

Supporting Information

Gravity-induced single-layer gradient structure of Ni@Ti₃C₂T_x/PVA for enhanced microwave absorption

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Table S1. Viscosity of PVA solutions.

Concentration	Viscosity at room temperature (cP)	Viscosity at 60 °C (cP)	Rotation speed used during test (rpm)
5wt.%	10	2.5	100
15wt.%	1025	407	100
15wt.%	1050	400	10
25wt.%	25750	13350	10

Note: Different rotation speed is used due to the different viscosity of sample. The higher rotation speed represents lower measuring range and higher precision. For rotation speed of 100 rpm, the measuring range is 0 ~ 5000 cP, which is not enough for the viscosity of 25wt.% PVA. The difference between viscosity measured using different rotation speed is acceptable, which can be seen from the data of 15wt.% PVA.

Table S2. Effect of distribution of Ni@Ti₃C₂T_x on microwave absorbing performance of the SLGS films.

SLGS Film	Distribution of Ni@Ti ₃ C ₂ T _x	RL _{min} (dB)
NTP-1	Small gradient	-15.75
NTP-2	Medium gradient	-57.7
NTP-3	Large gradient	-3.85

Table S3. Microwave absorbing performance of reported film materials.

Absorber	RL _{min} (dB)	EAB (GHz)	Tested frequency range (GHz)	Thickness (mm)	Reference
Ti ₃ C ₂ T _x /polymer	-26.1	4.2	8.2-12.4	7.3	[1]
V ₄ C ₃ T _x /PU	-39	4.2	8.2-12.4	3.4	[2]
SiC/MXene/polymer	-45.5	3.5	2-18	1.58	[3]
CNF/MXene	-42.2	7.12	2-18	4.7	[4]
MXene/Co/PVDF	-38.2	4.8	18-26.5	3	[5]
CNTs/Fe ₃ O ₄ /PU/PET	-17.19	2	12-18	1	[6]
RGO/epoxy	-32	10	8-18	3.2	[7]
PVB/Co ₂ Z/MXene	-46.3	1.6	2-18	2.8	[8]
PANI-PTSA/PU	-37	1.8	8.2-12.4	1.2	[9]
GO/CNT-Fe ₃ O ₄	-37.25	1	2-18	5	[10]
Ni@Ti ₃ C ₂ T _x /PVA	-57.7	3.2	8.2-12.4	2.22	This work

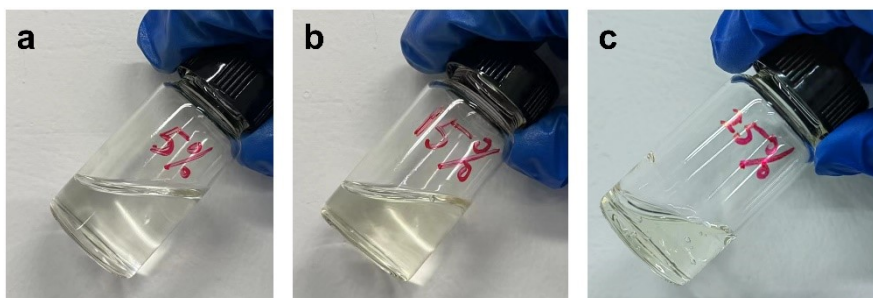


Figure S1. Photos of PVA solutions with concentration of **a** 5wt.%, **b** 15wt.%, **c** 25wt.%.

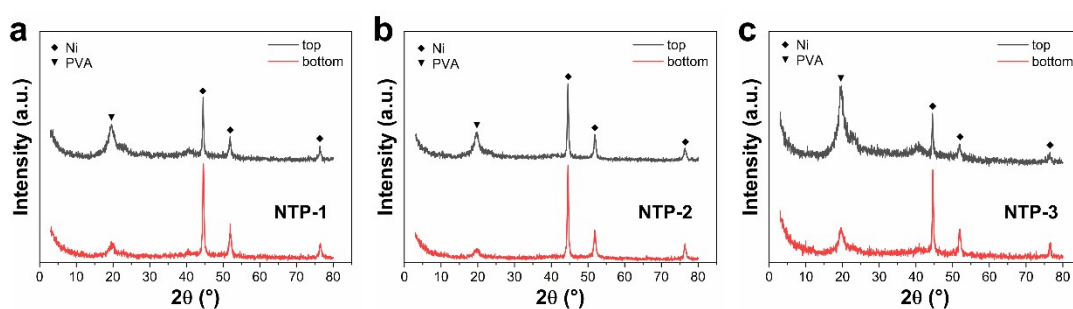


Figure S2. XRD patterns of top and bottom sides of **a** NTP-1, **b** NTP-2, **c** NTP-3.

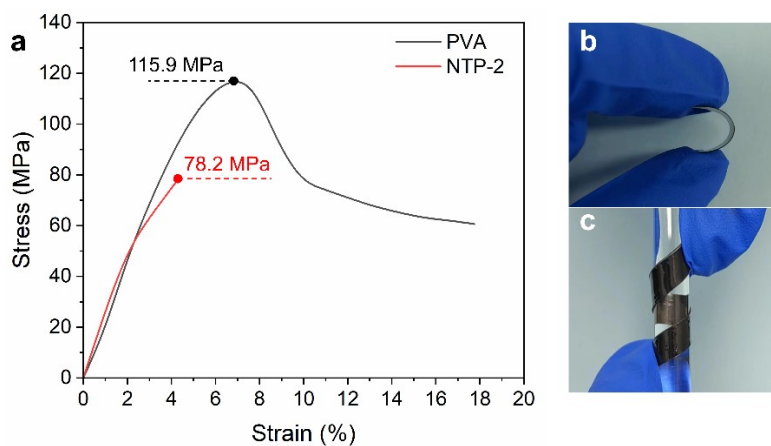


Figure S3. **a** Strain-Stress curves of pure PVA film and NTP-2. **b**, **c** Photos of NTP-2 showing its flexibility.

Experimental

Synthesis of Ti_3AlC_2 : TiC (2 ~ 4 μm , Aladdin Reagent), Ti (300 mesh, Aladdin Reagent) and Al (300 mesh, Zhongnuo Advanced Material) with a mole ratio of 1.8:1:1 were mixed by a 3D powder mixer for 24 h. Afterwards, the mixture was put in an alumina crucible and heated to 1450 $^{\circ}\text{C}$ for 2 h in an argon atmosphere. The Ti_3AlC_2 powder was obtained from crushing the derived bulk ceramic and sieving through a 300-mesh screen. SEM image and XRD pattern of the obtained Ti_3AlC_2 is shown in **Figure S3**.

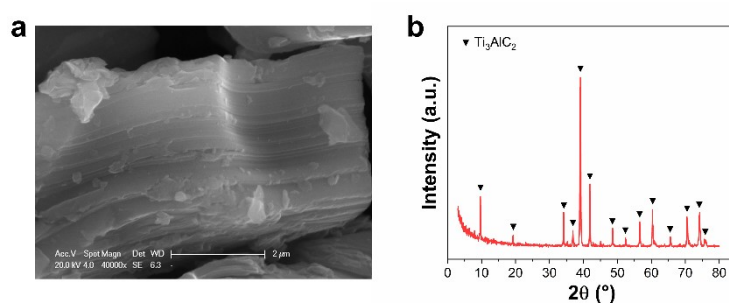


Figure S4. **a** SEM image and **b** XRD pattern of Ti_3AlC_2 .

Synthesis of few-layer $Ti_3C_2T_x$ colloid: First, 5 g of LiF (Aladdin Reagent) was added into 100 mL HCl (Sinopharm Chemical Reagent) and magnetic stirred at 500 rpm for 10 min to fully dissolve LiF. Then 5 g of Ti_3AlC_2 was slowly added to the solution and kept stirring at 45 $^{\circ}\text{C}$ for 24 h. Afterwards, the product was repeatedly washed with deionized water by centrifugation at 4500 rpm for 5 min until the pH reached 6 ~ 7. The sediment was collected and added by 100 mL deionized water, followed by sonication in ice water bath and argon atmosphere for 60 min. The derived black liquid was centrifuged at 3500 rpm for 60 min and the supernatant is the few-layer $Ti_3C_2T_x$ colloid. The concentration of the colloid was measured by freeze-drying method, and the measured concentration is about 15 mg/mL.

Reference

- [1] B. Ji, S. Fan, S. Kou, X. Xia, J. Deng, L. Cheng, L. Zhang, *Carbon* **2021**, 181, 130.
- [2] M. Han, C. E. Shuck, A. Singh, Y. Yang, A. C. Foucher, A. Goad, B. McBride, S. J. May, V. B. Shenoy, E. A. Stach, Y. Gogotsi, *Cell Rep. Phys. Sci.* **2022**, 3, 101073.
- [3] Y. Zhou, M. Wu, J. Jiang, P. Yang, T. Rao, J. J. Liou, W. Liao, *Appl. Surf. Sci.* **2022**, 574, 151463.
- [4] H. Peng, M. He, Y. Zhou, Z. Song, Y. Wang, S. Feng, X. Chen, X. Zhang, H. Chen, *Chem. Eng. J.* **2022**, 433, 133269.
- [5] R. Li, Q. Gao, H. Xing, Y. Su, H. Zhang, D. Zeng, B. Fan, B. Zhao, *Carbon* **2021**, 183, 301.
- [6] W. Gu, R. Zhan, R. Li, J. Liu, J. Zhang, *Coatings* **2021**, 11, 982.
- [7] F. Ye, C. Song, Q. Zhou, X. Yin, M. Han, X. Li, L. Zhang, L. Cheng, *Materials* **2018**, 11, 1771.
- [8] H. Yang, J. Dai, X. Liu, Y. Lin, J. Wang, L. Wang, F. Wang, *Mater. Chem. Phys.* **2017**, 200, 179.
- [9] S. Zeghina, J.-L. Wojkiewicz, S. Lamouri, B. Belaabed, N. Redon, *J. Appl. Polym. Sci.* **2014**, 131, 40961.
- [10] L. Wang, X. Jia, Y. Li, F. Yang, L. Zhang, L. Liu, X. Ren, H. Yang, *J. Mater. Chem. A* **2014**, 2, 14940.