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# **Supporting Information**

# High-Efficiency Electromagnetic Interference Shielding Capability of

Magnetic Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene/CNT Composite Film

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#### **S1: Impedance Matching**

According to Eqs. (1) and (2) in manuscript, optimal impedance matching can be obtained if  $Z_{in} = Z_0$  and the *RL* value tends to negative infinity. Moreover, the impedance matching can be changed *via* adjusting the composition or conductivity of absorbers. On the other hand, impedance matching can be expressed as:<sup>1</sup>

$$\left|\Delta\right| = \left|\sinh^2\left(Kfd\right) - M\right| \tag{S1}$$

$$K = \frac{4\pi \sqrt{\varepsilon' \mu' \sin \frac{\delta_{\varepsilon} + \delta_{\mu}}{2}}}{c \cos \delta_{\varepsilon} \cos \delta_{\mu}}$$
(S2)

$$M = \frac{4\mu'\varepsilon'\cos\delta_{\varepsilon}\cos\delta_{\mu}}{\left(\mu'\cos\delta_{\varepsilon} - \varepsilon'\cos\delta_{\mu}\right) + \left[\tan\left(\frac{\delta_{\mu}}{2} - \frac{\delta_{\varepsilon}}{2}\right)\right]^{2}\left(\mu'\cos\delta_{\varepsilon} + \varepsilon'\cos\delta_{\mu}\right)^{2}}$$
(S3)

here,  $\tan \delta_{\varepsilon} (\tan \delta_{\varepsilon} = \varepsilon''/\varepsilon')$  and  $\tan \delta_{\mu} (\tan \delta_{\mu} = \mu''/\mu')$  are the abilities of dielectric and magnetic losses, respectively. In fact, smaller  $|\Delta|$  value (close to zero) always represents a lower possibility of reflection, *i.e.*, a better impedance matching.

### S2: Electrical/Magnetic Loss Simulation Method

COMSOL was used as the electromagnetic wave simulation software. Based on Fig. S9, a two-dimensional structure was established. Meanwhile, plane wave with a frequency of 10 GHz illuminates this structure from above in the Y-axis and a perfectly matched layer directly in the Y direction was outside the structural region. In X direction, the periodic boundary condition is used to reproduce the array mode. Due to the need of consistency between simulation and experiment, the measured frequency-dependent permittivity and permeability are imported to the corresponding structure. The wave transmission equation is shown as follows:

$$\nabla \times \mu_{\rm r}^{-1} (\nabla \times E) - k_0^2 (\varepsilon_{\rm r} - \frac{j\sigma}{\omega \varepsilon_0}) E = 0$$
(S4)

$$E(x, y, z) = \widetilde{E}(x, y)e^{-ik_z z}$$
(S5)

Among them, *E* is electric field intensity (N/C, V/m),  $K_0$  is the free space wave number (rad/m),  $\omega$  is the angular frequency (rad/s),  $\sigma$  stands for the conductivity (S/m) and  $\varepsilon_0$  represents the vacuum dielectric constant (8.8542×10<sup>-2</sup> F/m).

### **S3: EMI Shielding Efficiency**

The EMI shielding efficiency (%) can be calculated using the equation as:<sup>2</sup>

Shielding Efficiency (%) = 100 - 
$$\left(\frac{1}{10^{\frac{SE}{10}}}\right) \times 100$$
 (S6)

S4: Figures S1-S17



**Fig. S1.** High-resolution XPS spectra of  $Ti_3C_2T_x$  MXene and NiCo/MX hybrid at (a) Ni 2p and Co 2p, (b) C 1s, (c) O 1s and (d) Ti 2p regions.



Fig. S2. (a) SEM and (b) TEM images of  $Ti_3C_2T_x$  MXene and photograph of Tyndall effect (insert picture).



Fig. S3. SEM of (a) NiCo/MX 1:2 hybrid, (b) NiCo/MX 1:1 hybrid (c) NiCo/MX2:1hybrid (a) NiCo/MX 3:1 hybrid.



Fig. S4. Frequency dependence of (a) real and (b) imaginary parts of relative complex permittivity ( $\epsilon$ ',  $\epsilon$ "). (c) Real and (d) imaginary parts of complex permeability ( $\mu$ ',  $\mu$ ") of NiCo/MXene hybrids with different NiCo content.



**Fig. S5.** Reflection loss curves of (a) NiCo/MX 1:2 hybrid, (b) NiCo/MX 1:1 hybrid (c) NiCo/MX 2:1 hybrid (a) NiCo/MX 3:1 hybrid with different thickness.



Fig. S6. (a) Attenuation constant and (b) impedance match of NiCo/MXene hybrids with different NiCo content.



**Fig. S7.** Calculated  $|\Delta|$  value maps of (a) NiCo/MX 1:2 hybrid, (b) NiCo/MX 1:1 hybrid (c)

NiCo/MX 2:1 hybrid (a) NiCo/MX 3:1 hybrid.



**Fig. S8.** XRD patterns of NiCo/MX, CNT and NiCo/MX-CNT films with different NiCo/MX:CNT ratios.



**Fig. S9.** Cross section SEM images of NiCo/MX-CNT film with different NiCo/MX:CNT ratio: (a) 9-1, (b) 8-2, (c) 7-3, (d) 5-5, (e) 4-6, (f) 3-7, (g) 2-8, (h) 1-9.



Fig. S10. Film thickness of  $Ti_3C_2T_x$  MXene, NiCo/MX, CNT and NiCO/MX-CNT films with

different component ratios at total mass of 50 mg.



Fig. S11. VSM curves of NiCo/MX-CNT films with different component ratios.



Fig. S12. R-A coefficients of NiCo/MX, CNT and NiCo/MX-CNT films.



**Fig. S13.** EMI SET, SEA, and SER values of NiCO/MX-CNT films with different CNT ratios of (a) 9-1, (b) 8-2, (c) 7-3, (d) 6-4, (e) 5-5, (f) 4-6, (g) 3-7, (h) 2-8 and (i) 1-9.



Fig. S14. EMI (a)  $SE_A$  and (b)  $SE_R$  values of NiCo/MX-CNT at different thickness.



Fig. S15. EMI SE<sub>T</sub>, SE<sub>A</sub>, and SE<sub>R</sub> values of NiCo/MX-CNT and MX ene-CNT films.



Fig. S16. Finite element model of NiCo/MX-CNT film.



Fig. S17. (a) Tensile stress-strain curves and (b) digital photo of NiCo/MX film.

Туре	Samples	Thickness [mm]	Frequency range [GHZ]	EMI SE [dB]	Conductivity [S/cm]	Ref.
Carbon fibres and tubes	CNT composite films	0.13	8-13	65	200	3
	MCMB-MWCNTs	0.6	8-12	56	11	4
	MWCNT/WPU Composites	1	8-12	21.1	/	5
	CNT sponge	1.8	8-12	54.8	/	6
	CNTs/CPE	0.5	8-12	18	0.0035	7
	CNT foam	2	8-12	33	5.16	8
	PUDA/CNT	2	8-12	30.7	0.021	9
	PEBA/MWCNT film	0.4	8-12	41	5.6 *10-4	10
Graphene	Graphene /CNTs	1.6	8-12	38	3.4	11
	Graphene /CNTs	1.6	8-12	36	/	12
	rGO	2	8-12	54.2	/	13
	rGO	1	8-12	38	/	14
	Expanded graphite	0.06	8-12	60.4	15	15
	RGO@AC/PVA	0.8	8-12	14	0.11	16
	Graphene foam	1.5	8-12	61	8.3	17
Metals	Ag nanowire	2.3	8-12	64	10	18
	PVDF/Ni	1.95	8-12	20	6.19 *10 <sup>-6</sup>	19
	Epoxy/Ag-CF	2.5	8-12	35	0.8	20
	PP/Ni	3	1-12.4	20	1	21
	Stainless steel	3.1	8-12	48	/	22
Others	Carbon Foam	2	8-12	51.2	2.4	23
	$MoS_2$	1.5	8-12	24.2	2	24
	rGO-BaTiO <sub>3</sub>	1.5	8-12	41.7	/	25
	PVA/MXene film	0.027	8-12	44.4	7.16	26
	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> @Ni	2	2-18	74.17	/	27

**S5: Table S1** EMI shielding performance of various EMI shielding materials.

VGSs/PI film	0.151	8–12	31.37	2.2	28
CL@GC/CE	2	8–12	35.57	32.66	29
MXene-AgNW film	0.45	8-12	27.8	/	30
MXene@Wood	10	8–12	72	0.37	31
PANI/BF	0.4	8-12	12.29	0.1	32
CCB	1.2	8-12	82	15.6	33
CNW	0.3	8-12	47	29.6	34
Mxene foam	2	8-12	62	19	35
MXene/epoxy aerogel	2	8-12	52.7	/	36
MXene/CNF paper	0.167	8-12	23	/	37
$Ti_3C_2T_x\!/\!rGO~film$	0.06	8-12	59	/	38
NiCo/MX-CNT	0.009	8.2-12.4	46	1851	This work
NiCo/MX-CNT	0.053	8.2-12.4	90.7	1851	This work
NiCo/MX-CNT	0.116	8.2-12.4	105	1851	This work

### **S6: References**

- 1. L. Liang, Q. Li, X. Yan, Y. Feng, Y. Wang, H.-B. Zhang, X. Zhou, C. Liu, C. Shen and X. Xie, *ACS Nano*, 2021, **15**, 6622-6632.
- 2. F. Shahzad, M. Alhabeb, C. B. Hatter, B. Anasori, S. Man Hong, C. M. Koo and Y. Gogotsi, *Science*, 2016, **353**, 1137-1140.
- 3. S. Lu, J. Shao, K. Ma, D. Chen, X. Wang, L. Zhang, Q. Meng and J. Ma, *Carbon*, 2018, **136**, 387-394.
- 4. A. Chaudhary, S. Kumari, R. Kumar, S. Teotia, B. P. Singh, D. A. Singh, S. Dhawan and S. Dhakate, *ACS Appl. Mater. Interfaces*, 2016, **8**, 10600-10608.
- 5. Z. Zeng, H. Jin, M. Chen, W. Li, L. Zhou and Z. Zhang, *Adv. Funct. Mater.*, 2016, **26**, 303-310.
- D. Lu, Z. Mo, B. Liang, L. Yang, Z. He, H. Zhu, Z. Tang and X. Gui, *Carbon*, 2018, 133, 457-463.
- 7. S. Mondal, P. Das, S. Ganguly, R. Ravindren, S. Remanan, P. Bhawal, T. K. Das and N. C. Das, *Compos. Part A Appl. Sci. Manuf.*, 2018, **107**, 447-460.
- 8. Y. Chen, H.-B. Zhang, Y. Yang, M. Wang, A. Cao and Z.-Z. Yu, *Adv. Funct. Mater.*, 2016, **26**, 447-455.
- T. Wang, W.-C. Yu, C.-G. Zhou, W.-J. Sun, Y.-P. Zhang, L.-C. Jia, J.-F. Gao, K. Dai, D.-X. Yan and Z.-M. Li, *Compos. B. Eng.*, 2020, **193**, 108015.
- 10. G. Wang, J. Zhao, C. Ge, G. Zhao and C. B. Park, J. Mater. Chem. C, 2021, 9, 1245-1258.
- 11. L. Kong, X. Yin, M. Han, X. Yuan, Z. Hou, F. Ye, L. Zhang, L. Cheng, Z. Xu and J. Huang, *Carbon*, 2017, **111**, 94-102.
- 12. Q. Song, F. Ye, X. Yin, W. Li, H. Li, Y. Liu, K. Li, K. Xie, X. Li, Q. Fu, L. Cheng, L.

Zhang and B. Wei, Adv. Mater., 2017, 29, 1701583.

- 13. F. Xu, R. Chen, Z. Lin, Y. Qin, Y. Yuan, Y. Li, X. Zhao, M. Yang, X. Sun, S. Wang, Q. Peng, Y. Li and X. He, *ACS Omega*, 2018, **3**, 3599-3607.
- 14. Y. Wu, Z. Wang, X. Liu, X. Shen, Q. Zheng, Q. Xue and J.-K. Kim, ACS Appl. Mater. Interfaces, 2017, 9, 9059-9069.
- 15. Y. Liu, K. Zhang, Y. Mo, L. Zhu, B. Yu, F. Chen and Q. Fu, *Compos Sci Technol*, 2018, **168**, 28-37.
- 16. D. Lai, X. Chen, X. Liu and Y. Wang, ACS Appl. Nano Mater., 2018, 1, 5854-5864.
- 17. Y. Li, J. Liu, S. Wang, L. Zhang and B. Shen, *Compos. B. Eng.*, 2020, **182**, 107615.
- 18. Z. Zeng, M. Chen, Y. Pei, S. I. Seyed Shahabadi, B. Che, P. Wang and X. Lu, ACS Appl. Mater. Interfaces, 2017, 9, 32211-32219.
- 19. H. Gargama, A. K. Thakur and S. K. Chaturvedi, J. Appl. Phys., 2015, 117, 224903.
- W. Zhu, Q. Song, L. Yan, W. Zhang, P.-C. Wu, L. K. Chin, H. Cai, D. P. Tsai, Z. X. Shen, T. W. Deng, S. K. Ting, Y. Gu, G. Q. Lo, D. L. Kwong, Z. C. Yang, R. Huang, A.-Q. Liu and N. Zheludev, *Adv. Mater.*, 2015, 27, 4739-4743.
- 21. S. H. Lee, J. Y. Kim, C. M. Koo and W. N. Kim, *Macromol Res*, 2017, 25, 936-943.
- 22. A. Ameli, M. Nofar, S. Wang and C. B. Park, *ACS Appl. Mater. Interfaces*, 2014, **6**, 11091-11100.
- 23. L. Zhang, M. Liu, S. Roy, E. K. Chu, K. Y. See and X. Hu, ACS Appl. Mater. Interfaces, 2016, 8, 7422-7430.
- 24. Q. Wen, W. Zhou, J. Su, Y. Qing, F. Luo and D. Zhu, J. Alloys Compd., 2016, 666, 359-365.
- 25. Q. Yuchang, W. Qinlong, L. Fa, Z. Wancheng and Z. Dongmei, *J. Mater. Chem. C*, 2016, **4**, 371-375.
- 26. X. Jin, J. Wang, L. Dai, X. Liu, L. Li, Y. Yang, Y. Cao, W. Wang, H. Wu and S. Guo, *Chem. Eng. J.*, 2020, **380**, 122475.
- 27. S. Hu, S. Li, W. Xu, W. Yu and Y. Zhou, *Ceram. Int.*, **2021**, DOI: https://doi.org/10.1016/j.ceramint.2021.07.174.
- 28. Z. Li, Z. Lin, M. Han, Y. Zhang and J. Yu, ACS Appl. Nano Mater., 2021, 4, 7461-7470.
- 29. Z. Zong, F. Ren, Z. Guo, Z. Lu, Y. Jin, Y. Zhao and P. Ren, *Compos. B. Eng.*, 2021, **223**, 109132.
- 30. S. Bai, X. Guo, X. Zhang, X. Zhao and H. Yang, *Compos. Part A Appl. Sci. Manuf.*, 2021, **149**, 106545.
- 31. M. Zhu, X. Yan, H. Xu, Y. Xu and L. Kong, *Carbon*, 2021, 182, 806-814.
- 32. Y. Zhang, Z. Yang, Y. Yu, B. Wen, Y. Liu and M. Qiu, ACS Appl. Polym. Mater., 2019, 1, 737-745.
- 33. X. Jia, B. Shen, Z. Chen, L. Zhang and W. Zheng, ACS Sustain. Chem. Eng., 2019, 7, 18718-18725.
- 34. X. Ma, Y. Li, B. Shen, L. Zhang, Z. Chen, Y. Liu, W. Zhai and W. Zheng, *ACS Appl. Mater. Interfaces*, 2018, **10**, 38255-38263.
- 35. X. Wu, B. Han, H.-B. Zhang, X. Xie, T. Tu, Y. Zhang, Y. Dai, R. Yang and Z.-Z. Yu, *Chem. Eng. J.*, 2020, **381**, 122622.
- 36. S. Zhao, H.-B. Zhang, J.-Q. Luo, Q.-W. Wang, B. Xu, S. Hong and Z.-Z. Yu, *ACS Nano*, 2018, **12**, 11193-11202.
- 37. W.-T. Cao, F.-F. Chen, Y.-J. Zhu, Y.-G. Zhang, Y.-Y. Jiang, M.-G. Ma and F. Chen, *ACS Nano*, 2018, **12**, 4583-4593.
- 38. Y. Zhang, K. Ruan, X. Shi, H. Qiu, Y. Pan, Y. Yan and J. Gu, *Carbon*, 2021, 175, 271-280.