

**Simple preparation of 1D hierarchical magnetic CNTs/hollow porous
macroscopic carbon fiber composites for efficient microwave absorption**

Minghang Yang ^{a,b,c}, Yu Deng ^{a,b,c}, Mingguang Zhang ^{a,b,c}, Shuaining Zhou ^{a,b,c},
Cheng Liu ^{a,b,c*}, Xigao Jian ^{a,b,c}, Yousi Chen ^{a,b,c*}

^a State Key Laboratory of Fine Chemicals, Frontier Science Center for Smart Materials, Department of Polymer Science & Materials, School of Chemical Engineering, Dalian University of Technology, Dalian, 116024, China.

^b Technology Innovation Center of High Performance Resin Materials, Liaoning Province.

^c Dalian Basalt Fiber Resin Matrix Composite Engineering Research Center, Dalian.

* Corresponding author. E-mail address: liuch1115@dlut.edu.cn (L. Cheng),

* Corresponding author. E-mail address: chenyouisi@dlut.edu.cn (Y. S. Chen).

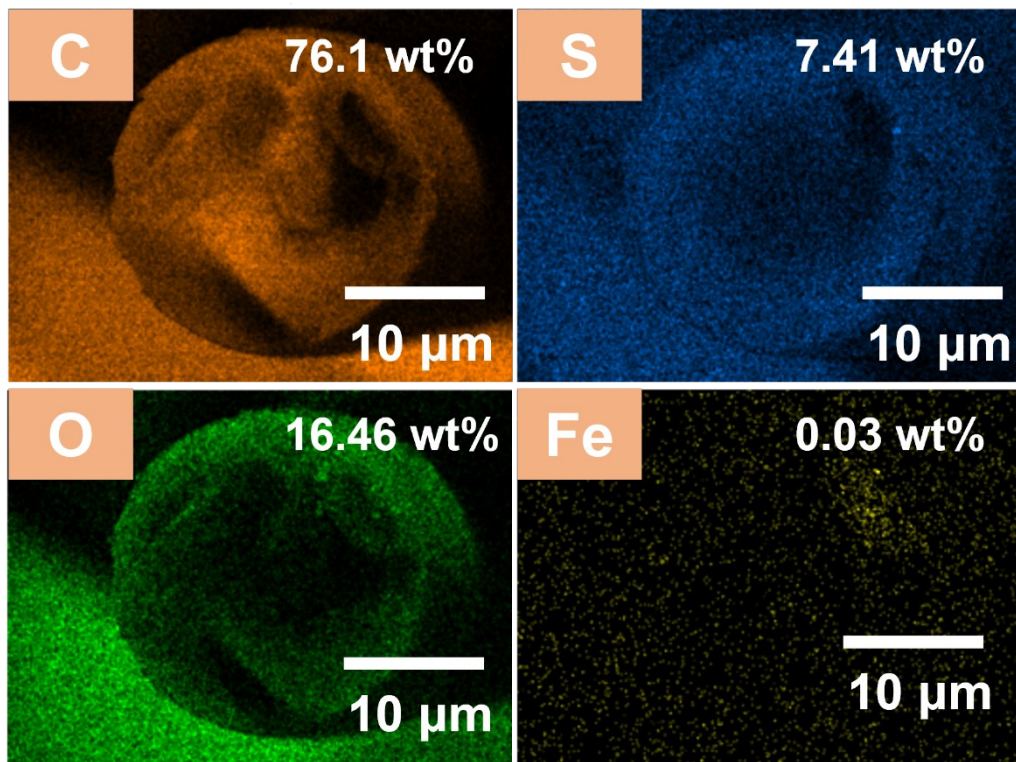


Fig S1.EDS images of Fe@SFs-1

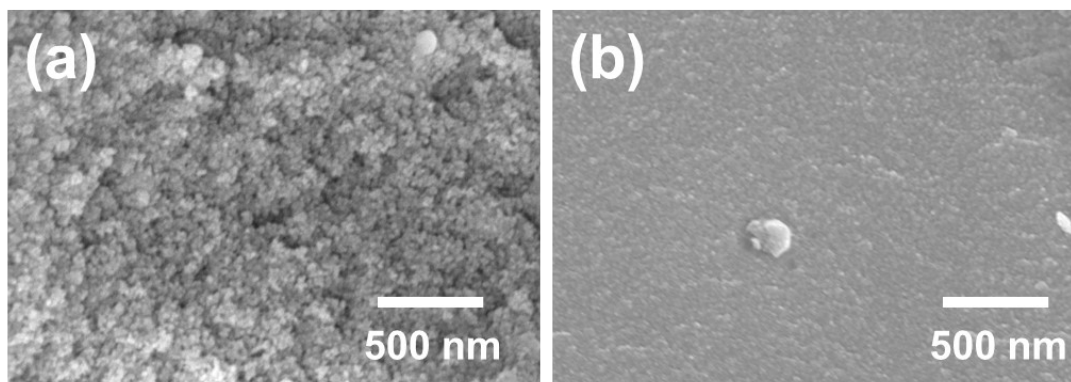


Fig S2. High magnification SEM images of (a) the porous layer in the cross section of HPCFs-1 and (b) the cross-section of HPCFs-3

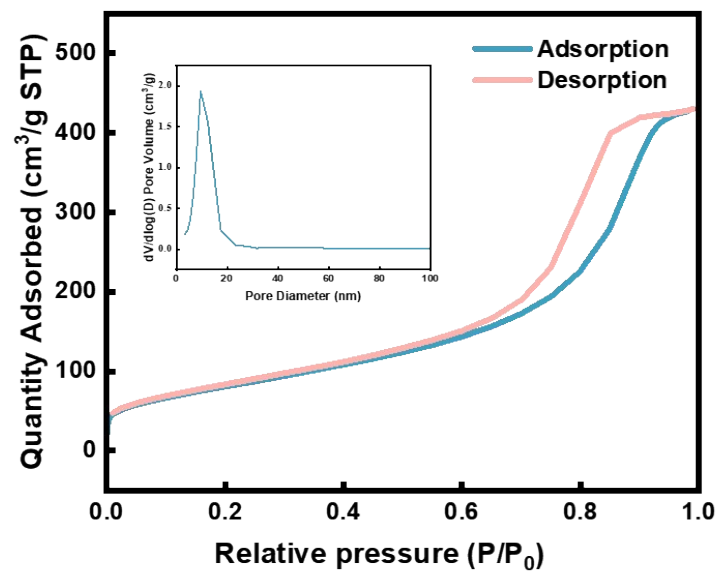


Fig S3. N₂ adsorption-desorption isotherms and pore size curves of HPCFs-1

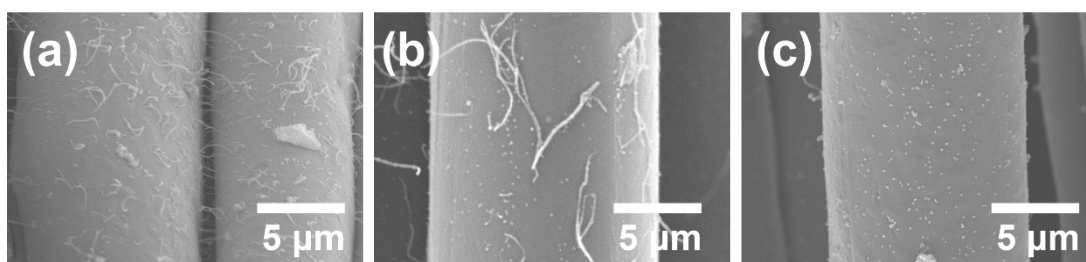


Fig S4. SEM images of surface of (a) CNTs@HPCFs-1, (b) CNTs@HPCFs-2, and (c) CNTs@HPCFs-3

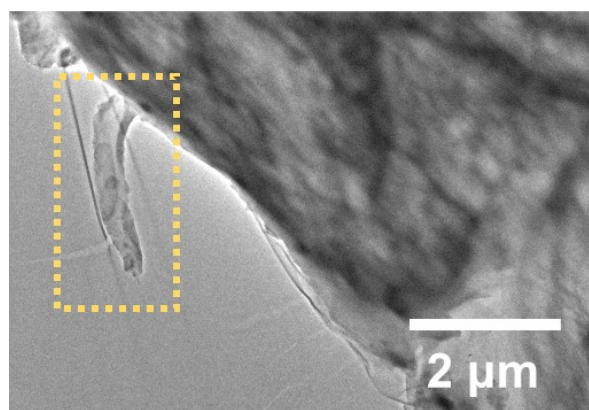


Fig S5. TEM images of surface of CNTs@HPCFs-1

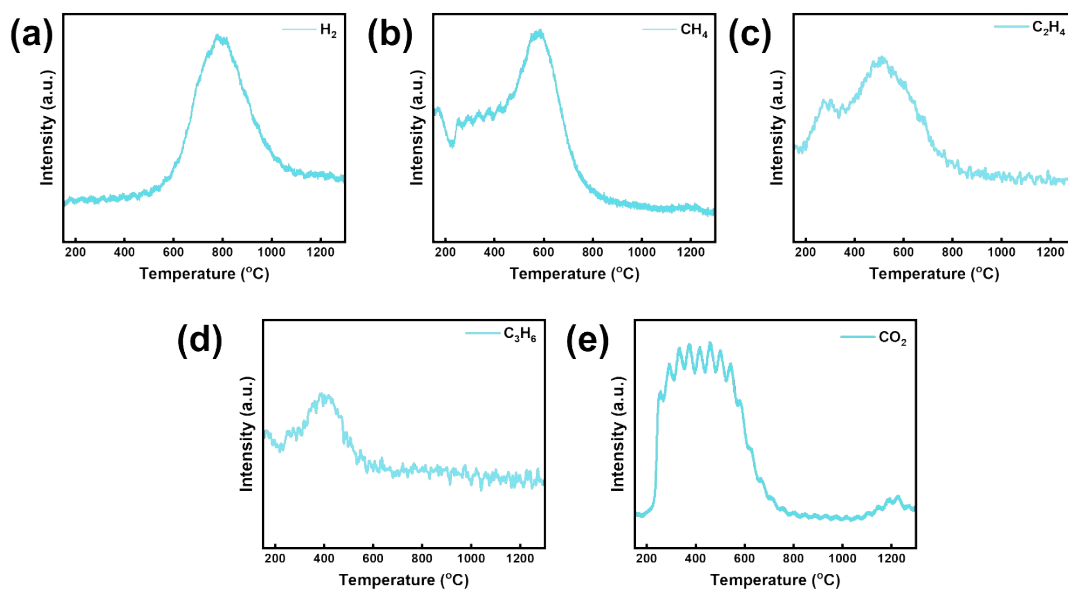


Fig S6. TGA-MS spectra of Fe@SFs-3

Table S1. Surface elemental content of fibers measured by XPS

Sample	C (Atomic %)	O (Atomic %)	S (Atomic %)	Fe (Atomic %)
HPCFs-1	95.88	3.87	0.26	-
CNTs@HPCFs-1	78.44	17.88	3.15	0.44

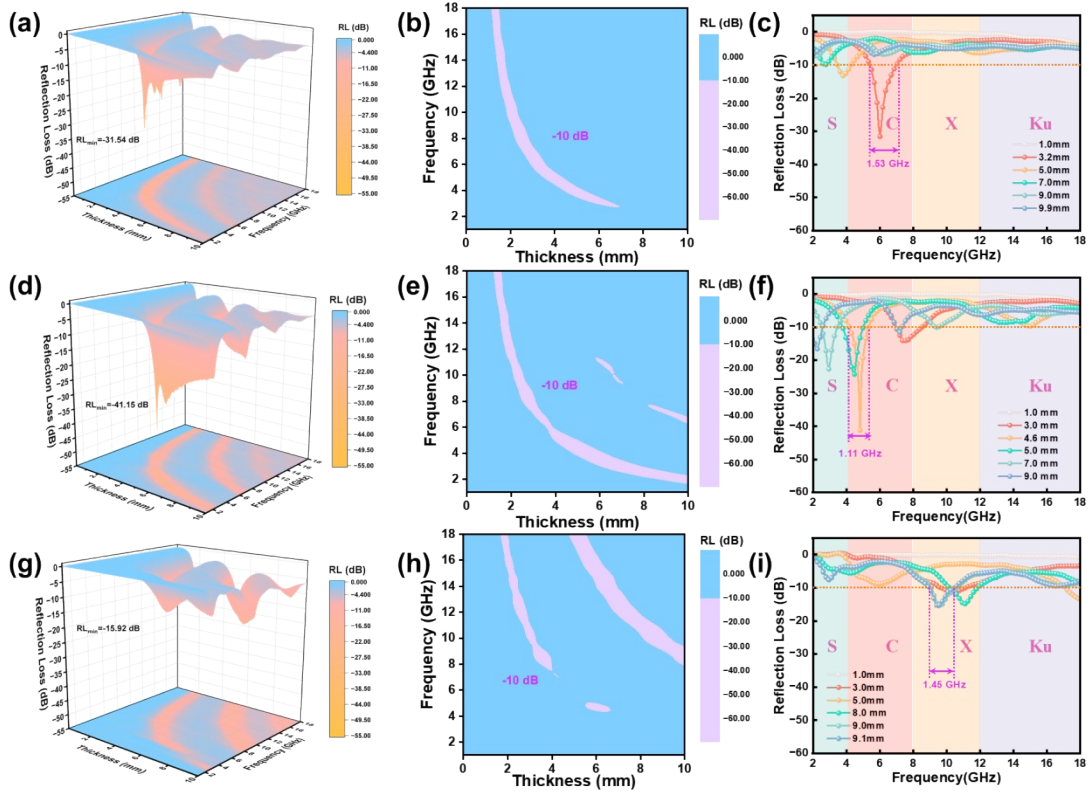


Fig S7. Calculated 3D/2D RL and contour maps of the RL of HPCFs-1 (a, b, c), HPCFs-2 (d, e, f), and HPCFs-3 (g, h, i)

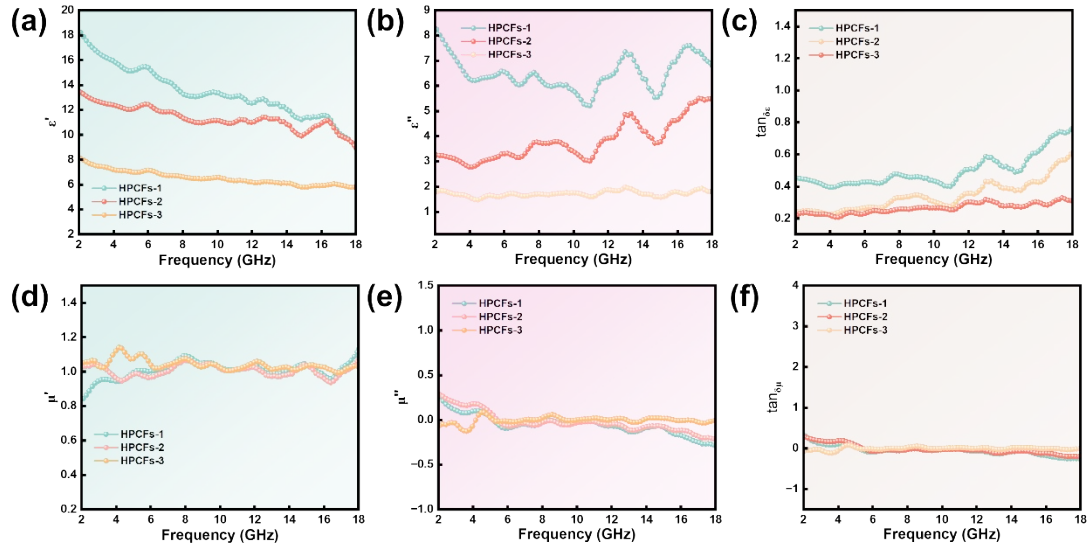


Fig S8. (a) Real permittivity (ϵ'), (b) imaginary permittivity (ϵ''), (c) dielectric loss tangent ($\tan\delta_\epsilon$), (d) Real permeability (μ'), (e) imaginary permeability (μ''), and (f) magnetic loss tangent ($\tan\delta_\mu$) of HPCFs

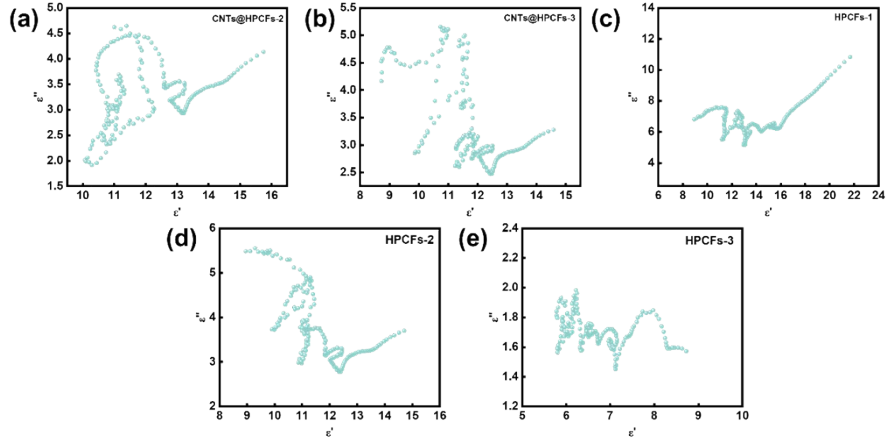


Fig S9. Cole-Cole curves of (a) CNTs@HPCFs-2, (b) CNTs@HPCFs-3, (c) HPCFs-1, (d) HPCFs-2, and (e) HPCFs-3

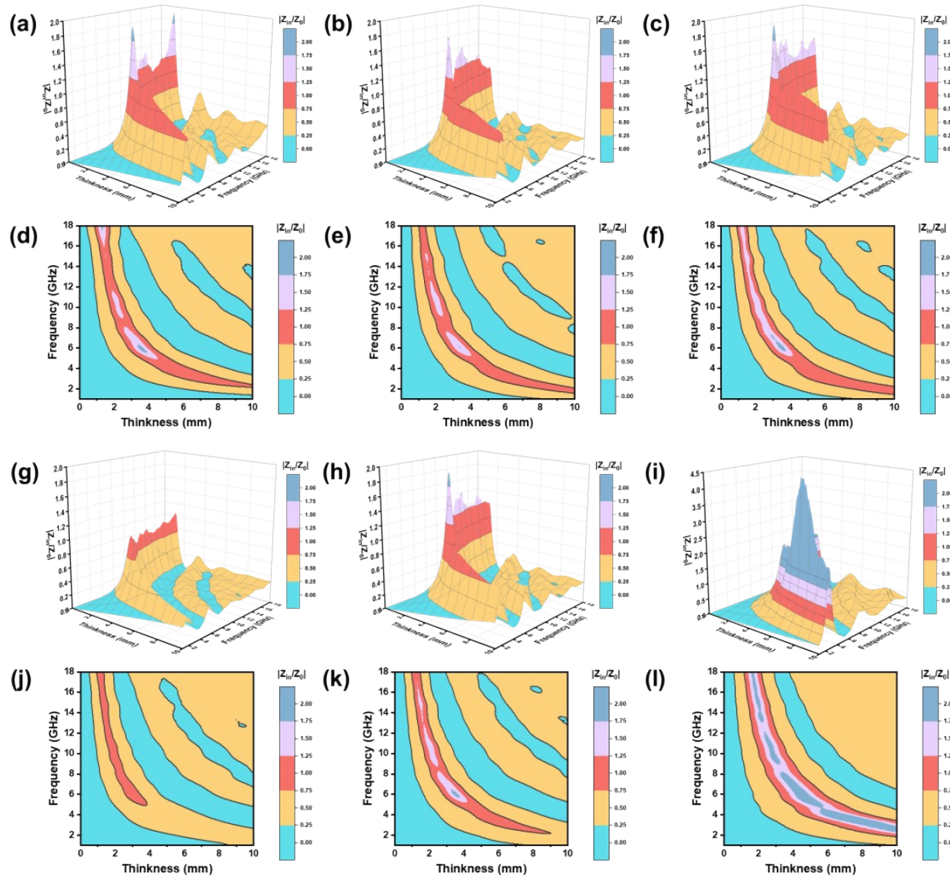


Fig S10. 3D and 2D contour maps of $|Z_{in}/Z_0|$ for (a, d) CNTs@HPCFs-1, (b, e) CNTs@HPCFs-2, (c, f) CNTs@HPCFs-3, (g, j) HPCFs-1, (h, k) HPCFs-2, and (i, l)

HPCFs-3

Table S2. Comparison with reported microwave properties of carbon materials

Microwave absorber	Thickness (mm)	RL _{min} (dB)	Effective bandwidth (GHz)	Reference
Carbon microtubes	2.06	-30.75	6.78	[1]
PAN-based carbon fiber	2.5	-39.90	1.30	[2]
CNTs/CF	1.18	-56.11	3.6	[3]
GO/CF	4.65	-57.3	4.88	[4]
Porous carbon	2.2	-30.46	5.44	[5]
Porous carbon nanosheets	1.70	-29.50	7.2	[6]
Porous carbon frameworks	1.75	-40.4	3.48	[7]
CNTs@HPCFs	1.90	-54.03	4.08	This work

Reference

[1] T. Ning, Q. Li, Q. Ren, J. Wang, Y. Sun and P. Zhang, Kapok fibers-derived carbon microtubes as efficient electromagnetic wave absorption materials, *Ceramics International*, 2023, 49, 29339-29347.

[2] S. Liu, J. Wang, B. Zhang, X. Su, X. Chen, Y. Chen, H. Yang, Q. Wu and S. Yang, Transformation of traditional carbon fibers from microwaves reflection to efficient

- absorption via carbon fiber microstructure modulation, *CARBON*, 2024, 219,
- [3] C. Wang, Y. Wang, H. Jiang, H. Tan and D. Liu, Continuous in-situ growth of carbon nanotubes on carbon fibers at various temperatures for efficient electromagnetic wave absorption, *CARBON*, 2022, 200, 94-107.
- [4] Y. Cao, Z. Cheng, R. Wang, X. Liu, T. Zhang, F. Fan and Y. Huang, Multifunctional graphene/carbon fiber aerogels toward compatible electromagnetic wave absorption and shielding in gigahertz and terahertz bands with optimized radar cross section, *CARBON*, 2022, 199, 333-346.
- [5] S. Wei, Z. Shi, W. Wei, H. Wang, D. Dastan, M. Huang, J. Shi and S. Chen, Facile preparation of ultralight porous carbon hollow nanoboxes for electromagnetic wave absorption, *Ceramics International*, 2021, 47, 28014-28020.
- [6] C. Gao, H. Zhang, D. Zhang, F. Gao, Y. Liu, X. Chen, D. Wu, M. Terrones and Y. Wang, Sustainable synthesis of tunable 2d porous carbon nanosheets toward remarkable electromagnetic wave absorption performance, *Chemical Engineering Journal*, 2023, 476,
- [7] Y. Mao, K. Liu, Y. Sheng, J. Liu, S. Fu and C. Tang, Hierarchical porous carbon frameworks derived from *juncus effusus* biomass with robust electromagnetic wave absorption properties, *Journal of Materials Chemistry C*, 2024, 12, 4442-4452.