

Supplementary Materials

Tuning Dielectric Loss of SiO₂@CNTs for Electromagnetic Wave Absorption

Fenghui Cao ^{1,2}, Jia Xu ¹, Xinci Zhang ¹, Bei Li ¹, Xiao Zhang ^{1,*}, Qiuyun Ouyang ¹, Xitian Zhang ³ and Yujin Chen ^{1,4,*}

¹ Key Laboratory of In-Fiber Integrated Optics, College of Physics and Optoelectronic Engineering, Harbin Engineering University, Harbin 150001, China; caofenghui@hrbeu.edu.cn (F.C.); xuja110006@hrbeu.edu.cn (J.X.); zhangxinci@hrbeu.edu.cn (X.Z.); 1284034781@hrbeu.edu.cn (B.L.); qyouyang7823@aliyun.com (Q.O.)

² School of Mechatronic Engineering, Daqing Normal University, Daqing 163712, China

³ Key Laboratory for Photonic and Electronic Bandgap Materials, Ministry of Education, School of Physics and Electronic Engineering, Harbin Normal University, Harbin 150025, China; xtzhazhang@hotmail.com

⁴ School of Materials Science and Engineering, Zhengzhou University, Zhengzhou 450001, China

* Correspondence: zhangxiaochn@hrbeu.edu.cn (X.Z.); chenyujin@hrbeu.edu.cn (Y.C.)

EXPERIMENTAL SECTION

Structure Characterizations

The magnetic property of NCNTs samples were measured by a vibrating sample magnetometer (VSM; Lakeshore 7410, Columbus, USA) at room temperature.

Electromagnetic parameter measurement

The electromagnetic wave absorption properties of the absorbing materials were measured by using a vector network analyzer (Anritsu MS4644A Vectorstar) in the 2–18 GHz range at the room temperature. The cylindrical sample (with the inner diameter and outer diameter are 3.04 mm and 7.00 mm respectively, and 3.00 mm thickness) was prepared by mixing absorbing materials with paraffin matrix was controlled to be 25 wt.% for SiO₂@Fe₃C/Fe@NCNT-GT.

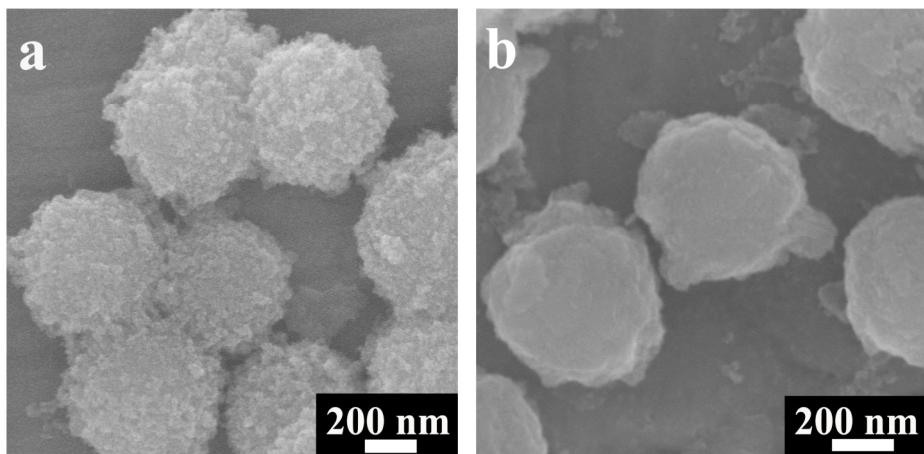


Figure S1. SEM images of SiO₂@Fe(OH)₃ and SiO₂@Fe(OH)₃-GT.

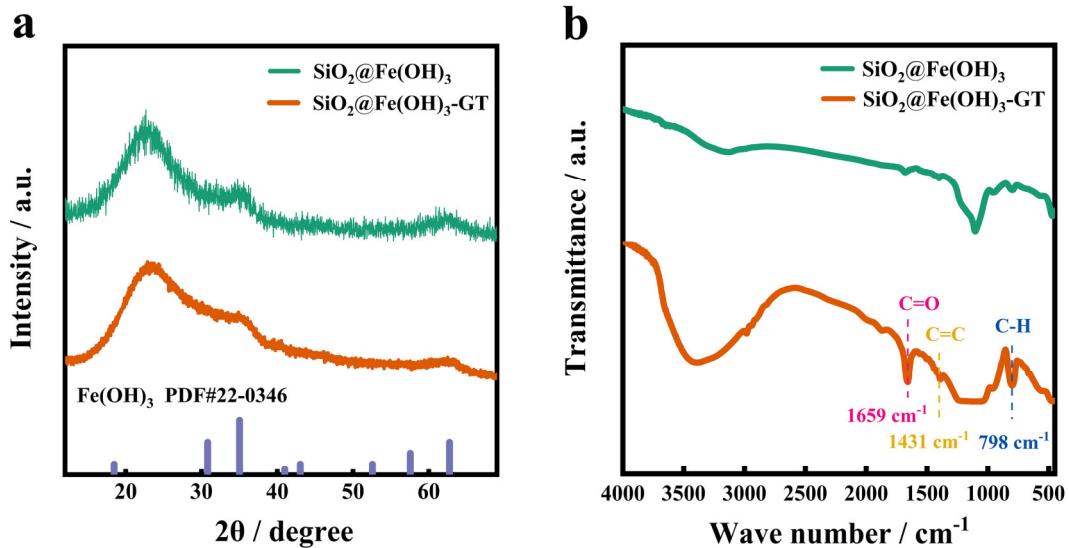


Figure S2. XRD patterns and FTIR spectra of SiO₂@Fe(OH)₃ and SiO₂@Fe(OH)₃-GT.

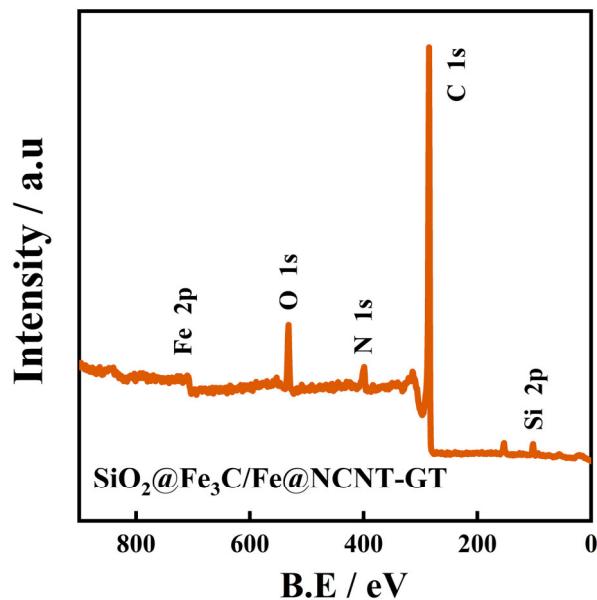


Figure S3. XPS spectra of the SiO₂@Fe₃C/Fe@NCNT-GT.

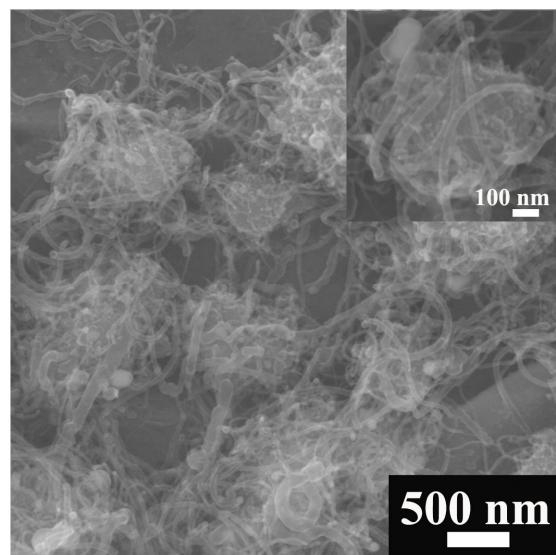


Figure S4. SEM image of $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT-GT}$.

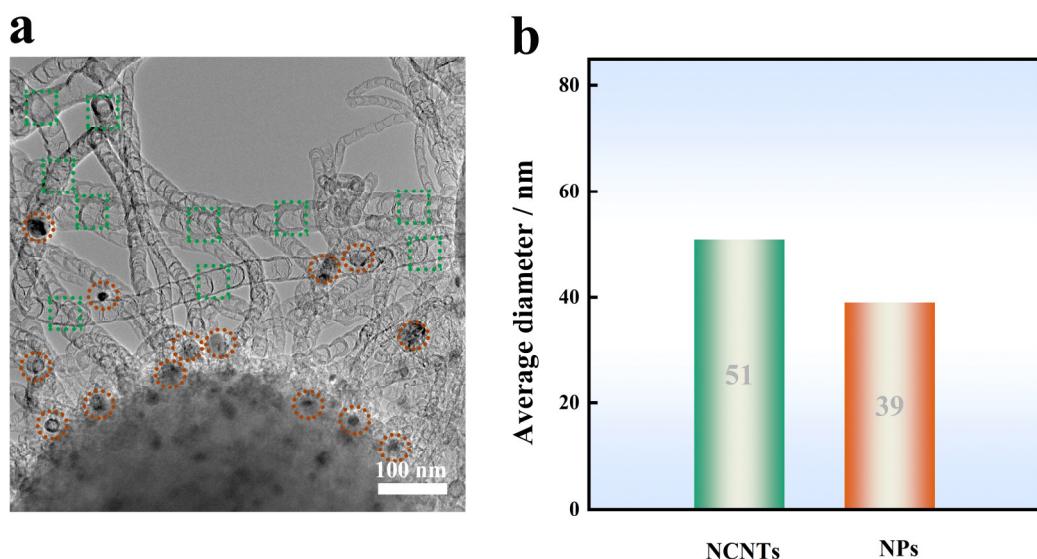


Figure S5. (a) TEM image, (b) Average diameter of NCNTs and NPs of $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT-GT}$.

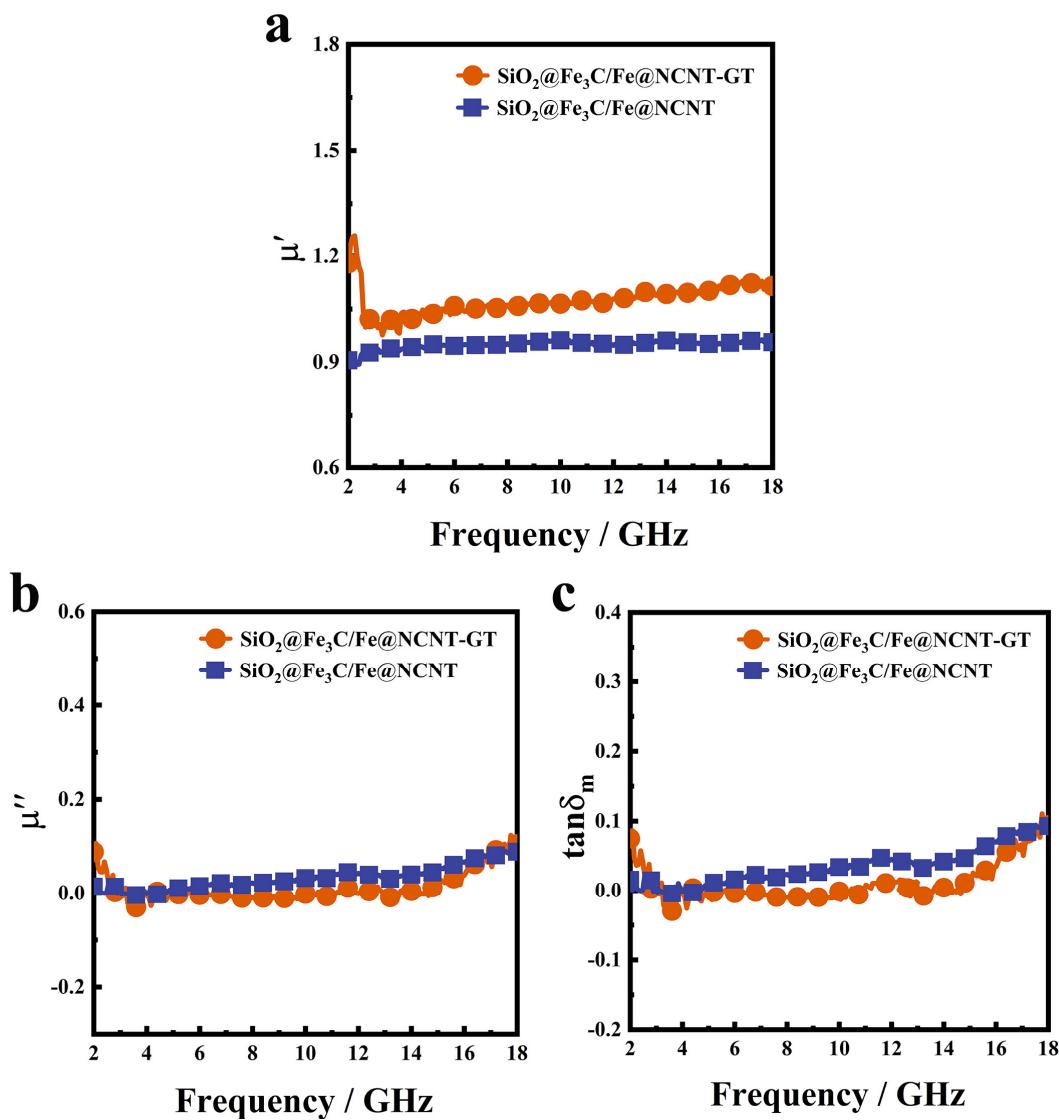


Figure S6. (a) μ' - f curves, (b) μ'' - f curves, and (c) $\tan \delta_m$ - f of $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT-GT}$ and $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT}$.

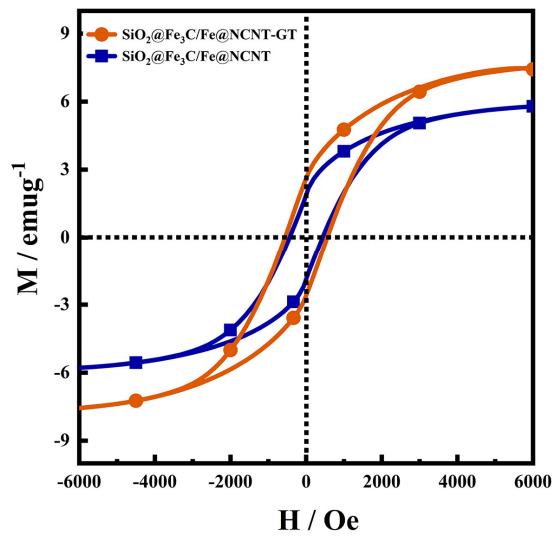


Figure S7. Magnetization hysteresis loops of the $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT-GT}$ and $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT}$.

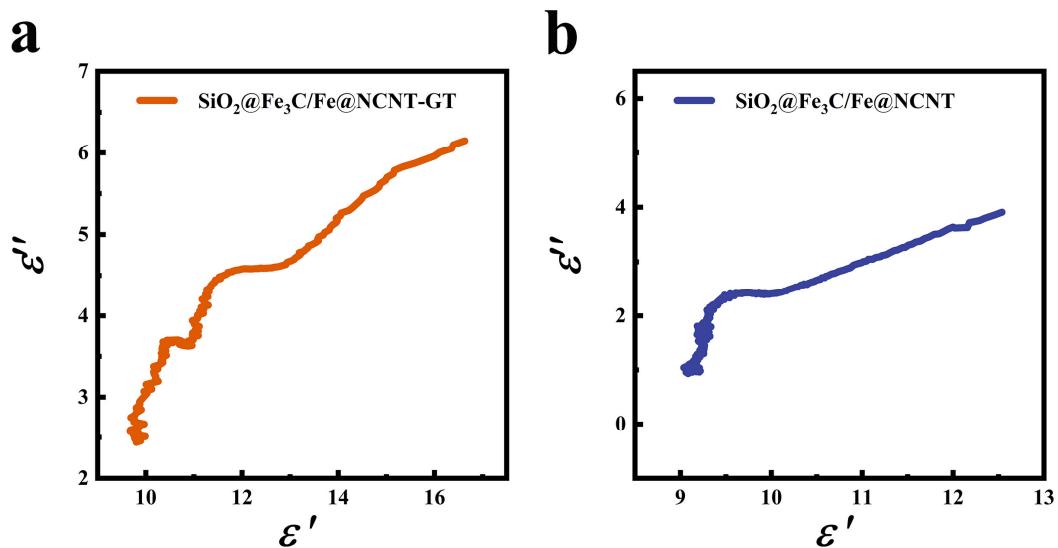


Figure S8. Cole-Cole semicircles of the (a) $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT-GT}$ and (b) $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT}$.

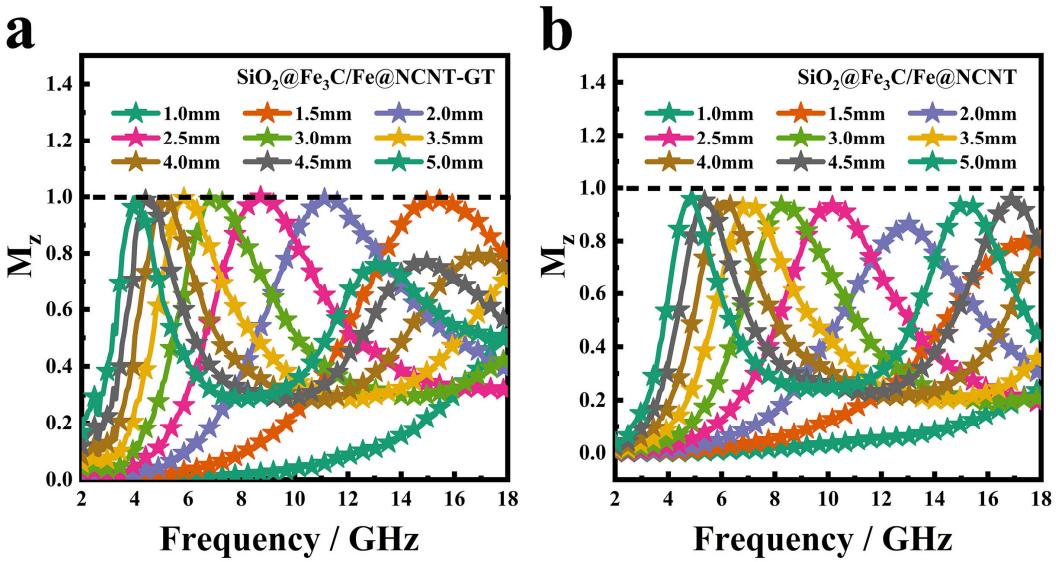


Figure S9. The M_z-f curves of the (a) $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT-GT}$ and (b) $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT}$.

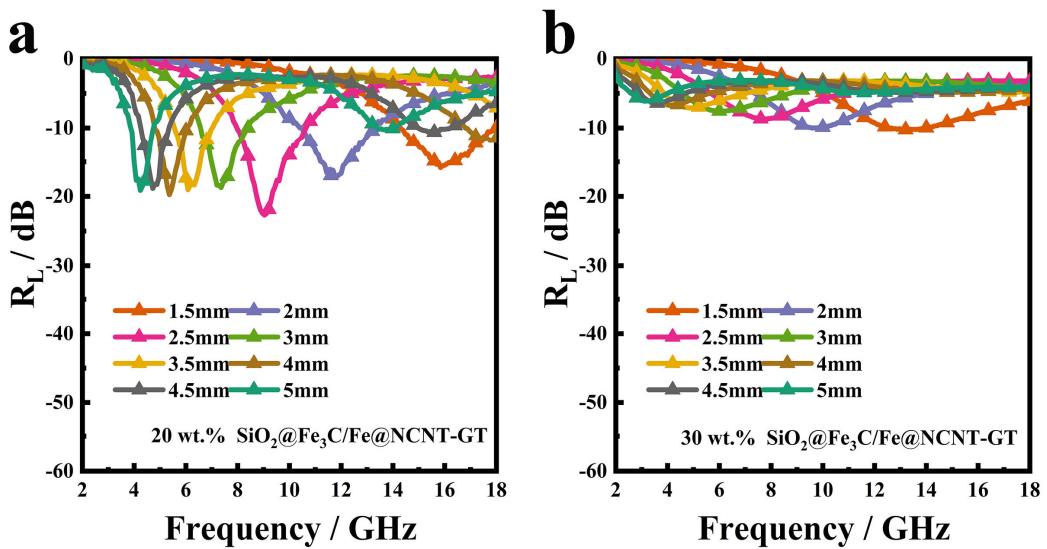


Figure S10. R_L-f curves of (a) the $\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT-GT}$ with a filler ratio of 20 wt.% and (b) 30 wt.%.

Table S1. Electrical conductivity of absorbing materials.

Sample	Electrical conductivity (S m^{-1})
$\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT}$	0.435
$\text{SiO}_2@\text{Fe}_3\text{C}/\text{Fe}@\text{NCNT-GT}$	0.686

Table S2. EMW absorption properties of some representative materials.

Sample	<i>f_r</i> wt.%	<i>d</i> mm	EAB ₁₀ GHz	R _{L, min} dB	SRL dB/m	SEAB GHz/m	Ref.
CoNi/NG hybrids	30	2.0	~5.5	-22.0	3.7	0.9	[2]
CNT/RGO/BaFe ₁₂ O ₁₉	20	2.5	3.8	-19.0	3.8	0.7	[3]
Co NPs/porous C	25	3.0	4.93	-30.3	4.0	0.6	[4]
Ni/C	50	2	4.3	-32.0	3.2	0.4	[5]
Fe ₃ O ₄ /C core–shell nanospindles	60	2.1	~3.0	-38.8	3.1	0.2	[6]
CNT@TiO ₂	30	2.0	3.1	-31.8	5.3	0.5	[7]
MWCNTs/Ni	60	~5.2	-	-37.0	1.2	-	[8]
MWCNTs/Fe	60	~4.3	-	-39.0	1.5	-	[8]
MWCNTs/Co	60	~5.3	-	-37.0	1.2	-	[8]
Fe ₃ O ₄ /polypyrrole/CNT	20	3.0	4.5	-25.9	4.3	0.7	[9]
GO/CNT–Fe ₃ O ₄	30	5.0	~1.5	-37.2	2.5	0.1	[10]
raw CNT's	45	3.0	0	-9.9	0.7	0	[10]
purified CNT's	45	3.0	2.9	-14.9	1.1	0.2	[11]
Fe/CNT's	20	3.5	4.2	-22.7	3.2	0.6	[12]
Fe ₃ O ₄ /BN CNT's	-	2.0	8.1	-42.2	-	-	[13]
Fe ₃ O ₄ /BN CNT's	-	2.5	13.1	-47.9	-	-	[13]
Ni/MWNT	5	4.0	4.4	-23.1	11.5	2.2	[14]
Ni/MWNT	10	4.0	3.4	-17.8	4.4	0.8	[14]
Fe/Fe ₃ C/MWCNT	50	2.0	2.4	-12.5	1.2	0.2	[15]
Fe:Fe ₃ C/MWCNT	50	3.5	2.3	-14.1	0.8	0.1	[15]
Er ₂ O ₃ /MWCNT	20	2.0	2.3	-27.7	6.9	0.6	[16]
Sm ₂ O ₃ /MWCNT	20	2.0	1.6	-21.5	5.4	0.4	[17]
PPy (30 wt%)	30	2.6	6.8	-56.3	7.2	0.9	[18]
PANI	17.5	2.1	5.5	-25.2	6.8	1.4	[19]
SiO ₂ @Fe ₃ C/Fe@NCNT-GT	25	1.5	4.51	-48.43	12.9	1.2	This work

References

- Stöber, W.; Fink, A.; Bohn, E. Controlled growth of monodisperse silica spheres in the micron size range. *J. Colloid Interf. Sci.* **1968**, *26*, 62–69.
- Feng, J.; Pu, F. Z.; Li, Z. X.; Li, X. H.; Hu, X. Y.; Bai, J. T. Interfacial interactions and synergistic effect of CoNi nanocrystals and nitrogen-doped graphene in a composite microwave absorber. *Carbon*. **2016**, *104*, 214–225.
- Zhao, T. K.; Ji, X. L.; Jin, W. B.; Wang, C.; Ma, W. X.; Gao, J. J.; et al. Direct in situ synthesis of a 3D interlinked amorphous carbon nanotube/graphene/BaFe₁₂O₁₉ composite and its electromagnetic wave absorbing properties. *Rsc Advances*. **2017**, *7*, 15903–15910.
- Wang, H.C.; Xiang, L.; Wei, W.; An, J.; He, J.; Gong, C.H.; et al., Efficient and lightweight electromagnetic wave absorber derived from metal organic framework-encapsulated cobalt nanoparticles. *ACS Appl. Mater. Interfaces*. **2017**, *9*, 42102–42110.
- Zhang, X. F.; Dong, X. L.; Huang, H.; Liu, Y. Y.; Wang, W. N.; Zhu, X. G.; et al. Microwave absorption properties of the carbon-coated nickel nanocapsules. *Appl Phys Lett*. **2006**, *89*, 053115.
- Liu, X. F.; Cui, X. R.; Chen, Y. X.; Zhang, X. J.; Yu, R. H.; Wang, G. S.; et al. Modulation of electromagnetic wave absorption by carbon shell thickness in carbon encapsulated magnetite nanospindles-poly(vinylidene fluoride) composites. *Carbon*. **2015**, *95*, 870–878.
- Mo, Z.C.; Yang, R.L.; Lu, D.W.; Yang, L.L.; Hu, Q.M.; Li, H.B. et al., Lightweight, three-dimensional carbon nanotube@TiO₂ sponge with enhanced microwave absorption performance. *Carbon*. **2019**, *144*, 433–439.
- Wen, F. S.; Zhang, F.; Liu, Z. Y. Investigation on microwave absorption properties for multiwalled carbon nanotubes/Fe/Co/Ni nanopowders as lightweight absorbers. *J Phys Chem C*. **2011**, *115*, 14025–14030.
- Zhou, Y.; Miao, J.; Shen, Y.; Xie, A. Novel porous Fe_xC_yN_z/N-doped CNT nanocomposites with excellent bifunctions for catalyzing oxygen reduction reaction and absorbing electromagnetic wave. *Applied Surface Science*. **2018**, *453*, 83–92.
- Wang, L. N.; Jia, X. L.; Li, Y. F.; Yang, F.; Zhang, L. Q.; Liu, L. P.; et al. Synthesis and microwave absorption property of flexible magnetic film based on graphene oxide/carbon nanotubes and Fe₃O₄ nanoparticles. *Journal Of Materials Chemistry A*. **2014**, *2*, 14940–14946.
- Qi, X.; Xu, J.; Hu, Q.; Deng, Y.; Xie, R.; Jiang, Y.; et al. Metal-free carbon nanotubes: synthesis, and enhanced intrinsic microwave absorption properties. *Scientific reports*. **2016**, *6*, 28310.
- Lin, H.; Zhu, H.; Guo, H.; Yu, L. Investigation of the microwave-absorbing properties of Fe-filled carbon nanotubes. *Materials Letters*. **2007**, *61*, 3547–3550.
- Zhang, T.; Zhong, B.; Yang, J. Q.; Huang, X. X.; Wen, G. Boron and nitrogen doped carbon nanotubes/Fe₃O₄ composite architectures with microwave absorption property. *Ceramics International*. **2015**, *41*, 8163–8170.

14. Zou, T.; Li, H.; Zhao, N.; Shi, C. Electromagnetic and microwave absorbing properties of multi-walled carbon nanotubes filled with Ni nanowire. *Journal of Alloys and Compounds*. **2010**, *496*, L22–L4.
15. Xu, P.; Han, X. J.; Liu, X. R.; Zhang, B.; Wang, C.; Wang, X. H. A study of the magnetic and electromagnetic properties of γ -Fe₂O₃–multiwalled carbon nanotubes (MWCNT) and Fe/Fe₃C–MWCNT composites. *Materials Chemistry and Physics*. **2009**, *114*, 556–560.
16. Zhang, L.; Zhu, H.; Song, Y.; Zhang, Y.; Huang, Y. The electromagnetic characteristics and absorbing properties of multi-walled carbon nanotubes filled with Er₂O₃ nanoparticles as microwave absorbers. *Materials Science and Engineering: B*. **2008**, *153*, 78–82.
17. Zhang, L.; Zhu, H. Dielectric, magnetic, and microwave absorbing properties of multi-walled carbon nanotubes filled with Sm₂O₃ nanoparticles. *Materials Letters*. **2009**, *63*, 272–274.
18. Green, M.; Tran, A. T. V.; Chen, X. Maximizing the microwave absorption performance of polypyrrole by data-driven discovery. *Composites Science and Technology*. **2020**, *199*, 108332..
19. Green, M.; Tran, A. T.; Chen, X. Obtaining Strong, Broadband microwave absorption of polyaniline through data-driven materials discovery. *Advanced Materials Interfaces*. **2020**, *7*, 2000658.