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Supporting Information

A Novel Two-Dimensional Ti₃C₂T_x MXenes/Nano-Carbon Spheres Hybrids for High-Performance Microwave Absorption

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| Etching time | S1 | S2 | S3 | S4 | S5 |
|---|--------|--------|--------|--------|--------|
| 20 (degree) | 8.823 | 8.746 | 8.563 | 8.464 | 8.323 |
| $d = n\lambda/2\sin\theta_{(\text{Å})}$ | 10.011 | 10.099 | 10.314 | 10.434 | 10.611 |

Table S1. The interlayer spacing of the $Ti_3C_2T_x$ with different etching time

 $(\lambda = 0.154 \text{ nm}; n=1)$



Figure S1. FESEM image of Ti₃AlC₂ powder after HF treatment for 12 h (S5).



Figure S2. FTIR spectra of the as-prepared $Ti_3C_2T_x$ after HF treatment at room temperature for different etching time.



Figure S3. Raman spectra of Ti_3AlC_2 before and after HF treatment at room temperature for different etching time.



Table S2. The I_D/I_G ratio of the $Ti_3C_2T_x$ with different etching time

Figure S4. High-resolution XPS. (a) XPS spectra of Ti_3AlC_2 . (b) Ti 2p spectra (c) C1s spectra and (d)

Al 2p spectra of Ti₃AlC_{2.}



Figure S5. High-resolution O 1s XPS spectra of Ti₃C₂T_x MXenes/nano-carbon-spheres hybrids with etching for 12 h.





Figure S6. (a) TEM micrograph of Ti₃AlC₂ powder after HF treatment for 12 h (S5). (b) High-resolution

TEM image of the denoted rectangular region in (a).



Figure S7. (a) The real part (ϵ') and (b) imaginary part (ϵ'') of the complex permittivity of loading 20%, 30%, 40%, 50% and 60% Ti₃C₂T_x MXenes/nano-carbon-spheres hybrids (12 h) mixed with paraffin wax at 2–18 GHz. (c), (d), (e) and (f) show reflection loss (RL) curves of 20%, 30%, 40% and 60% Ti₃C₂T_x MXenes/nano-carbon-spheres hybrids (12 h) mixed with paraffin wax at 2–18 GHz.

Figure S7 shows (a) the real part (ϵ') and (b) imaginary part (ϵ'') of the complex permittivity of loading 20%, 30%, 40%, 50% and 60% Ti₃C₂T_xMXenes/nano-carbon-spheres hybrids (12 h) mixed with paraffin wax at 2–18 GHz. With the increase of sample content, the real part (ϵ') and imaginary part (ϵ'')

of the complex permittivity increase gradually and the case of loading 60% $Ti_3C_2T_x$ MXenes/nanocarbon-spheres hybrids (12 h) is the highest. However, the loading 60% $Ti_3C_2T_x$ MXenes/nano-carbonspheres hybrids (12 h) did not exhibit excellent microwave absorption (MA) properties (shown in Figure S7f), while the loading 50% $Ti_3C_2T_x$ MXenes/nano-carbon-spheres hybrids (12 h) exhibited the best MA properties (shown in Figure 7). This is related to another important determinant of MA materials-impedance matching. The complex dielectric constant of the material is too large and is not conducive to the impedance matching of MA material. In addition, the imaginary part of the dielectric constant,

according to the free electron theory, $\varepsilon'' \approx \frac{1}{2\pi\varepsilon_0 \rho f}$ where ρ is the resistivity, suggests that high ε'' value means high conductivity, which may cause the reflection of electromagnetic wave instead of the incident, is not conducive to the absorption of electromagnetic waves.¹ Furthermore, EM absorbing materials need a suitable relative complex permittivity to meet impedance match and energy loss. Generally speaking, the real part of permittivity is required to be smaller than 30 while the imaginary part should be lower than 15.² Figure 7S(c), (d) and (e) show reflection loss (RL) curves of 20%, 30% and 40% Ti₃C₂T_x MXenes/nano-carbon-spheres hybrids (12 h) mixed with paraffin wax at 2–18 GHz.

| Samples | Matrix | Filler loading | Thickness | EAB | RL _{min} | Ref. |
|---|--------|----------------|-----------|-------|-------------------|------|
| | | (wt%) | (d;mm) | (GHz) | (dB) | |
| $Ti_3C_2T_x$ MXenes | Wax | 55 | 1.7 | 2.8 | -30 | 3 |
| Annealed- $Ti_3C_2T_x$ | Wax | 50 | 2.0 | 2.8 | -48.4 | 3 |
| Ti ₃ C ₂ T _x /CNTs | Wax | 35 | 1.55 | 4.46 | -52.9 | 4 |
| $Ti_{3}C_{2}T_{x}/Ni_{0.5}Zn_{0.5}Fe_{2}O_{4}$ | Wax | 5 | 6.5 | 3 | -42.3 | 5 |
| Co ₂ Z/Ti ₃ C ₂ Mxenes | PVB | / | 2.8 | 1.6 | -46.3 | 6 |
| Ti ₃ C ₂ | PVB | / | 2 | 0.8 | -14.4 | 6 |
| $ZnO-Ti_3C_2T_x$ | Wax | 25 | 4.0 | 1.4 | -26.3 | 7 |
| $Ti_3C_2T_x$ nanosheets | Wax | 50 | 1.4 | 5.6 | -18 | 8 |
| 2 wt%N-GP+30wt% | | / | 1.4 | 3 | -52 | 9 |
| Ti_3C_2 nanosheets | epoxy | | | | | , |
| Ti ₃ C ₂ T _x /nano-carbon- | Way | 50 | 4.8 | 1.1 | -54.7 | This |
| spheres hybrids | w ax | | | | | work |

Table S3. Comparison of EM wave absorbing properties of $Ti_3C_2T_x$ MXenes based composites.

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