Supplementary Materials

Dual magnetic particles modified carbon nanosheets in CoFe/Co@NC heterostructure for efficient electromagnetic synergy

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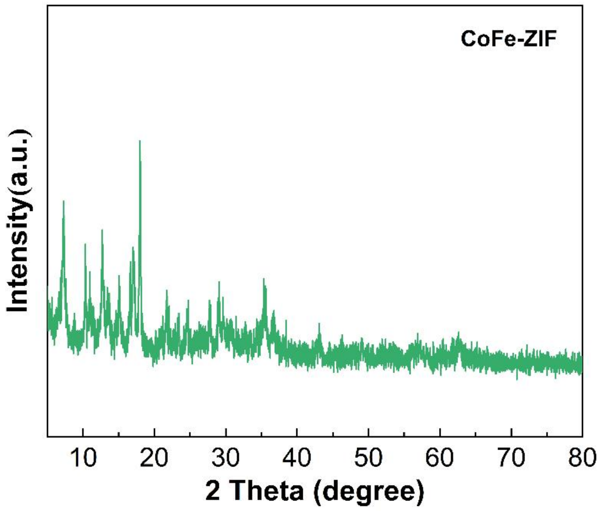
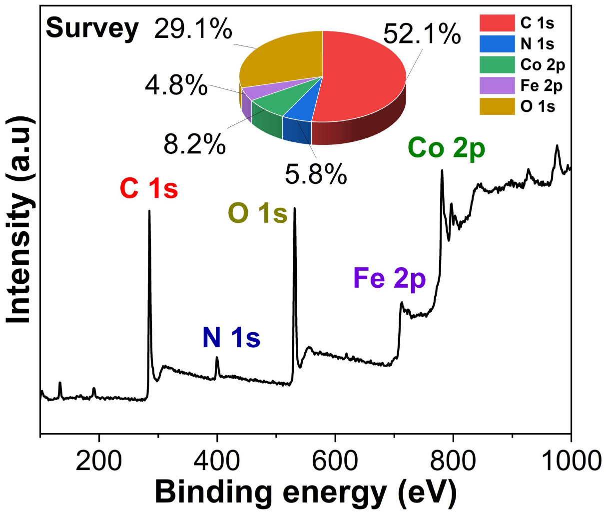
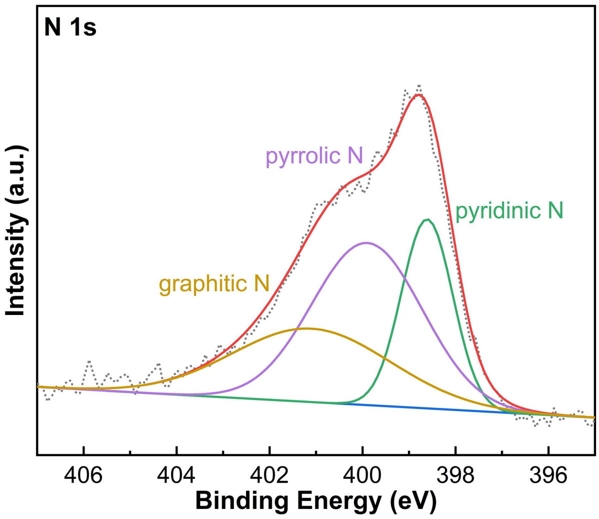


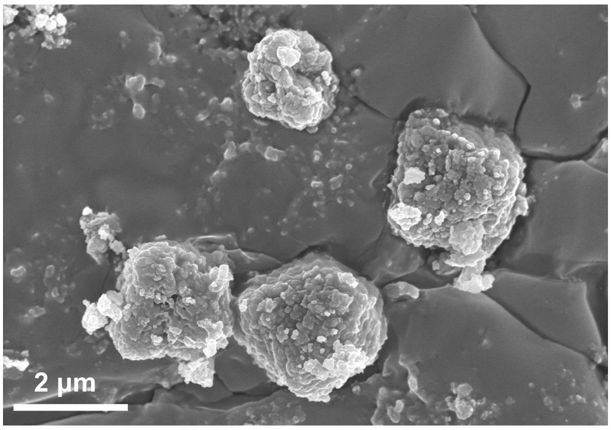
Supplementary Figure 1. XRD patterns of CoFe-MOF.



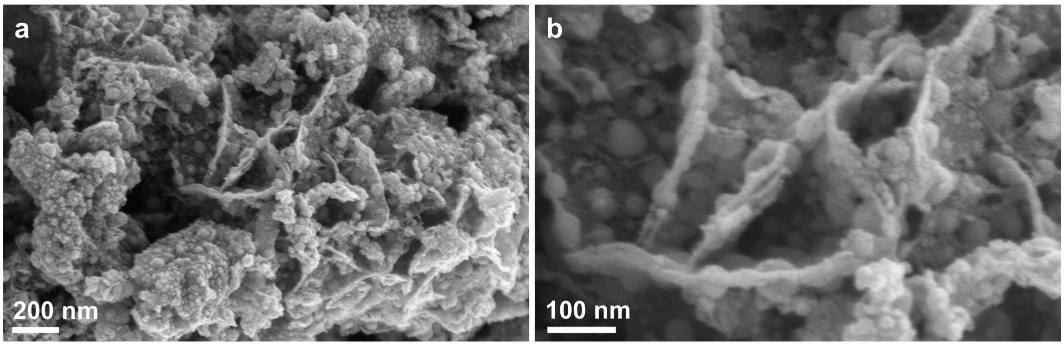
Supplementary Figure 2. XPS survey spectra of CoFe/Co@NC.



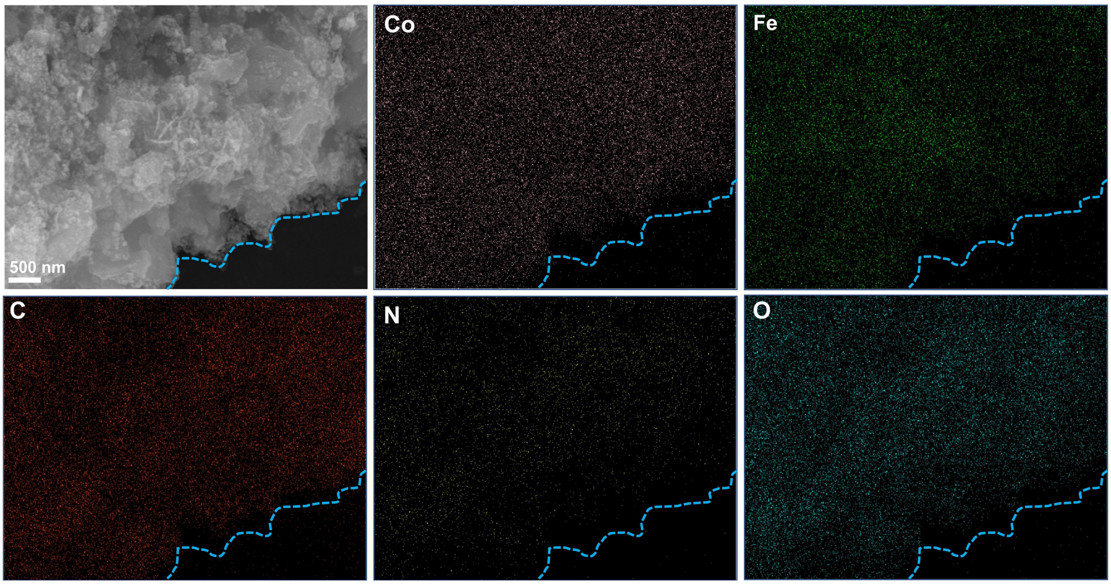
Supplementary Figure 3. High-resolution XPS spectra of N 1s for CoFe/Co@NC.



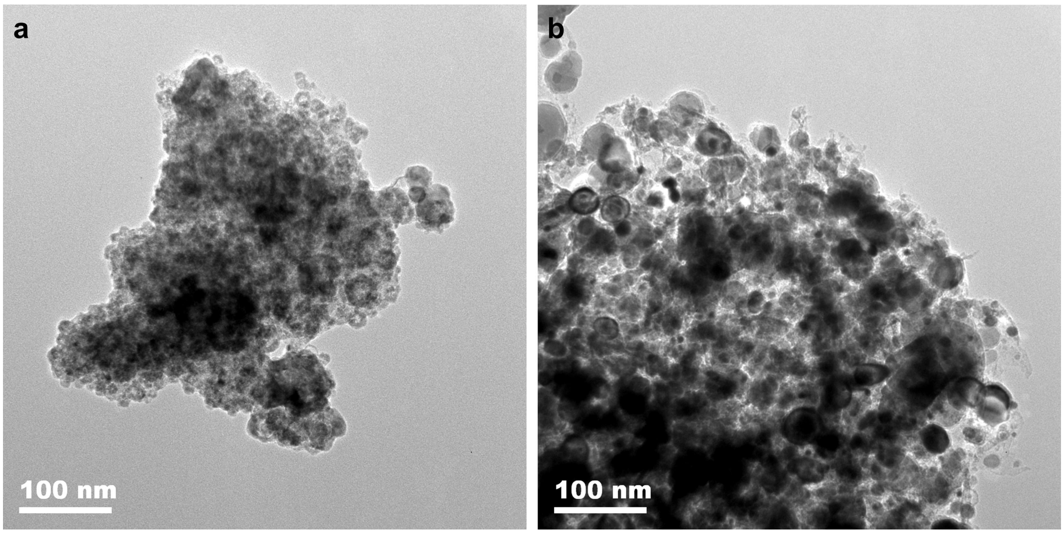
Supplementary Figure 4. SEM images of CoFe-MOF.



Supplementary Figure 5. SEM images of CoFe/Co@NC.



Supplementary Figure 6. Element mapping image of CoFe/Co@NC.



Supplementary Figure 7. TEM images of CoFe/Co@NC.

Supplementary Table 1. EMW absorption performance of CoFe/Co@NC and recently advanced absorbers

Samples	Materials	Shapes	Thickness /mm	R_L /dB	Bandwidth (< -10 dB) /GHz	Ref.
S1	ZIF-800	Cactus-like	1.50	-15.37	4.50	[1]
S2	Co/C	Octahedron	1.70	-15.70	5.40	[2]
S3	$FeNi/Ti_3C_2T_x$	Layered	1.60	-16.96	6.20	[3]
S4	S-Co/C	Sponge-like	1.60	-19.86	3.84	[4]
S5	$Ti_3C_2T_x$@MoS_2@C	Nanosheets	4.80	-20.80	1.00	[5]
S6	FeCo-LDO/RCs	Spheres	2.20	-21.20	7.40	[6]
S7	CF@MoS_2	Fibers	3.80	-21.40	10.85	[7]
S8	SiC_w-BDC	Whiskers	2.55	-24.60	6.80	[8]
S9	Fe_3C/NCFs	Hollow beaded	2.16	-24.71	5.28	[9]
S10	$Ti_3C_2T_x$	Layered	4.00	-27.50	3.00	[10]
S11	CoCN@CNT	Cubic porous	2.50	-29.05	5.73	[11]
S12	$MoO_3/TiO_2/Mo_2TiC_2T_x$	Layered	2.30	-30.76	8.60	[12]
S13	$CoFe_2O_4$-Ti_3C_2	Layered	1.50	-30.90	8.50	[13]
S14	Fe-doped Ti_3AlC_2	Ternary layered	1.50	-33.30	3.90	[14]
S15	Ni@C	Waxberry-like	1.70	-41.50	5.20	[15]
S16	$NiFe_2O_4$/MXene	Nanosheets	2.90	-41.83	1.60	[16]
S17	MXene/FeCo	Film	2.00	-43.70	1.00	[17]
S18	HPPy	Fiber	3.60	-44.50	5.40	[18]
S19	Si_3N_4/SiC	Multilayered	3.20	-45.00	8.40	[19]
S20	$NiCo_2O_4$	Dandelion-like	2.10	-45.08	3.06	[20]
S21	PPy/Fe_3O_4	Planar microhelixes	2.10	-45.48	10.24	[21]
S22	SiC@C	Microsphere	2.10	-46.61	5.61	[22]
S23	Ni@C	Microspheres	3.50	-46.90	2.50	[23]

S24	$CoO/NiCo_2O_4$/MXene	Flower-like	2.90	-47.17	5.44	[24]
S25	N-Ni_xS_y/Co_xS_y@C	Nanoparticles	2.50	-47.20	3.70	[25]
S26	Ni/NiO/C	Hydrangea-like	1.90	-47.72	5.67	[26]
S27	$CuFe_2O_4/MoS_2$	Nanoflower	2.70	-49.43	8.16	[27]
S28	CoNi-MOF-71	Nanosheet arrays	2.60	-49.80	7.60	[28]
S29	$Ti_3C_2T_x$	Foam	1.80	-50.60	4.20	[29]
S30	Fe&TiO_2@C	Sheets	3.00	-51.80	6.50	[30]
S31	$CoFe_2O_4$/carbon	Nanosheets	2.00	-52.29	5.36	[31]
S32	Ni@C	Prisms	2.40	-52.90	4.60	[32]
S33	FeNiZn@C NFs	Octagonal nanoflower	1.90	-53.10	6.00	[33]
S34	Fe_3O_4-rGO	Nanoflowers	6.16	-53.30	5.68	[34]
S35	$NiCo_2O_4$@agaric	Tremella-like	4.09	-54.60	5.70	[35]
S36	Fe_3O_4@SiO_2@MnO_2	Nanochains	3.20	-55.01	4.26	[36]
S37	$ZnCo_2O_4$@$CoSnO_3$	Flower-like	2.60	-56.00	6.96	[37]
S38	C/Co@CNT	Nanotubes	3.80	-56.80	4.80	[38]
S39	MXene nanoribbons-NiCo@NC	hierarchical network	4.82	-57.10	4.82	[39]
S40	NiFe LDH/MXene	Sheet-fiber	2.50	-58.00	7.00	[40]
S41	BSA@Mil-100	Spheres	4.00	-58.00	6.79	[41]
S42	Co@NC	Nanoparticles	2.01	-68.18	4.20	[42]
S43	Cu_9S_5/C	octahedral	1.83	-69.60	5.81	[43]
S44	CeO_2/Co/C	dendritic	1.70	71.50	5.20	[44]
-	CoFe/Co@NC	Nanosheets	1.78	-73.8	5.4	This work

Note: The exact R_L values, thickness, and bandwidth were not presented in some references, thus, those values were dug out according to the R_L-f curves.

References

[1] X.Q. Xu, F.T. Ran, Z.M. Fan, H. Lai, Z.J. Cheng, T. Lv, L. Shao, Y.Y. Liu, Cactus-inspired bimetallic metal-organic framework-derived 1D-2D hierarchical Co/N-decorated carbon architecture toward enhanced electromagnetic wave absorbing performance, ACS Appl. Mater. Interfaces 2019, 11, 13564-13573.

[2] B.Y. Zhu, P. Miao, J. Kong, X.L. Zhang, G.Y. Wang, K.J. Chen, Co/C composite derived from a newly constructed metal-organic framework for effective microwave absorption, Cryst. Growth Des. 2019, 19, 1518-1524.

[3] J. He, X.Y. Liu, Y.H. Deng, Y.H. Peng, L.W. Deng, H. Luo, C.P. Cheng, S.Q. Yan, Improved magnetic loss and impedance matching of the FeNi-decorated $Ti_3C_2T_x$ MXene composite toward the broadband microwave absorption performance, J. Alloys Compd. 2021, 862, 158684.

[4] W. Liu, S.J. Tan, Z.H. Yang, G.B. Ji, Enhanced low-frequency electromagnetic properties of MOF-derived cobalt through interface design, ACS Appl. Mater. Interfaces 2018, 10, 31610-31622.

[5] X. Zhou, H. Han, H. Yan, Y. Wang, C. Zhang, H. Lv, Z. Lou, Multi-interface self-assembling on MXenes skeleton towards wideband electromagnetic dissipation, Mater. Today Phys. 2022, 24, 100685.

[6] H.J. Wu, Z.H. Zhao, G.L. Wu, Facile synthesis of FeCo layered double oxide/raspberry-like carbon microspheres with hierarchical structure for electromagnetic wave absorption, J. Colloid Interf. Sci. 2020, 566, 21-32.

[7] W.D. Zhang, X. Zhang, Q. Zhu, Y. Zheng, L.F. Liotta, H.J. Wu, High-efficiency and wide-bandwidth microwave absorbers based on MoS_2-coated carbon fiber, J. Colloid Interf. Sci. 2021, 586, 457-468.

[8] S. Dong, X.H. Zhang, P.T. Hu, W.Z. Zhang, J.C. Han, P. Hu, Biomass-derived carbon and polypyrrole addition on SiC whiskers for enhancement of electromagnetic wave absorption, Chem. Eng. J. 2019, 359, 882-893.

[9] R.D. Guo, D. Su, F. Chen, Y.Z. Cheng, X. Wang, R.Z. Gong, H. Luo, Hollow beaded Fe_3C/N-doped carbon fibers toward broadband microwave absorption, ACS Appl. Mater. Interfaces 2022, 14, 3084-3094.

[10] G.Z. Cui, X.D. Sun, G.Y. Zhang, Z. Zhang, H. Liu, J. Gu, G.X. Gu, Electromagnetic absorption performance of two-dimensional MXene $Ti_3C_2T_x$ exfoliated by HCl+LiF etchant with diverse etching times, Mater. Lett. 2019, 252, 8-10.

[11] P.S. Yi, X.F. Zhang, L.Q. Jin, P. Chen, J.Q. Tao, J.T. Zhou, Z.G. Yao, Regulating pyrolysis strategy to construct CNTs-linked porous cubic Prussian blue analogue derivatives for lightweight and broadband microwave absorption, Chem. Eng. J. 2022, 430, 132879.

[12] F.Y. Hu, F. Zhang, X.H. Wang, Y.Y. Li, H.L. Wang, R. Zhang, H.X. Li, B.B. Fan, Ultrabroad band microwave absorption from hierarchical $MoO_3/TiO_2/Mo_2TiC_2T_x$ hybrids via annealing treatment, J. Adv. Ceram. 2022, 11, 1466-1478.

[13] J. He, S. Liu, L.W. Deng, D.Y. Shan, C. Cao, H. Luo, S.Q. Yan, Tunable electromagnetic and enhanced microwave absorption properties in $CoFe_2O_4$ decorated Ti_3C_2 MXene composites, Appl. Surf. Sci. 2020, 504, 144210.

[14] J. Li, T.T. Xu, H. Bai, Z.Y. Shen, Y.Y. Huang, W.W. Xing, Z.X. Zhou, Structural modifications and electromagnetic property regulations of Ti_3AlC_2 MAX for enhancing microwave absorption through the strategy of Fe doping, Adv. Mater. Interfaces 2022, 9, 2101510.

[15] D.W. Liu, Y.C. Du, P. Xu, F.Y. Wang, Y.H. Wang, L.R. Cui, H.H. Zhao, X.J. Han, Rationally designed hierarchical N-doped carbon nanotubes wrapping waxberry-like Ni@C microspheres for efficient microwave absorption, J. Mater. Chem. A 2021, 9, 5086-5096.

[16] Y. Guo, D.D. Wang, T.T. Bai, H. Liu, Y.J. Zheng, C.T. Liu, C.Y. Shen, Electrostatic self-assembled $NiFe_2O_4/Ti_3C_2T_x$ MXene nanocomposites for efficient electromagnetic wave absorption at ultralow loading level, Adv. Compos. Hybrid. Ma. 2021, 4, 602-613.

[17] X. Li, C.Y. Wen, L.T. Yang, R.X. Zhang, X.H. Li, Y.S. Li, R.C. Che, MXene/FeCo films with distinct and tunable electromagnetic wave absorption by morphology control and magnetic anisotropy, Carbon 2021, 175, 509-518.

[18] H.C. Ren, T.A. Li, H.G. Wang, Z.H. Guo, T.L. Chen, F.B. Meng, Two birds with one stone: Superhelical chiral polypyrrole towards high-performance electromagnetic wave absorption and corrosion protection, Chem. Eng. J. 2022, 427, 131582.

[19] Z.X. Cai, L. Su, H.J. Wang, M. Niu, L.T. Tao, D. Lu, L. Xu, M.Z. Li, H.F. Gao, Alternating multilayered Si_3N_4/SiC aerogels for broadband and high-temperature electromagnetic wave absorption up to 1000 °C, ACS Appl. Mater. Interfaces 2021, 13, 16704-16712.

[20] L. Chai, Y.Q. Wang, Z.R. Jia, Z.X. Liu, S.Y. Zhou, Q.C. He, H.Y. Du, G.L. Wu, Tunable defects and interfaces of hierarchical dandelion-like $NiCo_2O_4$ via Ostwald

ripening process for high-efficiency electromagnetic wave absorption, Chem. Eng. J. 2022, 429, 132547.

[21] X.F. Yang, B.X. Fan, X. Tang, J.L. Wang, G.X. Tong, D.B. Chen, J.G. Guan, Interface modulation of chiral PPy/Fe_3O_4 planar microhelixes to achieve electric/magnetic-coupling and wide-band microwave absorption, Chem. Eng. J. 2022, 430, 132747.

[22] J. Liu, J.Q. Tao, L.L. Gao, X.X. He, B. Wei, Y.S. Gu, Z.G. Yao, J.T. Zhou, Morphology-size synergy strategy of SiC@C nanoparticles towards lightweight and efficient microwave absorption, Chem. Eng. J. 2022, 433, 134484.

[23] Q.W. Zeng, L. Wang, X. Li, W.B. You, J. Zhang, X.H. Liu, M. Wang, R.C. Che, Double ligand MOF-derived pomegranate-like Ni@C microspheres as high-performance microwave absorber, Appl. Surf. Sci. 2021, 538, 148051.

[24] H.Y. Wang, X.B. Sun, G.S. Wang, A MXene-modulated 3D crosslinking network of hierarchical flower-like MOF derivatives towards ultra-efficient microwave absorption properties, J. Mater. Chem. A 2021, 9, 24571-24581.

[25] L.X. Gai, G.L. Song, Y.Y. Li, W.S. Niu, L.F. Qin, Q.D. An, Z.Y. Xiao, S.G. Zhai, Versatile bimetal sulfides nanoparticles-embedded N-doped hierarchical carbonaceous aerogels (N-Ni_xS_y/Co_xS_y@C) for excellent supercapacitors and microwave absorption, Carbon 2021, 179, 111-124.

[26] L. Lei, Z.J. Yao, J.T. Zhou, W.J. Zheng, B. Wei, J.Q. Zu, K.Y. Yan, Hydrangea-like Ni/NiO/C composites derived from metal-organic frameworks with superior microwave absorption, Carbon 2021, 173, 69-79.

[27] J.K. Liu, Z.R. Jia, W.H. Zhou, X.H. Liu, C.H. Zhang, B.H. Xu, G.L. Wu, Self-assembled MoS_2/magnetic ferrite $CuFe_2O_4$ nanocomposite for high-efficiency microwave absorption, Chem. Eng. J. 2022, 429, 132253.

[28] G. Liu, J.Q. Tu, C. Wu, Y.J. Fu, C.H. Chu, Z.H. Zhu, X.H. Wang, M. Yan, High-yield two-dimensional metal-organic framework derivatives for wideband electromagnetic wave absorption, ACS Appl. Mater. Interfaces 2021, 13, 20459-20466.

[29] K.X. Hu, H.H. Wang, X. Zhang, H. Huang, T. Qiu, Y. Wang, C.F. Zhang, L.M. Pan, J. Yang, Ultralight $Ti_3C_2T_x$ MXene foam with superior microwave absorption performance. Chem. Eng. J. 2021, 408, 127283.

[30] B.W. Deng, Z. Xiang, J. Xiong, Z.C. Liu, L.Z. Yu, W. Lu, Sandwich-like Fe&TiO_2@C nanocomposites derived from MXene/Fe-MOFs hybrids for

electromagnetic absorption, Nano-Micro Lett. 2020, 55, 12.

[31] R.X. Xu, D.W. Xu, Z. Zeng, D. Liu, $CoFe_2O_4$/porous carbon nanosheet composites for broadband microwave absorption, Chem. Eng. J. 2022, 427, 130796.

[32] X. Li, Z.L. Wang, Z. Xiang, X.J. Zhu, Y.Y. Dong, C. Huang, L. Cai, W. Lu, Biconical prisms Ni@C composites derived from metal-organic frameworks with an enhanced electromagnetic wave absorption, Carbon 2021, 184, 115-126.

[33] F. Wu, M.Y. Ling, L.Y. Wan, P. Liu, Y.B. Wang, Q.Y. Zhang, B.L. Zhang, Three-dimensional FeMZn (M = Co or Ni) MOFs: Ions coordinated self-assembling processes and boosting microwave absorption, Chem. Eng. J. 2022, 435, 134905.

[34] X.J. Zeng, L.Y. Zhu, G.M. Jiang, C.Y. Wang, Z.H. Xia, R.H. Yu, Template-free formation of uniform Fe_3O_4 hollow nanoflowers supported on reduced graphene oxide and their excellent microwave absorption performances, Phys. Status. Solidi. A 2018, 215, 1701049.

[35] Y.Y. Dong, X.J. Zhu, F. Pan, L. Cai, H.J. Jiang, J. Cheng, Z. Shi, Z. Xiang, W. Lu, Implanting $NiCo_2O_4$ equalizer with designable nanostructures in agaric aerogel-derived composites for efficient multiband electromagnetic wave absorption, Carbon 2022, 190, 68-79.

[36] M.L. Ma, W.T. Li, Z.Y. Tong, Y.Y. Yang, Y. Ma, Z.H. Cui, R.H. Wang, P. Lyu, W.B. Huang, 1D flower-like Fe_3O_4@SiO_2@MnO_2 nanochains inducing RGO self-assembly into aerogels for high-efficient microwave absorption, Mater. Design 2020, 188, 108462.

[37] C.X. Wang, B.B. Wang, X. Cao, J.W. Zhao, L. Chen, L.G. Shan, H.N. Wang, G.L Wu, 3D flower-like Co-based oxide composites with excellent wideband electromagnetic microwave absorption, Compos. B Eng.2021, 205, 108529.

[38] M.L. Dong, M.Y. Peng, W. Wei, H.J. Xu, C.T Liu, C.Y. Shen, Improved microwave absorption performance of double helical C/Co@CNT nanocomposite with hierarchical structures, J. Mater. Chem. C 2021, 9, 2178-2189.

[39] X.J. Zeng, C. Zhao, T.L. Nie, Z.Y. Shen, R.H. Yu, G.D. Stucky, Construction of 0D/1D/2D MXene nanoribbons-NiCo@NC hierarchical network and their coupling effect on electromagnetic wave absorption, Mater. Today Phys. 2022, 28, 100888.

[40] Z.H. Wang, L.X. Yang, Y. Zhou, C. Xu, M. Yan, C. Wu, NiFe LDH/MXene derivatives interconnected with carbon fabric for flexible electromagnetic wave absorption, ACS Appl. Mater. Interfaces 2021, 13, 16713-16721.

[41] S.U. Rehman, S. Xu, Z.H. Li, T.X. Tao, J. Zhang, H.N. Xia, H.T. Xu, K. Ma, J.F.

Wang, Hierarchical-bioinspired MOFs enhanced electromagnetic wave absorption, Small 2023, 20, 2306466.

[42] W.B. Deng, T.H. Li, H. Li, J. Abdul, L.T. Liu, A. Dang, X. Liu, M.F. Duan, H.J. Wu, MOF derivatives with gradient structure anchored on carbon foam for high-performance electromagnetic wave absorption, Small 2024, 21, 2309806.

[43] S.M. Wu, C.J. Wang, Y.X. Tang, J.Y.M. Jiang, H.T. Jiang, X.D. Xu, B.W. Cui, Y.Y. Jiang, Y.X. Wang, Metal-organic framework-derived hierarchical Cu_9S_5/C nanocomposite fibers for enhanced electromagnetic wave absorption, Adv. Fiber Mater. 2024, 6, 430-443.

[44] L.T. Li, J.R. Liu, F. Pan, J. Qiao, X. Zhang, J.P. Lin, W. Liu, Q.L. Wu, S.F. Tao, F. Wu, Z.H. Zeng, Structural engineering of rare earth metal-organic frameworks derivatives with high anisotropy for high-efficiency electromagnetic wave absorption, Chem. Eng. J. 2024, 481, 148383.