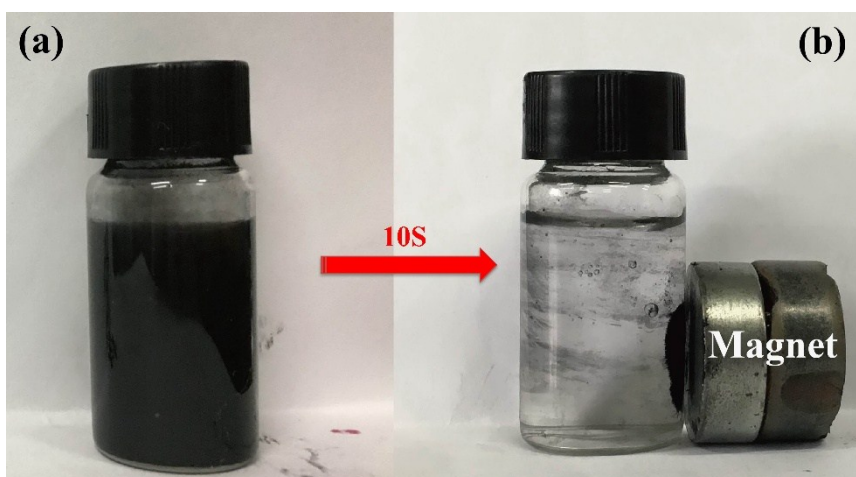


Fig. S3 Images of Co@Co₃O₄-G dispersion in a water solution (a) and its response to external magnetic field when a magnet is placed near the dispersion (b).

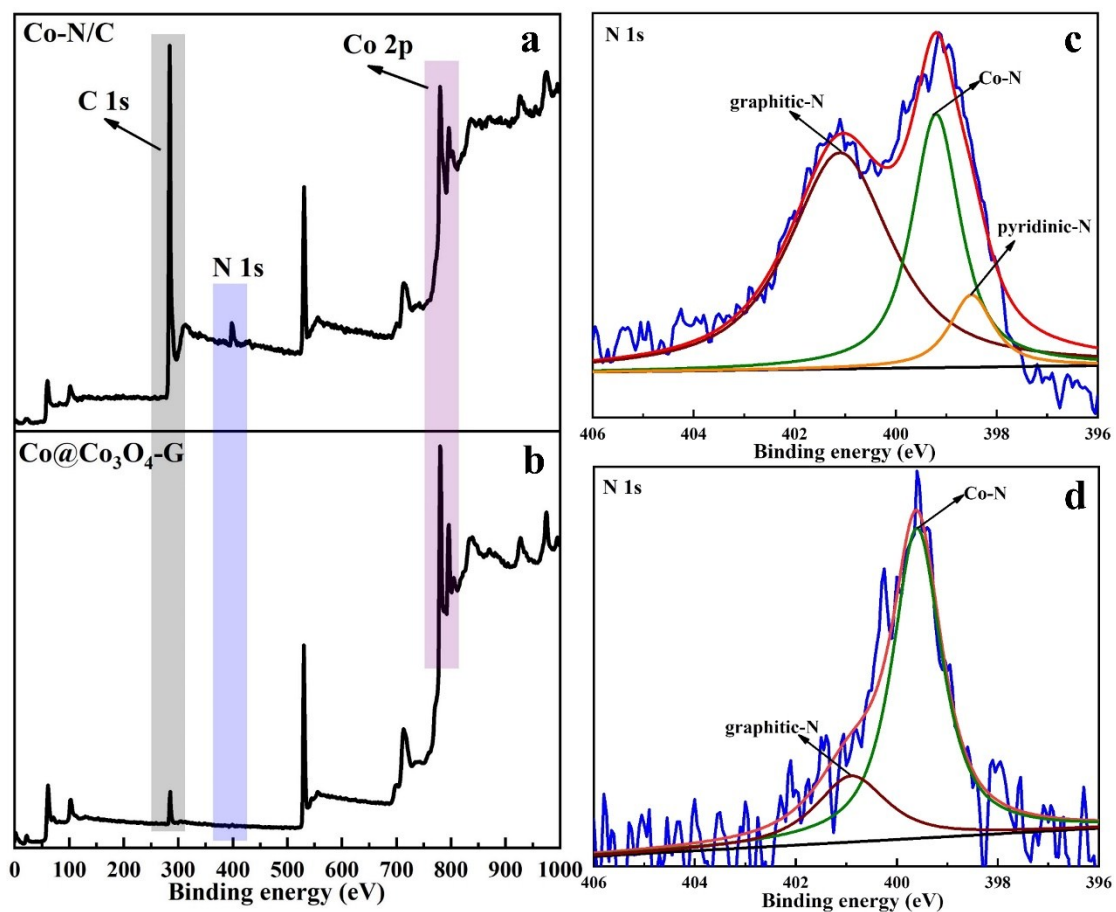


Fig. S4 Full survey XPS of Co-N/C (a) and Co@Co₃O₄-G(b), N 1s of Co-N/C (c) and Co@Co₃O₄-G(d).

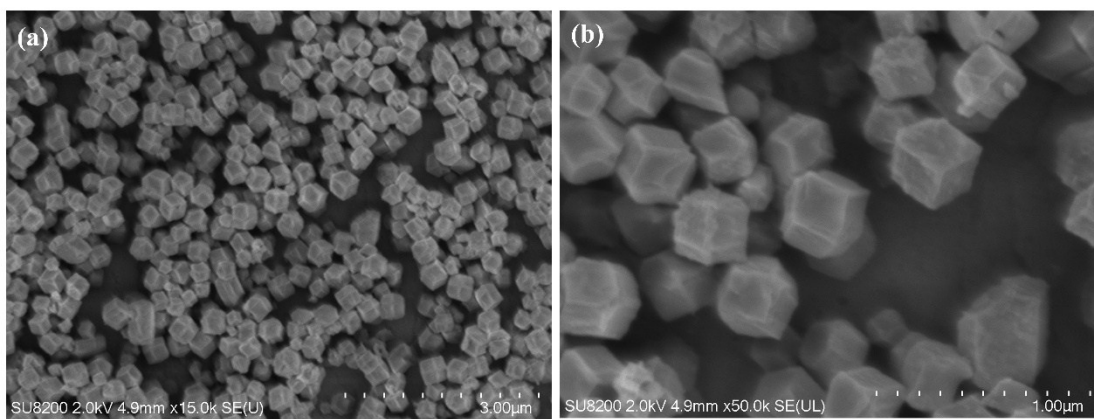


Fig. S5 SEM image of ZIF-67.

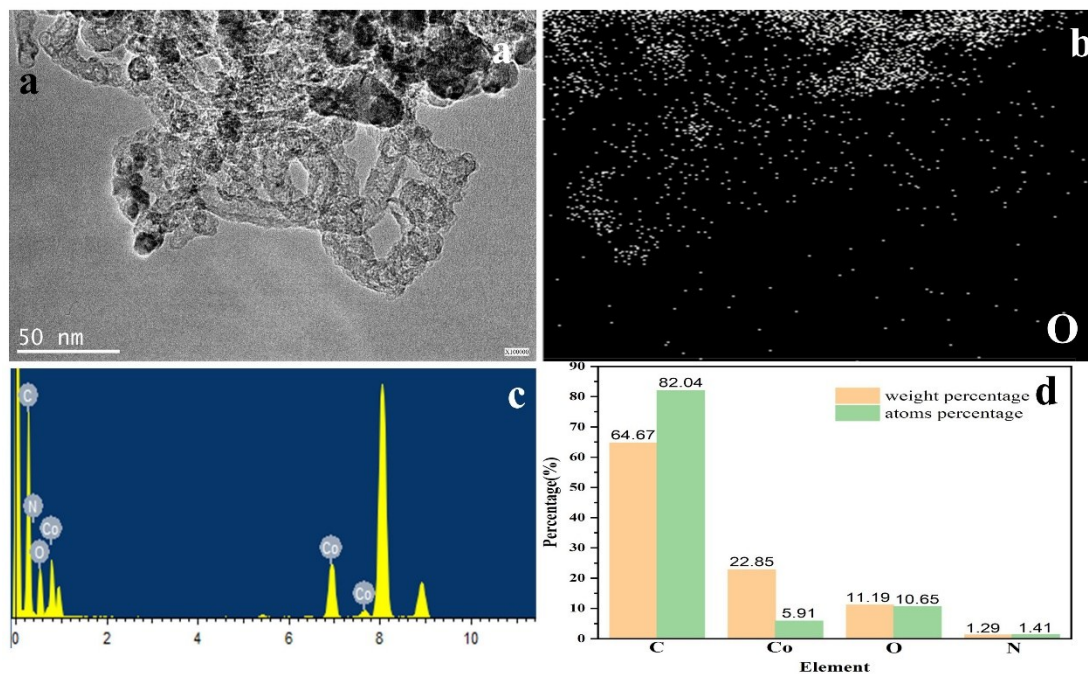


Fig. S6 (a) TEM of Co@Co₃O₄-G. (b) TEM-EDS elemental mapping of O atom in Co@Co₃O₄-G. (c) The corresponding EDS spectrum of Co@Co₃O₄-G. (d) The weight and atoms percentage of C, Co, O and N in the Co@Co₃O₄-G.

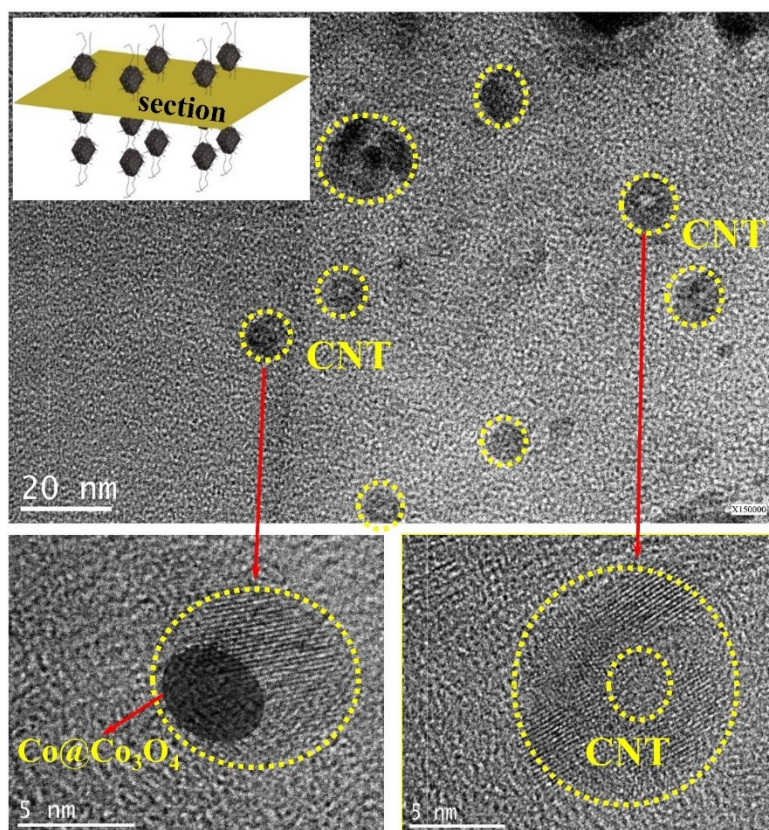


Fig. S7 The HRTEM of ultra-thin section of ER/Co@Co₃O₄-G with 8.7 vol% loading of Co@Co₃O₄-G.

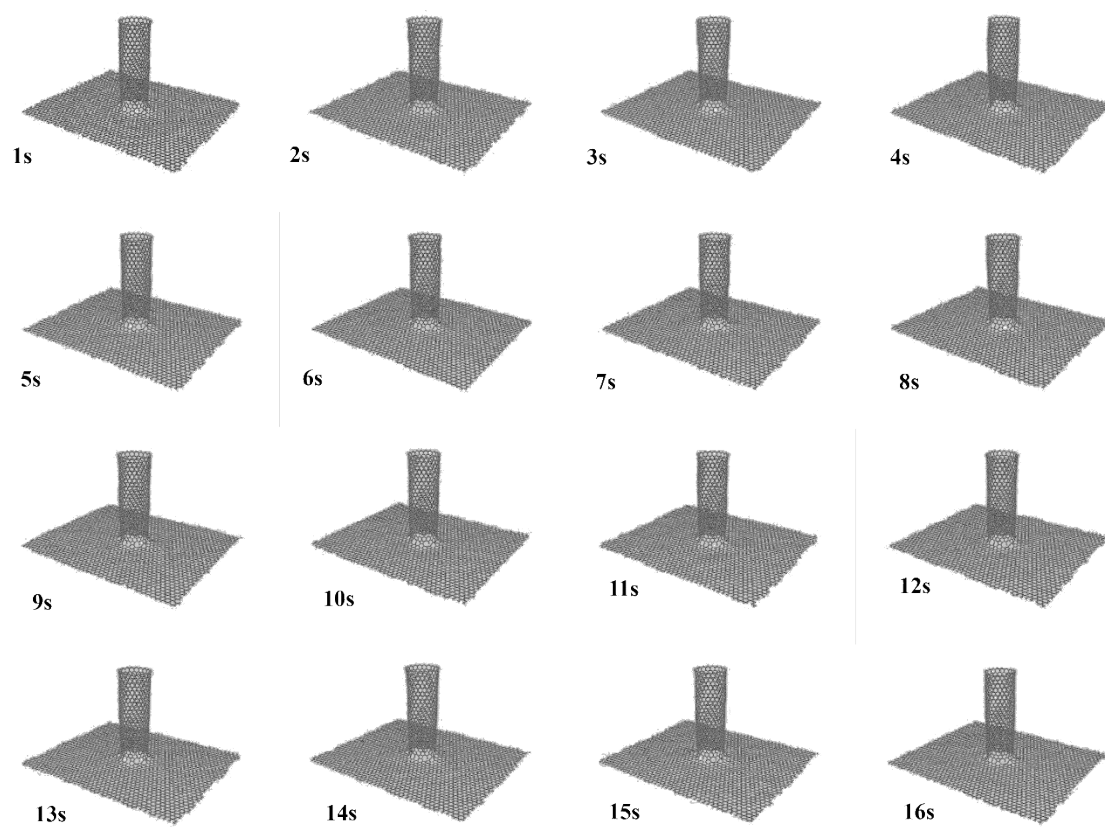


Fig. S8 the atomic vibrogram of G/CNT with time in EMD simulate.

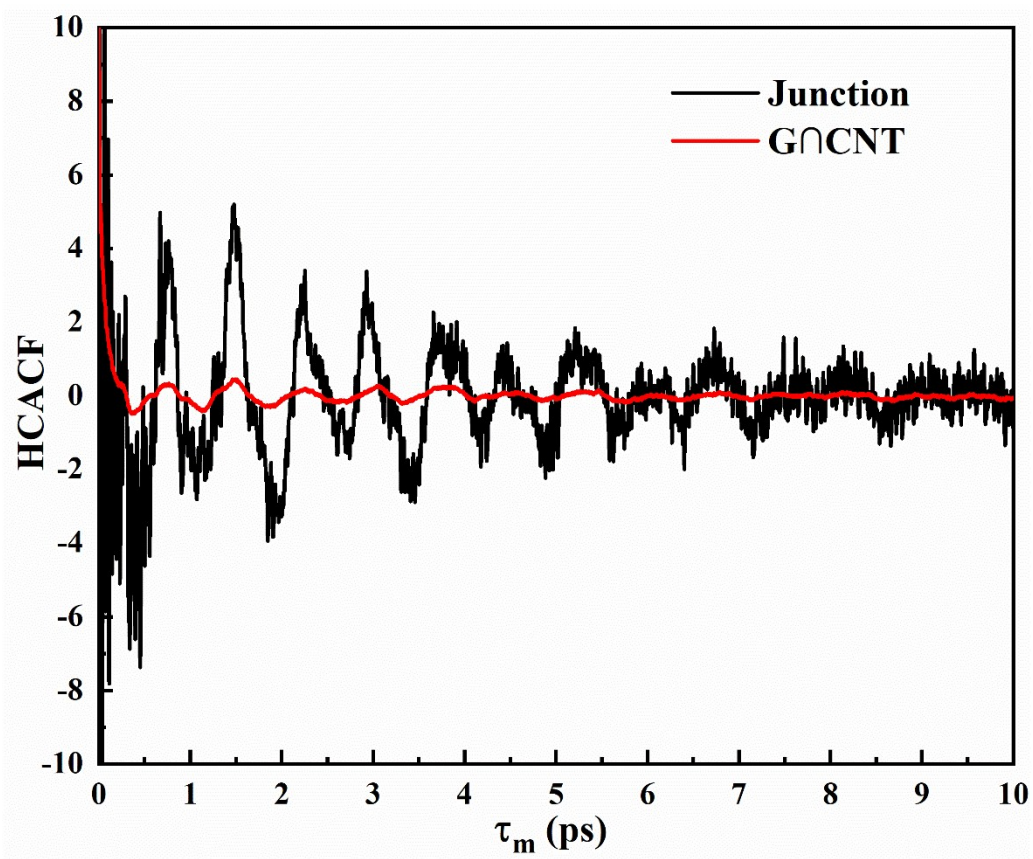


Fig. S9 The change of HCACF of the GNCNT and the junction with time.

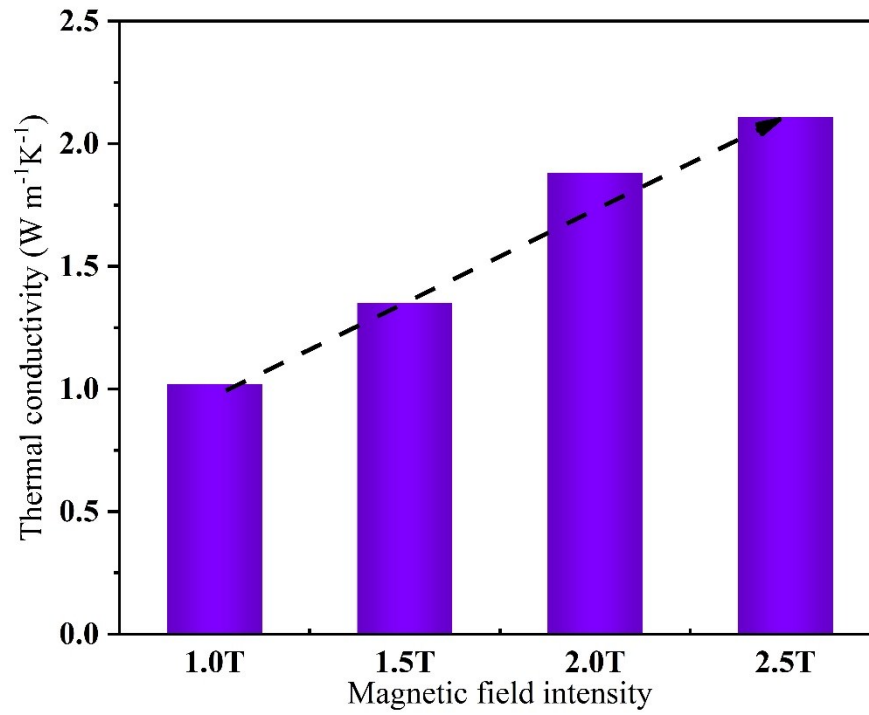


Fig. S10 The thermal conductivity of nanocomposites with 8.7 vol% loading with the change of magnetic field intensity

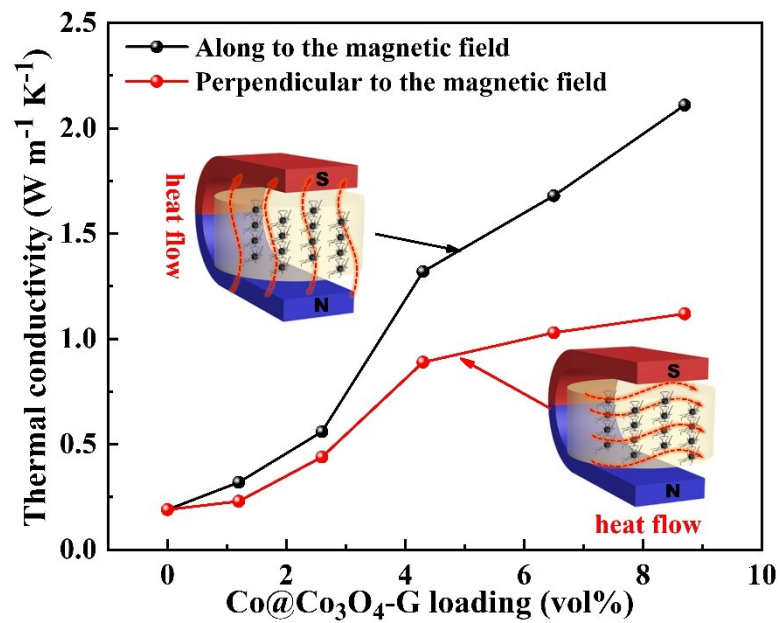


Fig. S11 the thermal conductivity of ER/Co@Co₃O₄-G with different loading along different heat flow.

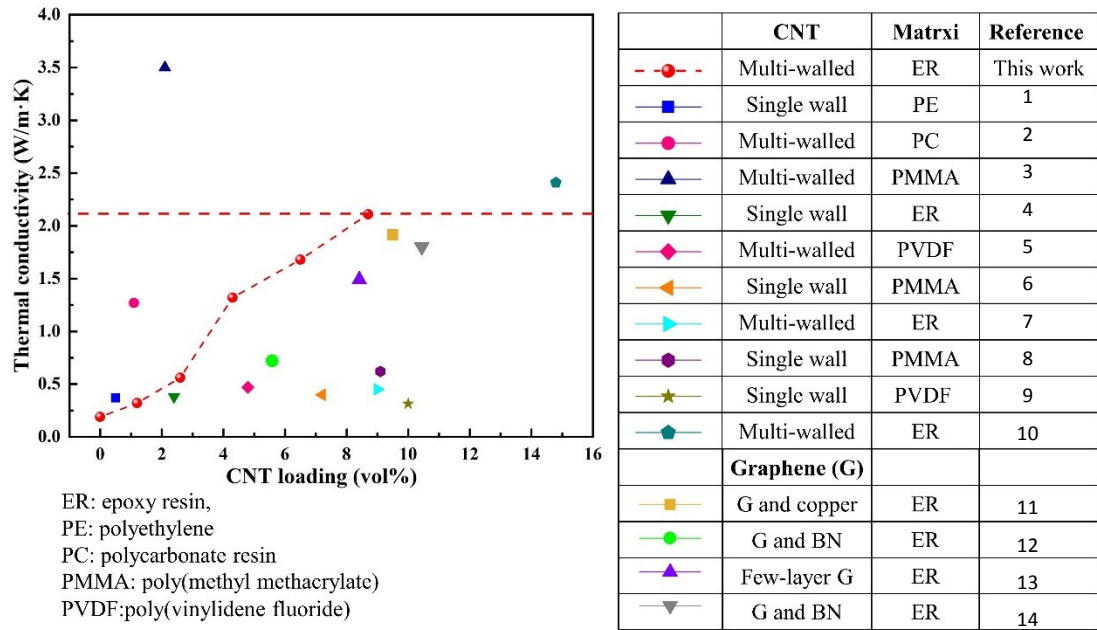


Fig. S12 Thermal conductivity of the ER/Co@Co₃O₄-G nanocomposites with different loading and other CNT and graphene-based composites reported in previous work.

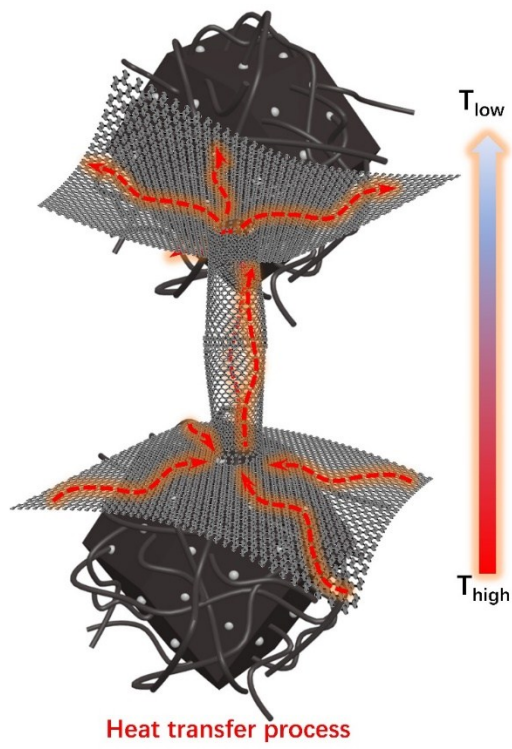


Fig. S13 The thermal conductive mechanism of Co@Co₃O₄-G.

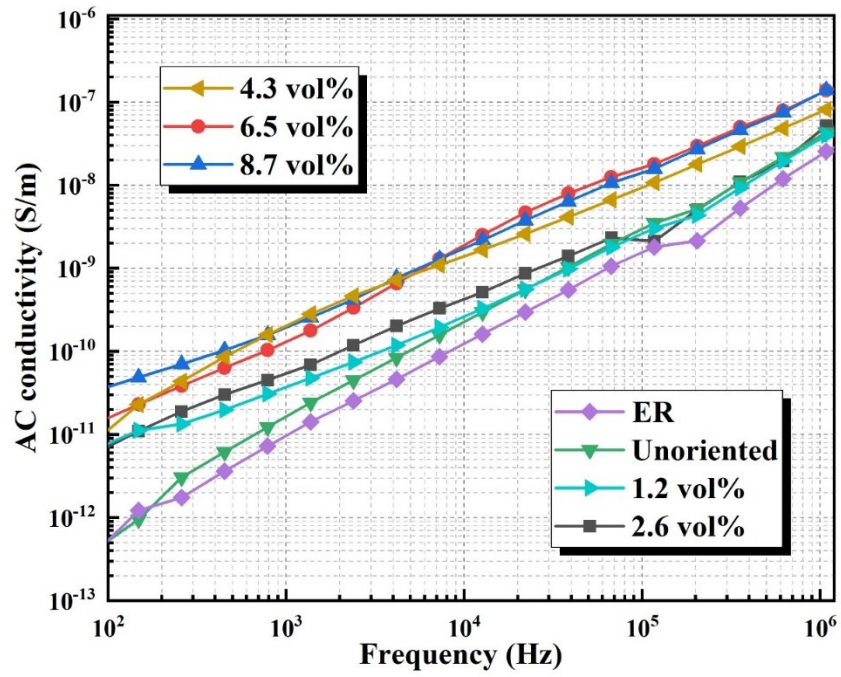


Fig. S14 Frequency dependence of electric conductivity of the nanocomposites with different loading.

Reference

1. R. Haggemueller, C. Guthy, J. R. Lukes, J. E. Fischer and K. I. Winey, *macromolecules*, 2007, **40**, 2417-2424.
2. H. S. Kim, J. U. Jang, J. Yu and S. Y. Kim, *Compos. Part B-Eng.*, 2015, **79**, 505-512.
3. W. T. Hong and N. H. Tai, *Diam. Relat. Mater.*, 2008, **17**, 1577-1581.
4. F. Du, C. Guthy, T. Kashiwagi, J. E. Fischer and K. I. Winey, *J. Polym. Sci. Pol. Phys.*, 2006, **44**, 1513-1519.
5. W. B. Zhang, Z. X. Zhang, J. H. Yang, T. Huang, N. Zhang, X. T. Zheng, Y. Wang and Z. W. Zhou, *Carbon*, 2015, **90**, 242-254.
6. P. Bonnet, D. Sireude, B. Garnier and O. Chauvet, *Appl. Phys. Lett.*, 2007, **91**, 201910.
7. F. H. Gojny, M. H. G. Wichmann, B. Fiedler, I. A. Kinloch, W. Bauhofer, A. H. Windle and K. Schulte, *Polymer*, 2006, **47**, 2036-2045.
8. C. Guthy, F. Du, S. Brand, K. I. Winey and J. E. Fischer, *J. Heat Transfer*, 2007, **129**, 1096-1099.
9. Y. Xu, G. Ray and B. Abdel-Magid, *Compos. Part A-Appl. S.*, 2006, **37**, 114-121.
10. A. M. Marconnet, N. Yamamoto, M. A. Panzer, B. L. Wardle and K. E. Goodson, *ACS Nano*, 2011, **5**, 4818-4825.
11. Z. Barani, A. Mohammadzadeh, A. Geremew, C. Y. Huang, D. Coleman, L. Mangolini, F. Kargar and A. A. Balandin, *Adv. Funct. Mater.*, 2019, 1904008.
12. F. Kargar, Z. Barani, R. Salgado, B. Debnath, J. S. Lewis, E. Aytan, R. K. Lake and A. A. Balandin, *ACS Appl. Mater. Inter.*, 2018, **10**, 37555-37565.
13. F. Kargar, Z. Barani, M. Balinskiy, A. S. Magana, J. S. Lewis and A. A. Balandin, *Adv. Electron. Mater.*, 2019, **5**, 1800558.
14. J. S. Lewis, Z. Barani, A. S. Magana, F. Kargar and A. A. Balandin, *Mater. Res. Express*, 2019, **6**, 085325.