

Supporting Information for

## Hierarchical Magnetic Network Constructed by CoFe Nanoparticles Suspended within “Tubes on Rods” Matrix Toward Enhanced Microwave Absorption

Chunyang Xu<sup>1</sup>, Lei Wang<sup>1</sup>, Xiao Li<sup>1</sup>, Xiang Qian<sup>1</sup>, Zhengchen Wu<sup>1</sup>, Wenbin You<sup>1</sup>, Ke Pei<sup>1</sup>, Gang Qin<sup>1</sup>, Qingwen Zeng<sup>1</sup>, Ziqi Yang<sup>1</sup>, Chen Jin<sup>1</sup>, Renchao Che<sup>1</sup>, \*

<sup>1</sup>Laboratory of Advanced Materials, Department of Materials Science and Collaborative Innovation Center of Chemistry for Energy Materials (iChem), Fudan University, Shanghai 200438, P. R. China

\*Corresponding author. E-mail: [rcche@fudan.edu.cn](mailto:rcche@fudan.edu.cn) (Renchao Che)

### Supplementary Figures and Table

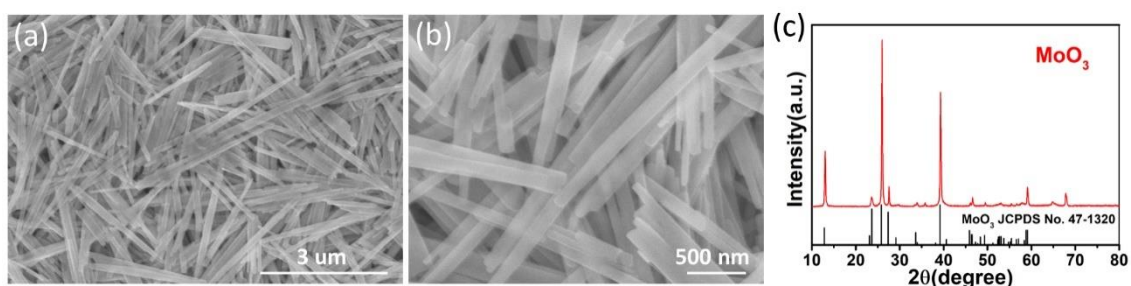


Fig. S1 a, b SEM images and c XRD patterns of MoO<sub>3</sub> samples

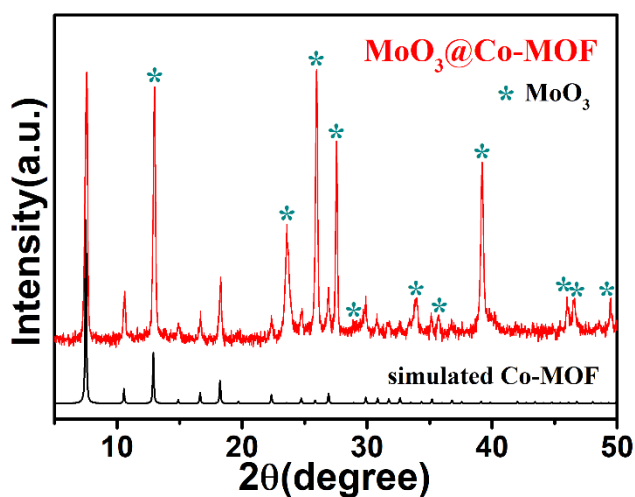


Fig. S2 XRD patterns of MoO<sub>3</sub>@Co-MOF

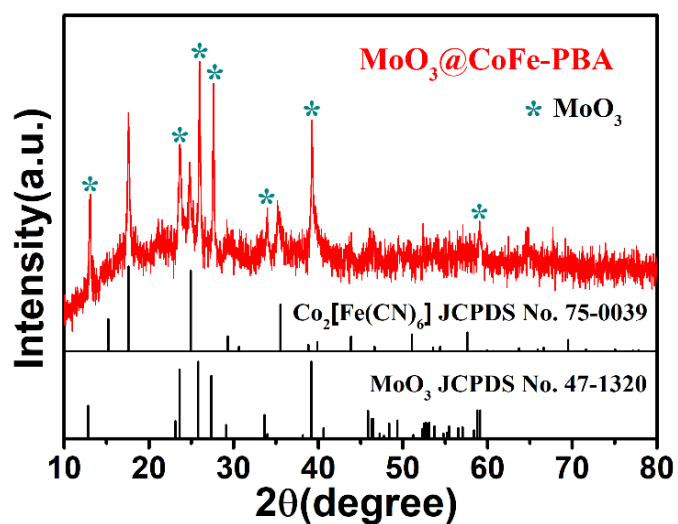


Fig. S3 XRD patterns of  $\text{MoO}_3@hollow\text{-CoFe-PBA}$  samples

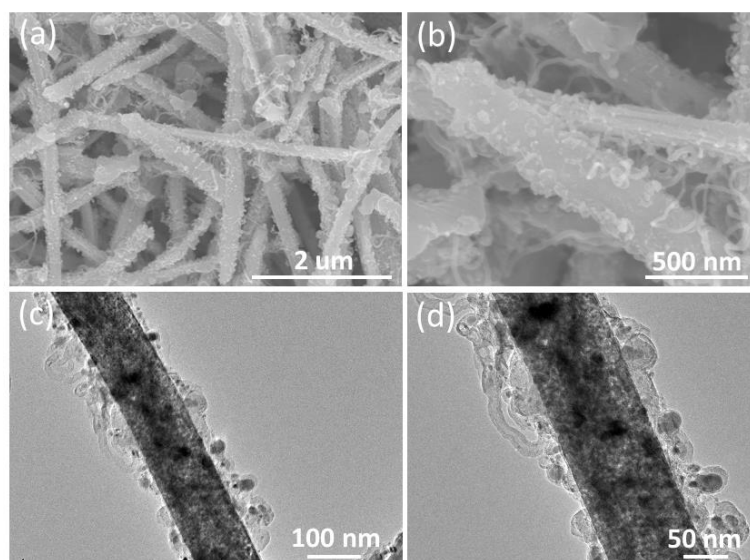


Fig. S4 a, b SEM c, d TEM images of  $\text{Mo}_2\text{N}@Co/CNT$  samples

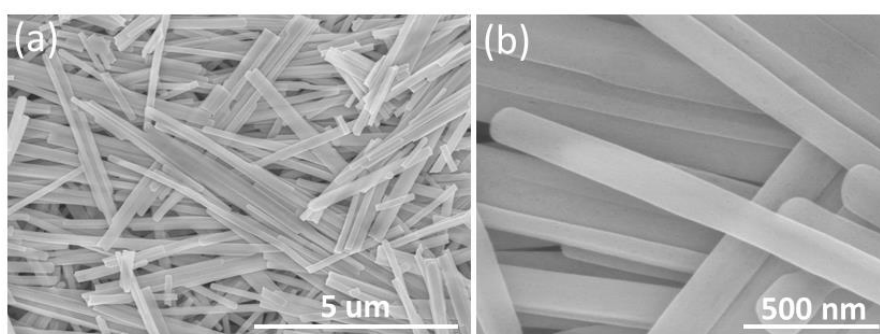
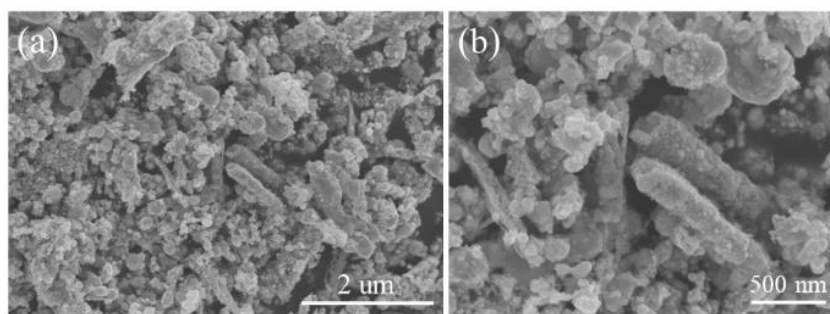
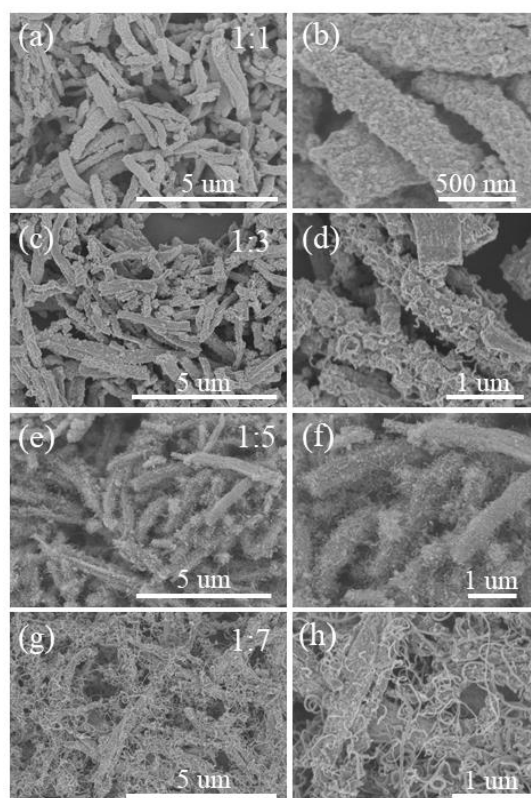


Fig. S5 a, b SEM images of  $\text{Mo}_2\text{N}$  samples

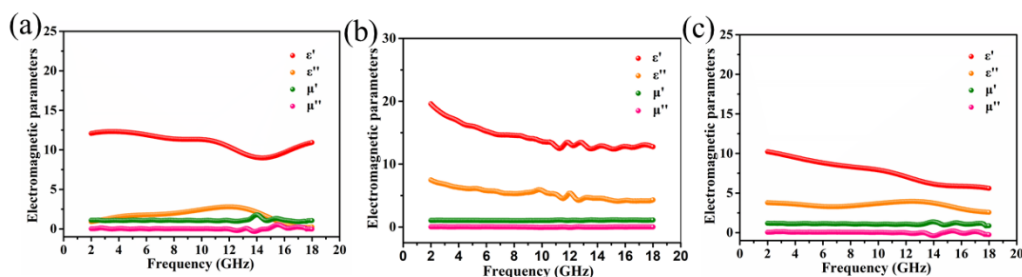


**Fig. S6 a, b** SEM images of samples calcined at 700 °C. We have also prepared the sample obtained under 700 °C. As displayed in Fig. S6,  $\text{MoO}_3$ @hollow-CoFe-PBA samples were unstable at 700 °C and decomposed into irregular structure. Even some samples could retain rod-like structure, there were no CNTs on the surface.

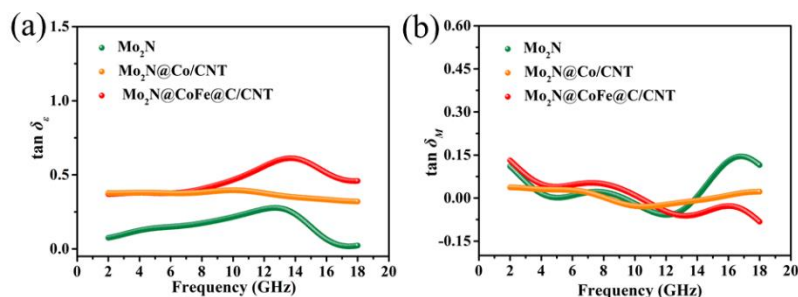


**Fig. S7** SEM images of samples calcined under different weight ratio of  $\text{MoO}_3$ @hollow-CoFe-PBA to melamine: **a-b** 1:1, **c-d** 1:3, **e-f** 1:5, **g-h** 1:7

We have prepared samples under different weight ratio of  $\text{MoO}_3$ @hollow-CoFe-PBA to melamine, including 1:1, 1:3, 1:5, and 1:7. Under the carbonization of  $\text{MoO}_3$ @hollow-CoFe-PBA with the same weight of melamine (1:1), there is no CNTs produced on the  $\text{Mo}_2\text{N}$  rod due to less melamine as carbon sources. Increase the content of melamine (1:3), few CNTs can be seen on the  $\text{Mo}_2\text{N}$  rod. When the weight ratio of  $\text{MoO}_3$ @hollow-CoFe-PBA to melamine is 1:5, a large number of CNTs are produced and deposited on the core of  $\text{Mo}_2\text{N}$  rod, forming  $\text{Mo}_2\text{N}$ @CoFe@C/CNT core-shell structure. Increasing the weight ratio to 1:7, too many CNTs are produced and core-shell structure cannot be maintained.



**Fig. S8** Electromagnetic parameters of **a** Mo<sub>2</sub>N, **b** Mo<sub>2</sub>N@Co/CNT and **c** Mo<sub>2</sub>N@CoFe@C/CNT samples



**Fig. S9** **a** Dielectric loss tangent and **b** magnetic loss tangent of Mo<sub>2</sub>N, Mo<sub>2</sub>N@Co/CNT, and Mo<sub>2</sub>N@CoFe@C/CNT samples

**Table S1** Microwave absorption performance of metal/C absorbents and molybdenum-based materials in previous references and this work.

Absorbents	Thickness (mm)	Maximum RL (dB)	EAB (GHz)	Refs.
FeCo/C/BaTiO <sub>3</sub>	2.0	-41.7	4.2	[S1]
NiFe@C	2.8	-51	3.97	[S2]
Co/NPC@Void@CI	2.2	-49.2	6.72	[S3]
MOF (Fe)/PANI	2.0	-41.4	5.5	[S4]
MWCNT/Fe	4.3	-39.0	2.7	[S5]
Fe/C	3.0	-36.0	0.9	[S6]
Fe/C	2.0	-22.6	5.3	[S7]
Co/N-C NF	2.0	-41.7	4.2	[S8]
CoZn@NCNTHS/G	1.5	-47.3	4.0	[S9]
CMT@CNT/Co	2.0	-52.2	5.1	[S10]
C@MoO <sub>2</sub> /G	1.5	-35.4	4.5	[S11]
Mo <sub>2</sub> C@C	1.9	-48.0	4.1	[S12]
Mo <sub>2</sub> C/C NCs	2.6	-49.2	4.6	[S13]
Mo <sub>2</sub> N@CoFe@C/CNT	2.0	-53.5	5.0	This work

## Supplementary References

- [S1] J. Jiang, D. Li, D. Geng, J. An, J. He et al., Microwave absorption properties of core double-shell FeCo/C/BaTiO<sub>3</sub> nanocomposites. *Nanoscale* **6**, 3967-3971 (2014). <https://doi.org/10.1039/C3NR04087A>
- [S2] Z. Yang, H. Lv, R. Wu, Rational construction of graphene oxide with mof-derived porous NiFe@C nanocubes for high-performance microwave attenuation. *Nano Res.* **9**, 3671-3682 (2016). <https://doi.org/10.1007/s12274->

016-1238-z

- [S3] B. Quan, X. Liang, G. Ji, J. Ma, P. Ouyang et al., Strong electromagnetic wave response derived from the construction of dielectric/magnetic media heterostructure and multiple interfaces. *ACS Appl. Mater. Inter.* **9**, 9964-9974 (2017). <https://doi.org/10.1021/acsami.6b15788>
- [S4] Y. Wang, W. Zhang, X. Wu, C. Luo, Q. Wang et al., Conducting polymer coated metal-organic framework nanoparticles: Facile synthesis and enhanced electromagnetic absorption properties. *Synthetic Met.* **228**, 18-24 (2017). <https://doi.org/10.1016/j.synthmet.2017.04.009>
- [S5] F. Wen, F. Zhang, Z. Liu, Investigation on microwave absorption properties for multiwalled carbon nanotubes/Fe/Co/Ni nanopowders as lightweight absorbers. *J. Phys. Chem. C* **115**, 14025-14030 (2011). <https://doi.org/10.1021/jp202078p>
- [S6] T. Wang, H. Wang, X. Chi, R. Li, J. Wang, Synthesis and microwave absorption properties of Fe-C nanofibers by electrospinning with disperse Fe nanoparticles parceled by carbon. *Carbon* **74**, 312-318 (2014). <https://doi.org/10.1016/j.carbon.2014.03.037>
- [S7] R. Qiang, Y. Du, H. Zhao, Y. Wang, C. Tian et al., Metal organic framework-derived Fe/C nanocubes toward efficient microwave absorption. *J. Mater. Chem. A* **3**, 13426-13434 (2015). <https://doi.org/10.1039/C5TA01457C>
- [S8] H. Liu, Y. Li, M. Yuan, G. Sun, H. Li et al., In situ preparation of cobalt nanoparticles decorated in n-doped carbon nanofibers as excellent electromagnetic wave absorbers. *ACS Appl. Mater. Inter.* **10**, 22591-22601 (2018). <https://doi.org/10.1021/acsami.8b05211>
- [S9] X. Zhang, J. Xu, X. Liu, S. Zhang, H. Yuan et al., Metal organic framework-derived three-dimensional graphene-supported nitrogen-doped carbon nanotube spheres for electromagnetic wave absorption with ultralow filler mass loading. *Carbon* **155**, 233-242 (2019). <https://doi.org/10.1016/j.carbon.2019.08.074>
- [S10] Z. Wu, K. Pei, L. Xing, X. Yu, W. You et al., Enhanced microwave absorption performance from magnetic coupling of magnetic nanoparticles suspended within hierarchically tubular composite. *Adv. Funct. Mater.* **29**, 1901448 (2019). <https://doi.org/10.1002/adfm.201901448>
- [S11] C. Wu, Z. Chen, M. Wang, X. Cao, Y. Zhang et al., Confining tiny MoO<sub>2</sub> clusters into reduced graphene oxide for highly efficient low frequency microwave absorption. *Small* **16**, 2001686 (2020). <https://doi.org/10.1002/sml.202001686>
- [S12] Y. Wang, X. Han, P. Xu, D. Liu, L. Cui et al., Synthesis of pomegranate-like Mo<sub>2</sub>C@C nanospheres for highly efficient microwave absorption. *Chem. Eng. J.* **372**, 312-320 (2019). <https://doi.org/10.1016/j.cej.2019.04.153>
- [S13] S. Dai, Y. Cheng, B. Quan, X. Liang, W. Liu et al., Porous-carbon-based Mo<sub>2</sub>C nanocomposites as excellent microwave absorber: A new exploration. *Nanoscale* **10**, 6945-6953 (2018). <https://doi.org/10.1039/C8NR01244J>