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# **Supporting Information**

## 2 Robust and flexible composite aerogel films with porous multilayered structures

## 3 toward broadband electromagnetic wave absorption

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### **Supporting Information Corresponding Formulas**

#### 17 S1. Electromagnetic parameters measurement

The electromagnetic parameters including complex permittivity and complex permeability of the composite aerogel films were measured within the frequency range of 2-18 GHz. The *RL-f* curve is calculated using the measured electromagnetic parameters based on the following equations:

$$R \quad \text{(d)} \quad B=2 \quad 0 \quad \begin{vmatrix} Z_i & \overline{n} & Z_0 \\ B & g \\ Z_i & -g \\ Z_i & -h & Z_0 \end{vmatrix}$$
(Eq. S1)

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$$\frac{Z_{i}}{Z_{0}} = \sqrt{\frac{\mu_{r}}{\varepsilon_{r}}} a \left[ \frac{2\pi}{jn - h} \sqrt{\frac{f}{\mu_{r}} \varepsilon_{r}} \right]$$
(Eq. S2)

where  $Z_{in}$  represents the input impedance of the absorber,  $Z_0$  is the impedance of free space,<sup>1</sup>  $\varepsilon_r$  is the relative complex permittivity,  $\mu_r$  is the relative complex permeability, *f* is the frequency, *d* is the sample thickness, and *c* is the speed of light.

### 26 S2. Debye relaxation

Dielectric polarization refers to the phenomenon where bound charges migrate along the direction of an applied electric field, resulting in electron displacement or alignment of dipoles. In highfrequency electromagnetic fields, when the motion of electrons or the reorientation of dipoles cannot keep up with the changes under the alternating electric field, a lagging effect of electron or dipole polarization relaxation occurs. The Cole-Cole semicircle equation based on the Debye relaxation theory can effectively contribute to understanding the relaxation processes that occur under the influence of alternating electromagnetic fields based on the following equations:

$$\varepsilon \stackrel{!}{=} \varepsilon_{\infty} + \frac{\varepsilon_{s} - \varepsilon_{\infty}}{1 + \omega^{2} \tau^{2}} \qquad (E q . S 3)$$

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$$\varepsilon \stackrel{\text{\tiny{def}}}{=} \frac{\varepsilon_{s} - \varepsilon_{\infty}}{1 + \omega^{2} \tau^{2}} \omega + \frac{\sigma}{\omega \varepsilon_{0}} \qquad (E q . S 4)$$

where  $\varepsilon_{\infty}$  represents the relative dielectric constant at high-frequency limit,  $\varepsilon_s$  denotes the static 36 dielectric constant,  $\omega$  refers to the angular frequency,  $\tau$  is the polarization relaxation time, and  $\sigma$  is the 37 conductivity. From Eq. S3 and Eq. S4, we can obtain the relationship between  $\varepsilon'$  and  $\varepsilon''$ :<sup>2</sup> 38

$$(\varepsilon' - \frac{\varepsilon_s + \varepsilon_{\infty}}{2})^2 + (-\varepsilon^2 = (\frac{\varepsilon_s - \varepsilon_{\infty}}{2})^2 \qquad (E q . S 5)$$

It should be noted that each Cole-Cole semicircle corresponds to a Debye relaxation 40 process. 41

#### S3. Quarter-wavelength resonance formula 42

$$f_m = \frac{(2 - N)}{4t_m n} (E q) S 6$$

where  $f_m$  is the resonance peak frequency and  $t_m$  is the resonance thickness of the sample. While *n* is 44 the real part of square root of the product of complex permittivity and permeability, and N is a positive 45 integer. 46

#### S4. Impedance matching 47

The attenuation constant ( $\alpha$ ), which mainly determines the attenuation capability of 48 the incident EMWs, can be expressed based on the following equation: 49

 $\alpha = \frac{\sqrt{2}}{c} \pi \sqrt{t} \left( \varepsilon \quad " -\mu \quad " \right) \frac{1}{\mu} \sqrt{(\varepsilon \quad u \in \mu' \quad ^2\mu (v \in \mu' \quad ^2\mu ) \quad (Eq. S7)}$ To achieve high-performance EMW absorption, it is quite important that the incident 51 EMWs should enter the absorber as much as possible. Therefore, a good impedance 52 matching at the air-material interface is extremely necessary and critical. According to 53 Eq. S1 and Eq. S2, the best impedance matching can be obtained when the  $Z(|Z_{in}/Z_0|)$ 54 55 value is close or equal to 1 ( $Z_{in} = Z_0$ ), and the *RL* will reach its optimal value.<sup>3</sup>



58 Fig. S1. (a) SEM image of the multilayered  $Ti_3C_2T_x$ . (b) Digital imaging of the Tyndall effect produced

59 by  $Ti_3C_2T_x$  suspensions. (c) XRD patterns of MAX phase and  $Ti_3C_2T_x$ .

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62 Fig. S2. TEM image of ANFs.

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65 Fig. S3. Digital images of the (a) pure ANF film, (b) pure MXene film, (c) AM, (d) AMN-1, and (e)

66 AMN-3.



68 Fig. S4. Cole-Cole curves of (a) AM, (b) AMN-1, (c) AMN-2, and (d) AMN-3.



**Fig. S5.** The values of  $Z = |Z_{in}/Z_0|$  of (a) AM, (b) AMN-1, (c) AMN-2, and (d) AMN-3.

| Samples   | <i>RL</i> (dB) | EAB (GHz) | Matching thickness | Ref.      |
|---|----------------|-----------|--------------------|-----------|
|   |                |           | (mm)               |           |
| $Fe_3O_4@Ti_3C_2T_x$                                      | -57.2          | 1.4       | 4.2                | 4         |
| MXene/Ni  | -49.9          | 2.1       | 1.75               | 5         |
| MXene/Ni  | -31.9          | 2.3       | 1.0                | 6         |
| MXene-CoC   | -46.48         | 3.0       | 1.02               | 7         |
| $Ti_3C_2T_x/Fe_3O_4$                                      | -50.18         | 3.2       | 1.3                | 8         |
| Ni@MXene  | -52.6          | 3.7       | 3.0                | 9         |
| MXene-CNTs/Ni   | -56.4          | 3.95      | 2.4                | 10        |
| Co/ZnO@CNTs/Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> | -46.0          | 4.0       | 1.9                | 11        |
| MXene/RGO   | -44.3          | 4.84      | 1.5                | 12        |
| MXene-rGO/CoNi  | -54.1          | 4.9       | 2.01               | 13        |
| MXene/Ni  | -50.5          | 5.28      | 3.5                | 14        |
| MXene/RGO   | -31.2          | 5.4       | 2.05               | 15        |
| ANF/MXene/Ni  | -48.6          | 5.8       | 1.5                | This work |

72 Table S1. Comparison of EMW absorption properties of the AMN aerogel film with other related73 MXene-based composites.

## 75 References

- 76 1. C. Wang, V. Murugadoss, J. Kong, Z. He, X. Mai, Q. Shao, Y. Chen, L. Guo, C. Liu, S. Angaiahand
- 77 Z. Guo, *Carbon*, 2018, **140**, 696-733.
- 78 2. N. Yang, Z.-X. Luo, G.-R. Zhu, S.-C. Chen, X.-L. Wang, G. Wu and Y.-Z. Wang, ACS Appl.
- 79 *Mater. Interfaces*, 2019, **11**, 35987-35998.
- 80 3. M. Green and X. Chen, J. Materiomics, 2019, 5, 503-541.
- X. Zhang, H. Wang, R. Hu, C. Huang, W. Zhong, L. Pan, Y. Feng, T. Qiu, C. Zhang and J. Yang,
   *Appl. Surf. Sci.*, 2019, 484, 383-391.
- L. Liang, G. Han, Y. Li, B. Zhao, B. Zhou, Y. Feng, J. Ma, Y. Wang, R. Zhang and C. Liu, ACS
   *Appl. Mater. Interfaces*, 2019, **11**, 25399-25409.
- F. Pan, Y. Rao, D. Batalu, L. Cai, Y. Dong, X. Zhu, Y. Shi, Z. Shi, Y. Liu and W. Lu, *Nanomicro lett.*, 2022, 14, 140.
- F. Pan, L. Yu, Z. Xiang, Z. Liu, B. Deng, E. Cui, Z. Shi, X. Li and W. Lu, *Carbon*, 2021, 172,
  506-515.
- 89 8. Y. Li, Y. Gao, B. Fan, L. Guan, B. Zhao and R. Zhang, J. Phys. Chem. C, 2021, 125, 1991490 19924.
- 91 9. L. Liang, R. Yang, G. Han, Y. Feng, B. Zhao, R. Zhang, Y. Wang and C. Liu, ACS Appl. Mater.
- 92 *Interfaces*, 2020, **12**, 2644-2654.
- 93 10. X. Li, W. You, C. Xu, L. Wang, L. Yang, Y. Li and R. Che, Nanomicro lett., 2021, 13, 157.
- 94 11. C. Sun, Q. Li, Z. Jia, G. Wu and P. Yin, Chem. Eng. J., 2023, 454, 140277.
- 95 12. X. Li, D. Xu, D. Zhou, S. Pang, C. Du, M. A. Darwish, T. Zhou and S.-K. Sun, *Carbon*, 2023,
  208, 374-383.

- 97 13. X. Li, Z. Wu, W. You, L. Yang and R. Che, Nanomicro lett., 2022, 14, 73.
- 98 14. X. Li, W. You, L. Wang, J. Liu, Z. Wu, K. Pei, Y. Li and R. Che, ACS Appl. Mater. Interfaces,
- 99 2019, **11**, 44536-44544.
- 100 15. L. Wang, H. Liu, X. Lv, G. Cui and G. Gu, J. Alloys Compd., 2020, 828, 154251.

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