

Supporting Information for

## Ultrathin, Lightweight, and Flexible CNT Buckypaper Enhanced Using MXenes for Electromagnetic Interference Shielding

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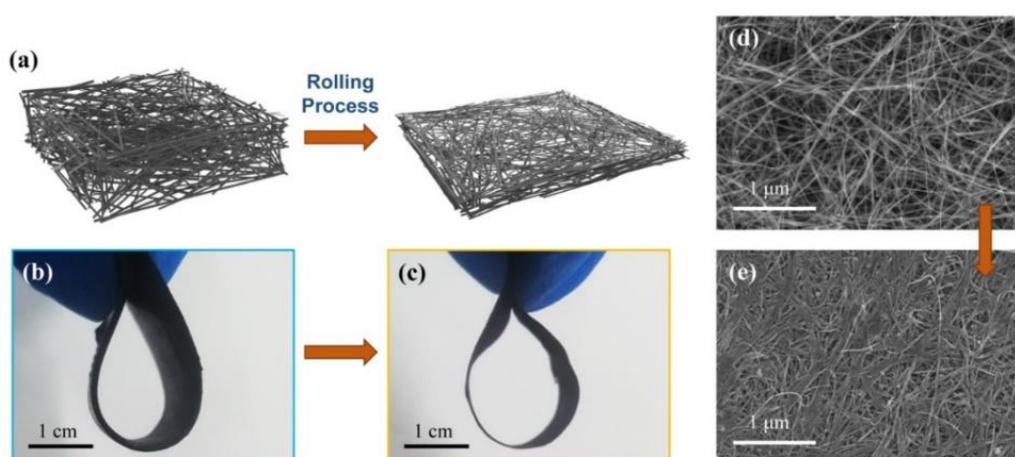
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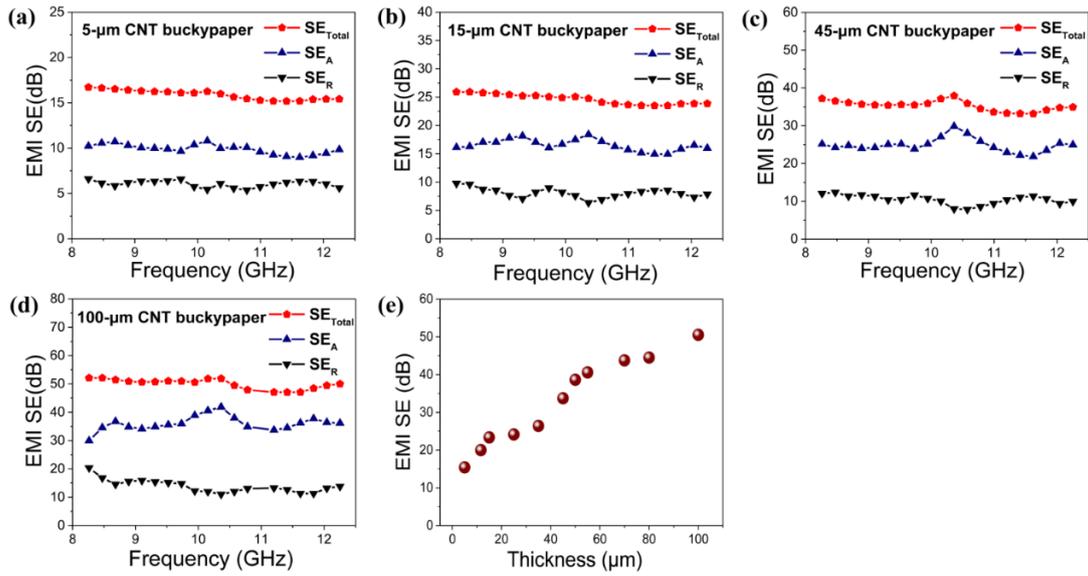
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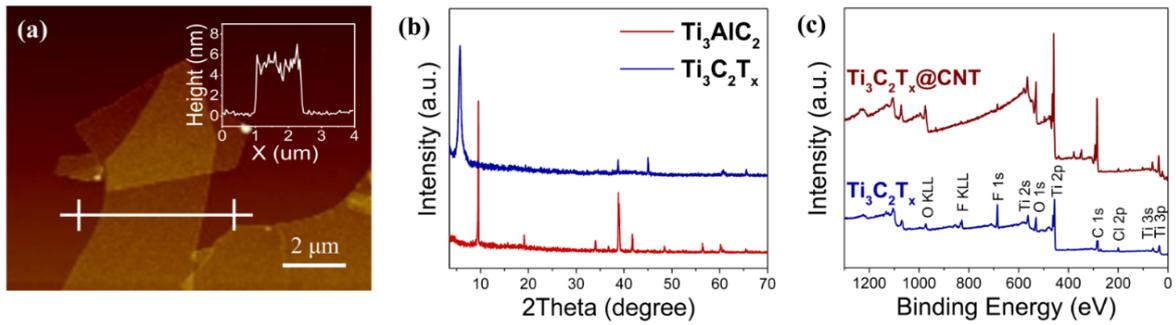
### Supplementary Tables and Figures



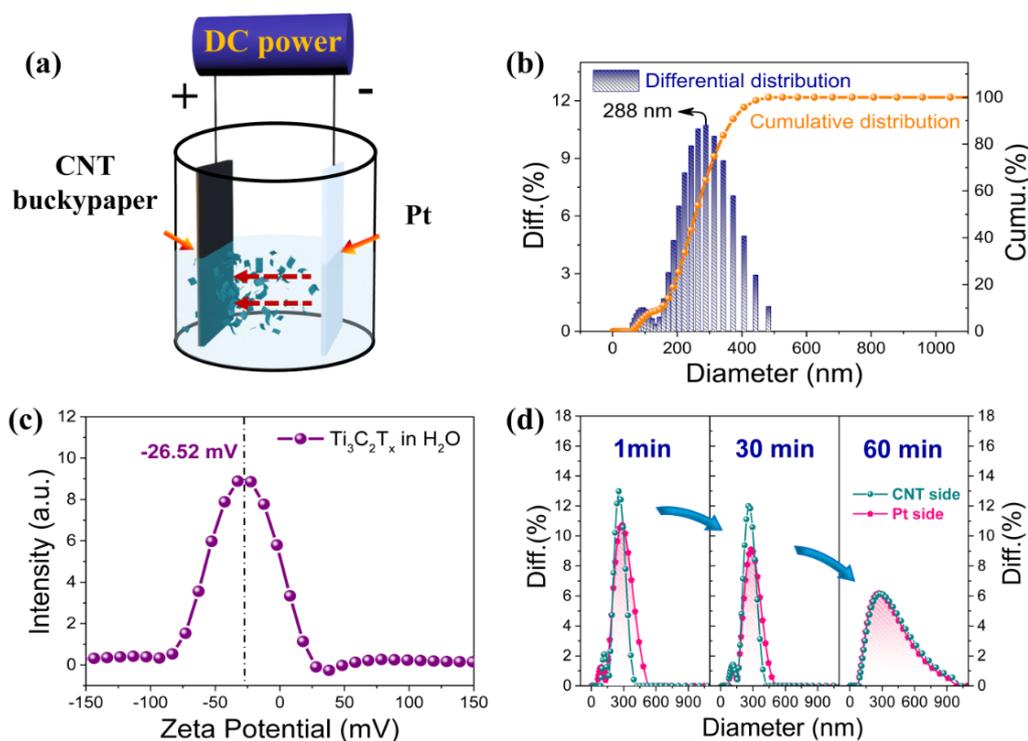
**Fig. S1** a Schematic illustration of rolling process for densifying the CNT buckypaper. Digital image of CNT buckypaper **b** before and **c** after densifying. Top-view SEM image of CNT buckypaper **d** before and **e** after densifying



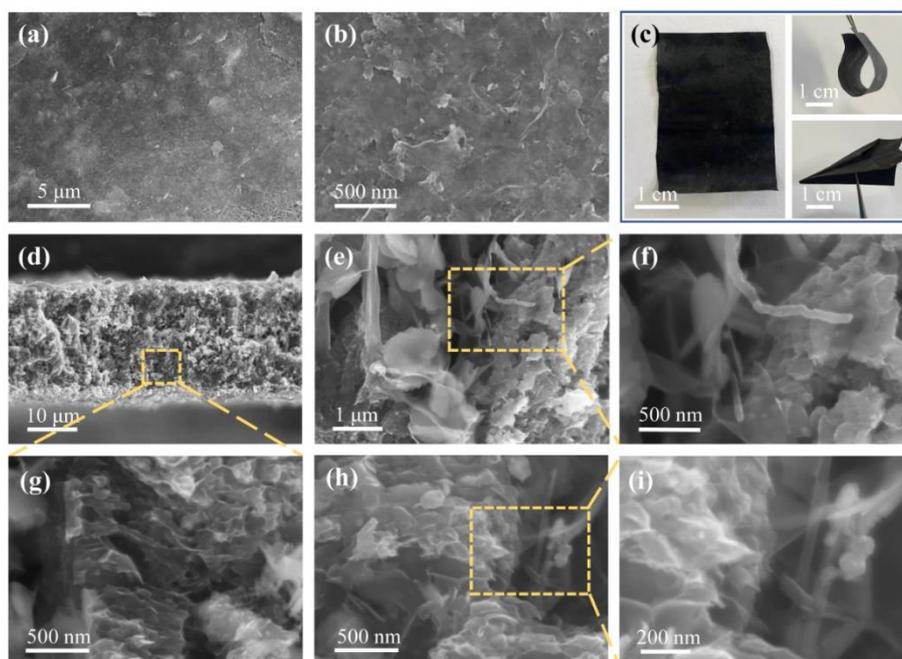
**Fig. S2** EMI SE of CNT buckypapers with thicknesses of **a** 5  $\mu\text{m}$ , **b** 15  $\mu\text{m}$ , **c** 45  $\mu\text{m}$ , and **d** 100  $\mu\text{m}$  in X-band region. **e** Comprehensive average  $SE_{\text{Total}}$  data *versus* thickness of CNT buckypapers



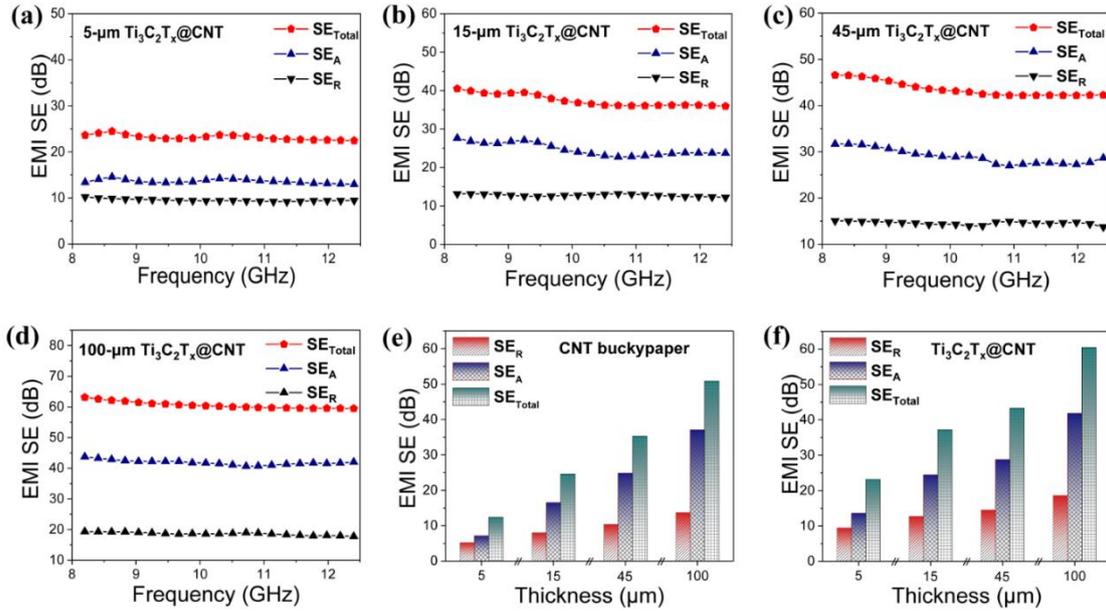
**Fig. S3** **a** AFM image of  $\text{Ti}_3\text{C}_2\text{T}_x$  nanosheets. **b** XRD patterns of  $\text{Ti}_3\text{AlC}_2$  powder and  $\text{Ti}_3\text{C}_2\text{T}_x$  nanosheets. **c** XPS survey spectra for  $\text{Ti}_3\text{C}_2\text{T}_x$  and  $\text{Ti}_3\text{C}_2\text{T}_x$ @CNT hybrid buckypaper



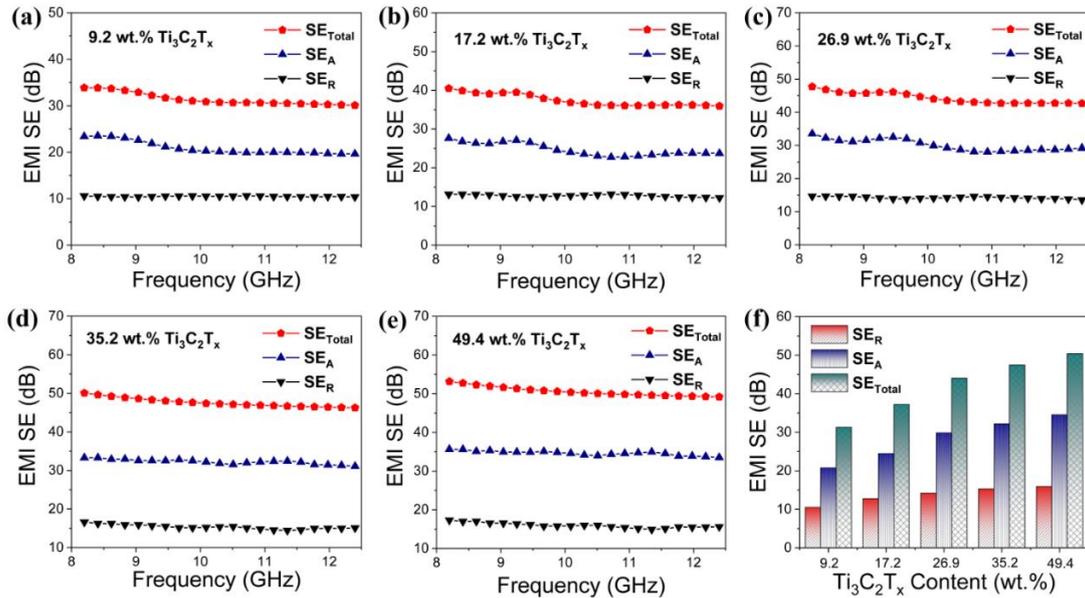
**Fig. S4** **a** Schematic illustration of electrophoretic deposition process. **b** Size distribution and **c** zeta potential of  $\text{Ti}_3\text{C}_2\text{T}_x$  nanoflakes in the  $\text{Ti}_3\text{C}_2\text{T}_x$  aqueous dispersion. **d** Variation of size distributions in  $\text{Ti}_3\text{C}_2\text{T}_x$  nanoflakes around two electrodes during the electrophoretic deposition process



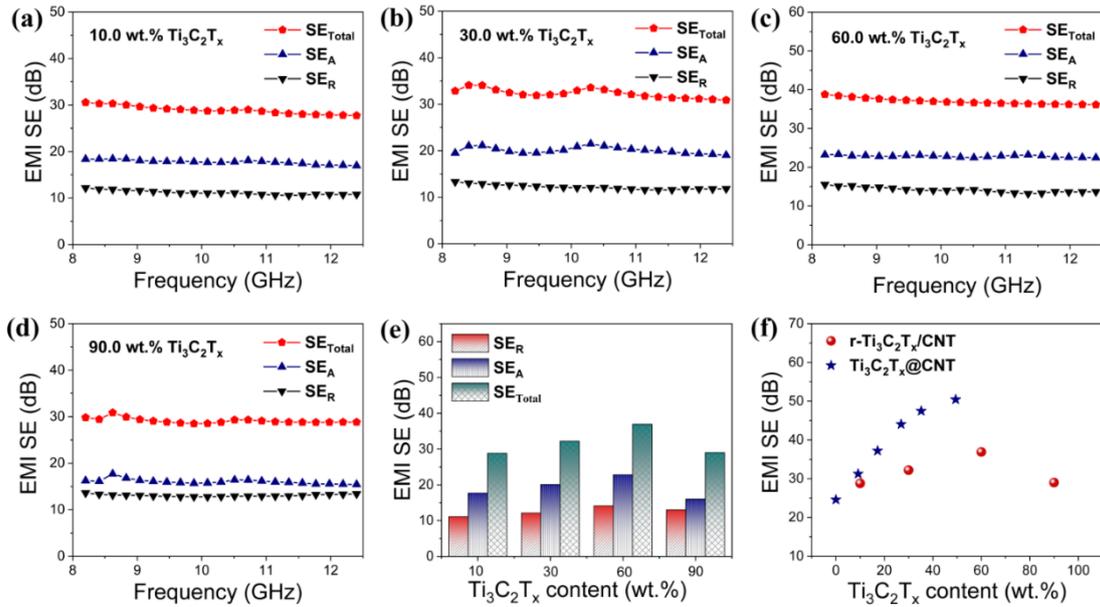
**Fig. S5** **a, b** Top-view SEM images of  $\text{Ti}_3\text{C}_2\text{T}_x$ @CNT hybrid buckypaper. **c** Digital images of  $\text{Ti}_3\text{C}_2\text{T}_x$ @CNT hybrid buckypapers. **d-i** Cross-sectional SEM images of  $\text{Ti}_3\text{C}_2\text{T}_x$ @CNT hybrid buckypaper



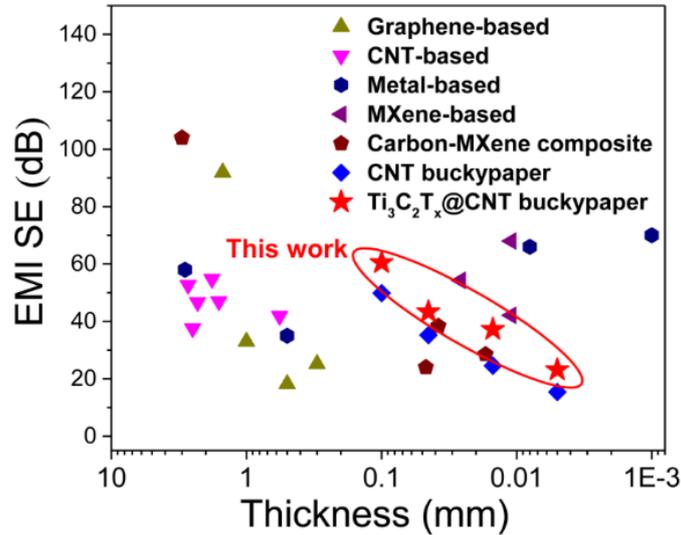
**Fig. S6** EMI SE of  $Ti_3C_2T_x@CNT$  hybrid buckypapers with thicknesses of **a** 5 μm, **b** 15 μm, **c** 45 μm, and **d** 100 μm in X-band region. Comparison of average  $SE_R$ ,  $SE_A$ , and  $SE_{Total}$  versus thickness in **e** CNT buckypapers and **f**  $Ti_3C_2T_x@CNT$  hybrid buckypapers



**Fig. S7** EMI SE of  $Ti_3C_2T_x@CNT$  hybrid buckypapers with  $Ti_3C_2T_x$  content of **a** 9.2 wt%, **b** 17.2 wt%, **c** 26.9 wt%, **d** 35.2 wt%, and **e** 49.4 wt%. **f** Comparison of average  $SE_R$ ,  $SE_A$ , and  $SE_{Total}$  versus  $Ti_3C_2T_x$  content in  $Ti_3C_2T_x@CNT$  hybrid buckypapers



**Fig. S8** EMI SE of randomly mixed  $\text{Ti}_3\text{C}_2\text{T}_x/\text{CNT}$  films with  $\text{Ti}_3\text{C}_2\text{T}_x$  content of **a** 10.0 wt%, **b** 30.0 wt%, **c** 60.0 wt%, and **d** 90.0 wt% in X-band region. **e** Comparison of average  $\text{SE}_R$ ,  $\text{SE}_A$ , and  $\text{SE}_{\text{Total}}$  versus  $\text{Ti}_3\text{C}_2\text{T}_x$  content in r- $\text{Ti}_3\text{C}_2\text{T}_x/\text{CNT}$  films. **f** Comparison of average  $\text{SE}_{\text{Total}}$  versus  $\text{Ti}_3\text{C}_2\text{T}_x$  content between  $\text{Ti}_3\text{C}_2\text{T}_x@\text{CNT}$  hybrid buckypapers and r- $\text{Ti}_3\text{C}_2\text{T}_x/\text{CNT}$  films



**Fig. S9** Comparison of SE versus thickness in  $\text{Ti}_3\text{C}_2\text{T}_x@\text{CNT}$  hybrid buckypapers and other shielding materials. Detailed data thereof is listed in Table S1

**Table S1** EMI shielding performance, thickness averaged specific EMI SE of various shielding materials

Type	Materials	Thickness (mm)	SE (dB)	SSE/t (dB cm <sup>2</sup> g <sup>-1</sup> )	Refs.
Graphene-based	Graphene foam/PDMS	1	33	3330	[S1]
	Graphene foam/PEDOT/PSS	1.5	91.9	20800	[S2]
	RGO/MWCNTs/PI	0.5	18.2	823	[S3]
	Graphene foam	0.3	25.2	14000	[S4]
CNT-based	CNT/WPU	2.3	46.7	10400	[S5]
	MWCNT/PDMS/hollow glass microspheres	2.7	52.7	260.4	[S6]
	CNT/Aramid Nanofiber	0.568	41.9	18304.6	[S7]
	CNT/Graphene Edge hybrid foam	1.6	47	33005.6	[S8]
	CNT/Chitosan	2.5	37.6	8556	[S9]
	CNT sponge	1.8	54.8	30444	[S10]
Metal-based	Ni fiber/PES	2.85	58	109	[S11]
	Ag NW	0.5	35	2416	[S12]
	Al Foil	0.008	66	30555	[S13]
	Cu Foil	0.001	70	7812	[S13]
MXene-based	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	0.011	68	25863	[S13]
	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /CA	0.026	54.3	17586	[S14]
	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /PEDOT/PSS	0.011	42.1	19497.8	[S15]
Carbon-MXene composite	Aramid nanofiber/Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	0.017	28.5	13377.1	[S16]
	Cellulose Nanofiber/Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	0.047	24	2647	[S17]
	CNTs/ Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> / Cellulose Nanofibrils	0.038	38.4	8020	[S18]
	CNTs/ Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> aerogel	3	104	8253.2	[S19]
This work	CNT buckypapers	0.1	49.8	11127.3	This work
		0.045	35.2	16721.7	
		0.015	24.6	33182.8	
		0.005	15.4	47512.8	
This work	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> @CNT hybrid buckypaper	0.1	60.5	13074.2	This work
		0.045	43.3	19138.4	
		0.015	37.2	41635.7	
		0.005	23.1	56945.8	

**Table S2** Thickness, density, SE, SSE, SSE/t of various ultrathin EMI shielding films

Materials	Thickness ( $\mu\text{m}$ )	Density ( $\text{g cm}^{-3}$ )	SE (dB)	SSE (dB $\text{cm}^3 \text{g}^{-1}$ )	SSE/t (dB $\text{cm}^2 \text{g}^{-1}$ )	Refs.
Graphene film	27	1.76	68	38.6	14309.8	[S20]
CNT film	20	1.34	67.4	50.3	25149.3	[S21]
Cu foil	10	8.97	70	7.8	7812	[S13]
MXene film	11	2.39	68	28.4	25863	[S13]
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> @CNT buckypaper	15	0.98	50.4	51.0	34003.7	This work

## Supplementary References

- [S1] Z. Chen, C. Xu, C. Ma, W. Ren, H. M. Cheng, Lightweight and flexible graphene foam composites for high-performance electromagnetic interference shielding. *Adv. Mater.* **25**, 1296 (2013).  
<https://doi.org/10.1002/adma.201204196>
- [S2] Y. Wu, Z. Wang, X. Liu, X. Shen, Q. Zheng et al., Ultralight graphene foam/conductive polymer composites for exceptional electromagnetic interference shielding. *ACS Appl. Mater. Interfaces* **9**, 9059 (2017).  
<https://doi.org/10.1021/acsami.7b01017>
- [S3] H. Yang, Z. Yu, P. Wu, H. Zou, P. Liu, Electromagnetic interference shielding effectiveness of microcellular polyimide / in situ thermally reduced graphene oxide / carbon nanotubes nanocomposites. *Appl. Surf. Sci.* **434**, 318 (2018).  
<https://doi.org/10.1016/j.apsusc.2017.10.191>
- [S4] B. Shen, Y. Li, D. Yi, W. Zhai, X. Wei et al., Microcellular graphene foam for improved broadband electromagnetic interference shielding. *Carbon* **102**, 154 (2016). <https://doi.org/10.1016/j.carbon.2016.02.040>
- [S5] Z. Zeng, H. Jin, M. Chen, W. Li, L. Zhou et al., Microstructure design of lightweight, flexible, and high electromagnetic shielding porous multiwalled carbon nanotube/polymer composites. *Small* **13**, 1 (2017).  
<https://doi.org/10.1002/sml.201701388>
- [S6] Y. J. Tan, J. Li, J. H. Cai, X. H. Tang, J. H. Liu et al., Comparative study on solid and hollow glass microspheres for enhanced electromagnetic interference shielding in polydimethylsiloxane/multi-walled carbon nanotube composites. *Compos. Part B* **177**, 107378 (2019).  
<https://doi.org/10.1016/j.compositesb.2019.107378>
- [S7] P. Hu, J. Lyu, C. Fu, W. Bin Gong, J. Liao et al., Multifunctional aramid nanofiber/carbon nanotube hybrid aerogel films. *ACS Nano* **14**, 688 (2020).  
<https://doi.org/10.1021/acsnano.9b07459>
- [S8] Q. Song, F. Ye, X. Yin, W. Li, H. Li, Y. Liu, K. Li, K. Xie, X. Li, Q. Fu, L. Cheng, L. Zhang, B. Wei, et al., Carbon nanotube–multilayered graphene edge plane core–shell hybrid foams for ultrahigh-performance electromagnetic-interference shielding. *Adv. Mater.* **29**, 1 (2017).  
<https://doi.org/10.1002/adma.201701583>

- [S9] M. Li, L. Jia, X. Zhang, D. Yan, Q. Zhang et al., Robust carbon nanotube foam for efficient electromagnetic interference shielding and microwave absorption. *J. Colloid Interface Sci.* **530**, 113 (2018).  
<https://doi.org/10.1016/j.jcis.2018.06.052>
- [S10] D. Lu, Z. Mo, B. Liang, L. Yang, Z. He et al., Flexible, Lightweight carbon nanotube sponges and composites for high-performance electromagnetic interference shielding. *Carbon* **133**, 457 (2018).  
<https://doi.org/10.1016/j.carbon.2018.03.061>
- [S11] X. Shui, D. D. L. Chung, Nickel filament polymer-matrix composites with low surface impedance and high electromagnetic interference shielding effectiveness. *J. Electron. Mater.* **26**, 928 (1997).  
<https://doi.org/10.1007/s11664-997-0276-4>
- [S12] J. Ma, K. Wang, M. Zhan, A comparative study of structure and electromagnetic interference shielding performance for silver nanostructure hybrid polyimide foams. *RSC Adv.* **5**, 65283 (2015).  
<https://doi.org/10.1039/c5ra09507g>
- [S13] F. Shahzad, M. Alhabeab, C. B. Hatter, B. Anasori, S. M. Hong et al., Electromagnetic interference shielding with 2D transition metal carbides (MXenes). *Science* **353**, 1137 (2016). <https://doi.org/10.1126/science.aag2421>
- [S14] Z. Zhou, J. Liu, X. Zhang, D. Tian, Z. Zhan et al., Ultrathin MXene/calcium alginate aerogel film for high-performance electromagnetic interference shielding. *Adv. Mater. Interfaces* **6**, 1 (2019).  
<https://doi.org/10.1002/admi.201802040>
- [S15] R. Liu, M. Miao, Y. Li, J. Zhang, S. Cao et al., Ultrathin biomimetic polymeric  $\text{Ti}_3\text{C}_2\text{T}_x$  MXene composite films for electromagnetic interference shielding. *ACS Appl. Mater. Interfaces* **10**, 44787 (2018).  
<https://doi.org/10.1021/acsami.8b18347>
- [S16] F. Xie, F. Jia, L. Zhuo, Z. Lu, L. Si et al., Ultrathin MXene/aramid nanofiber composite paper with excellent mechanical properties for efficient electromagnetic interference shielding. *Nanoscale* **11**, 23382 (2019).  
<https://doi.org/10.1039/c9nr07331k>
- [S17] W. T. Cao, F. F. Chen, Y. J. Zhu, Y. G. Zhang, Y. Y. Jiang et al., Binary strengthening and toughening of MXene/cellulose nanofiber composite paper with nacre-inspired structure and superior electromagnetic interference shielding properties. *ACS Nano* **12**, 4583 (2018).  
<https://doi.org/10.1021/acsnano.8b00997>
- [S18] W. Cao, C. Ma, S. Tan, M. Ma, P. Wan et al., Ultrathin and flexible CNTs/MXene/cellulose nanofibrils composite paper for electromagnetic interference shielding. *Nano-Micro Lett.* **11**, 1 (2019).  
<https://doi.org/10.1007/s40820-019-0304-y>
- [S19] P. Sambyal, A. Iqbal, J. Hong, H. Kim, M. K. Kim et al., Ultralight and mechanically robust  $\text{Ti}_3\text{C}_2\text{T}_x$  hybrid aerogel reinforced by carbon nanotubes for electromagnetic interference shielding. *ACS Appl. Mater. Interfaces* **11**, 38046 (2019). <https://doi.org/10.1021/acsami.9b12550>

- [S20] E. Zhou, J. Xi, Y. Liu, Z. Xu, Y. Guo, et al., Large-area potassium-doped highly conductive graphene films for electromagnetic interference shielding. *Nanoscale* **9**, 18613 (2017). <https://doi.org/10.1039/c7nr07030f>
- [S21] H. Li, X. Lu, D. Yuan, J. Sun, F. Erden et al., Lightweight flexible carbon nanotube/polyaniline films with outstanding EMI shielding properties. *J. Mater. Chem. C* **5**, 8694 (2017). <https://doi.org/10.1039/c7tc02394d>