Supporting Information

Self-assembly magnetized MXene avoid dual-agglomeration with enhanced interfaces for Strong Microwave Absorption through Tunable Electromagnetic Property

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**Figure S1.** XRD patterns of as-prepared MXene/Ni(OH)$_2$ composites (a) and MXene/Ni composites (b), magnification of XRD patterns of MAX, MXene, MXene/Ni(OH)$_2$-30 and MXene/Ni-30 nanocomposites (c).

**Figure S2.** SEM images of Ni(OH)$_2$ (a) and M/N-30 (b).

**Figure S3.** TEM images of MXene (a, b) and M/N-30 (c).

**Figure S4.** HRTEM images (a) and corresponding strain maps of the pure Ni (b, c). The color variation indicates the strain values ranging from -0.5 to 0.5.
Figure S5. Hysteresis loops of Ni and M/N-40 measured at 300K. The inset gives an enlarged view of the hysteresis loops (-400 Oe – 400 Oe).

Figure S6. 3D representations of RL of MAX (a) and compared RL curves about MAX, MXene and M/N-30 samples with highest microwave absorption performance (b).
**Figure S7.** Comparison of the maximum EAB of five different M/N composites.

**Figure S8.** Comparison of MXene-based composites with minimum microwave absorption value (a); comparison of MXene-based composites with RL and EAB (b).
Figure S9. TEM image (a, d), off-axis electron holograms of cutting sections (b, e), off-axis electron holograms (c, f) of pure agglomerated Ni nanoparticles and the phase amplification of the holograms are 2.
**Figure S10.** The cyclical variation of magnetic moments of the individual Ni nanoparticle (a) and numerous Ni nanoparticle (b) under an alternating magnetic field obtained from computational micromagnetic simulation. The size of circular Ni nanoparticle is 20*2 nm. The frequency of external magnetic field is 2 GHz.

**Table S1.** Comparison of microwave absorption performance among the reported MXene-based composites and the as-prepared MXenes/Ni.
<table>
<thead>
<tr>
<th>Absorber</th>
<th>$RL_{\text{min}}$ (dB)</th>
<th>Matching frequency (GHz)</th>
<th>EAB (GHz)</th>
<th>Thickness (mm)</th>
<th>refs</th>
</tr>
</thead>
<tbody>
<tr>
<td>MXene/amorphous carbon/TiO$_2$</td>
<td>-48.4</td>
<td>11.6</td>
<td>2.8</td>
<td>1.85</td>
<td>S1</td>
</tr>
<tr>
<td>MXene/ZnO</td>
<td>-26.3</td>
<td>17.4</td>
<td>1.4</td>
<td>4</td>
<td>S2</td>
</tr>
<tr>
<td>MXene/Ni$<em>{0.5}$Zn$</em>{0.5}$Fe$_2$O$_4$</td>
<td>-42.5</td>
<td>13.5</td>
<td>3</td>
<td>6.5</td>
<td>S3</td>
</tr>
<tr>
<td>MXene/PVB/Ba$<em>3$Co$<em>2$Fe$</em>{24}$O$</em>{41}$</td>
<td>-46.3</td>
<td>5.8</td>
<td>1.6</td>
<td>2.8</td>
<td>S4</td>
</tr>
<tr>
<td>MXene/Ni-modified</td>
<td>-18.2</td>
<td>16.2</td>
<td>6.3</td>
<td>1.5</td>
<td>S5</td>
</tr>
<tr>
<td>MXene/Co$_3$O$_4$</td>
<td>-34.5</td>
<td>14</td>
<td>6.3</td>
<td>2.0</td>
<td>S6</td>
</tr>
<tr>
<td>MXene/FeCo</td>
<td>-17.86</td>
<td>-</td>
<td>8.8</td>
<td>1.6</td>
<td>S7</td>
</tr>
<tr>
<td>MXene/CoFe</td>
<td>-36.29</td>
<td>8.56</td>
<td>2.64</td>
<td>2.2</td>
<td>S8</td>
</tr>
<tr>
<td>MXene/TiO$_2$/MoS$_2$</td>
<td>~ -16</td>
<td>~ 9.8</td>
<td>2.6</td>
<td>2.5</td>
<td>S9</td>
</tr>
<tr>
<td>MXene/Fe$_3$O$_4$/PANI</td>
<td>-40.3</td>
<td>15.3</td>
<td>5.2</td>
<td>1.9</td>
<td>S10</td>
</tr>
<tr>
<td>MXene/Ni chain</td>
<td>-49.9</td>
<td>11.9</td>
<td>2.1</td>
<td>1.75</td>
<td>S11</td>
</tr>
<tr>
<td>MXene/carbonyl iron</td>
<td>-15.52</td>
<td>12.8</td>
<td>8.16</td>
<td>1</td>
<td>S12</td>
</tr>
<tr>
<td><strong>MXene/Ni</strong></td>
<td><strong>-50.5</strong></td>
<td><strong>5.5</strong></td>
<td><strong>5.28</strong></td>
<td><strong>3.5</strong></td>
<td>this work</td>
</tr>
</tbody>
</table>

Compared with other MXene-based composites (Table S1), the M/N absorbers prepared in this work showed distinct advantages due to its both excellent absorption performance and wide absorption band.


S2. Qian, Y.; Wei, H.; Dong, J.; Du, Y.; Fang, X.; Zheng, W.; Sun, Y.; Jiang, Z., Fabrication of Urchin-Like ZnO-MXene Nanocomposites for High-Performance

S3. Li, Y.; Zhou, X.; Wang, J.; Deng, Q.; Li, M.; Du, S.; Han, Y.-H.; Lee, J.; Huang, Q., Facile Preparation of In-Situ Coated Ti$_3$C$_2$T$_x$/Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ Composites and Their Electromagnetic Performance. *RSC Adv.* 2017, 7 (40), 24698-24708.


**The basic principle of geometric phase analysis is as follows:**

For a perfect crystal, a HRTEM can be described as a Fourier series:

\[
I(r) = \sum_g H_g e^{2\pi ig \cdot r}
\]  
\(1\)

Where \(I(r)\) is the image intensity at the position \(r\), \(g\) is the periodicities of the Bragg reflections, the Fourier coefficients \(H_g\) can be described as:

\[
H(g) = A_g e^{ip_g}
\]  
\(2\)

Where \(A_g\) is the amplitude of the set of sinusoidal lattice fringes \(g\), \(p_g\) is the lateral position of the fringes in the original image.

In real image conditions, the Fourier coefficient \(H_g\) has conjugate symmetry.

Image strength can be expressed as the following real number function:

\[
I(r) = A_0 + \sum_{g > 0} 2A_g \cos(2\pi g \cdot r + p_g)
\]  
\(3\)
When processing the actually captured high-resolution image, the lattice image is subjected to fast Fourier transform processing to obtain an inverted space bitmap. A specific \( \pm g \) direction lattice is selected by a mask to obtain a specific direction stripe information, and then an inverse Fourier transform is performed to obtain a lattice fringe \( B_g(r) \) in the specific direction:

\[
B_g(r) = 2A_g \cos (2\pi g \cdot r + p_g)
\]  

(4)

In order to describe the lattice changes caused by distortion and defects in the material, the amplitude and phase of the lattice fringes should be expressed by the functions \( A_g(r) \) and \( P_g(r) \) for the position \( r \), which should be written as:

\[
B_g(r) = 2A_g(r) \cos (2\pi g \cdot r + p_g(r))
\]  

(5)

**The basic principle of electron holography analysis is as follows:**

According to the simplified Poisson’s equation about the electrostatic field theory, the charge density can be calculated from

\[
\rho(\chi) = -\varepsilon_\gamma \varepsilon_0 \frac{\partial^2 \psi(\chi)}{\partial \chi^2}
\]  

(6)

where \( \rho(\chi) \) is the charge density, \( \chi \) is the distance, \( \varepsilon_\gamma \) and \( \varepsilon_0 \) are the relative dielectric constant in sample and the dielectric constant in vacuum, respectively.