Supporting Information

High-compact MXene-based coatings by controllable interfacial structure

Jiheng Ding^a, Hao Wang^{a, c}, Hongran Zhao^{a, b}, Mohammad Raza Miah^{a, b}, Jinggang

Wang^{a, b, *}, Jin Zhu^{a, b}

a. Ningbo Institute of Materials Technology and Engineering, Chinese Academy of

Sciences, Ningbo 315201, P. R. China.

b. University of Chinese Academy of Science, Beijing 100049, PR China.

c. School of Materials Science and Chemical Engineering, Ningbo University, Ningbo

315201, China.

*Corresponding authors:

wangjg@nimte.ac.cn (J WANG).

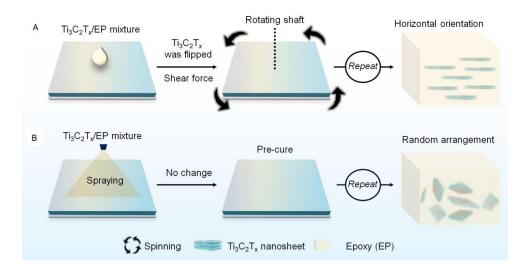


Fig. S1. Schematic illustration of the fabrication process and the structure of OMC (A) and RMC (B) coatings. The MXene nanosheets are flipped from the disordered distribution into well spread and parallelly aligned under the high-speed shear force during the rotating. For the spraying process, MXene nanosheets are partly re-stacking and aggregation during due to the intermolecular van der Waals forces.

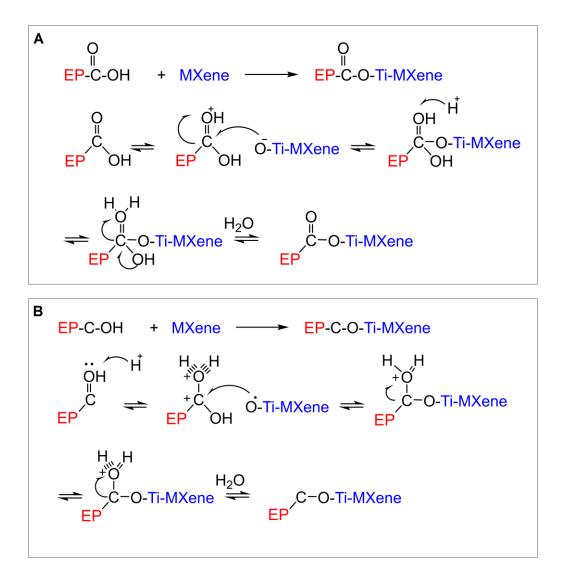


Fig. S2. The stabilization mechanisms of MXene nanosheets in EP matrix. The –OH and –COOH of EP can for Ti-O-C covalent bonds with MXene nanosheets to inhibit the oxidative degradation of MXene during the service time.

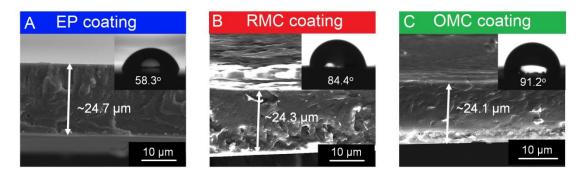


Fig. S3. Cross-sectional SEM images of EP (A), RMC (B), and OMC (C) coatings, insets shows the corresponding water contact angles. The thickness of the coatings is ca. 25 μm according to the cross-sectional SEM images in Fig.S2A-C. Besides, the dispersion states of MXene nanosheets can have an impact on the water contact angles. The uniform dispersion of MXene nanosheets can better cross-link with the EP matrix, and thus lead to a higher water contact angle than that of RMC.

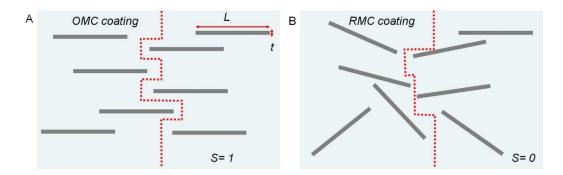


Fig. S4. The tortuosity of OMC (A) and RMC (B) coatings with different distributed MXene nanosheets.

The tortuosity (τ) of the coatings can be determined, as given by ^[1-3]:

$$\tau = 1 + \frac{L}{t} \times \varphi \times \frac{1}{3} \left(S + \frac{1}{2} \right) \tag{1}$$

Where, *L*, *t* and φ are the lateral size, thickness, and mass content of fillers, respectively. Accordingly, the orientation degree S of MXene in the coating matrix can be obtained as the following:

$$S = \frac{1}{2}(3\cos^2\theta - 1) \tag{2}$$

Where, θ is the orientation angle of MXenes in the coating matrix. Especially, when MXenes are parallel arrangement, the average orientation angle $\theta = 0^\circ$, S = 1; when MXenes are random distribution, the average orientation angle $\theta = 54.7^\circ$, S = 0^[4].

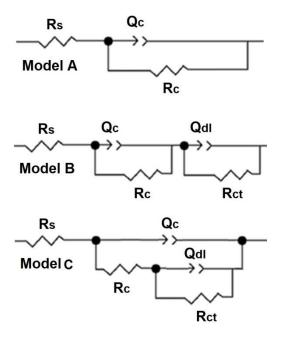


Fig. S5. The equivalent electric circuits for fitting EIS results. In the equivalent electric circuits, R_s , R_c , R_{ct} , Q_c , Q_{dl} represent solution resistance, coating resistance, charge-transfer resistance, coating capacitance, and double-layer capacitance, respectively ^[5]. The equivalent electric model A was used to fit the EIS results of complete coating system. The equivalent electric model B and C were for the EIS results of defected coatings.

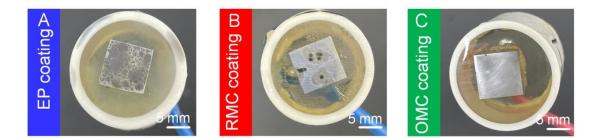


Fig. S6. Digital photos of EP (A), RMC (B), and OMC (C) coatings protected steels after immersion in 3.5wt% NaCl solutions for 15 days. Compared with the EP, the RMC shows a typical localized corrosion. This indicates that the essential conductivity of MXene can trigger galvanic corrosion behaviour during the long-term immersion. The digital photo is the direct evidence for the galvanic corrosion.

Table S1. EIS parameters of different coatings protected steel electrodes during the 15days immersion in 3.5wt.% NaCl solutions.

Sample	Time/d	Z _{0.01} /ohm cm ²	R _c /ohm cm ⁻²	f _b /Hz
EP	1	1.58×10^{6}	6.83×10^{5}	1122
	2	1.08×10^{6}	9.20×10^{5}	1175
	5	1.29×10^{6}	1.12×10^{6}	1202
	10	1.99×10^{6}	1.76×10^{6}	1230
	15	1.54×10^{6}	1.95×10^{5}	1585
RMC	1	1.37×10^{9}	1.18×10^{9}	1.25
	2	6.01×10^{7}	2.68×10^{7}	15.8
	5	1.74×10^{7}	1.39×10^{7}	63.1
	10	1.22×10^{7}	2.25×10^{6}	70.8
	15	1.89×10^{7}	2.15×10^{6}	112.2
ОМС	1	4.64×10^{9}	3.79×10^{9}	0.18
	2	6.72×10^{9}	5.53×10^{9}	0.16
	5	7.48×10^{9}	6.29×10^{9}	0.16
	10	9.11×10^{9}	8.15×10^{9}	0.16
	15	6.84×10^{9}	6.08×10^{9}	0.16

Sample	Time/d	Z _{0.01} /ohm cm ²	R _{ct} /ohm cm ⁻²
EP	1	1.11×10^{5}	1.65×10^{5}
	2	6.40×10^4	4.50×10^{4}
	3	4.35×10^{4}	4.21×10^{4}
-	1	1.79×10^{4}	3.14×10^{5}
RMC	2	1.51×10^{4}	1.97×10^{4}
	3	6.45×10^{3}	8.23×10^{3}
-	1	7.20×10^{5}	5.37×10^{5}
OMC	2	7.20×10^{5}	1.23×10^{5}
	3	5.39×10^{5}	1.23×10^{5}

Table S2. EIS parameters of different scratched coatings protected steel electrodesduring the 3 days of immersion in 3.5wt.% NaCl solutions.

Reference.

- [1] Y. Shi, C. Chen, Y. Li and W. Zhao, Carbon, 2023, 201, 1048–1060.
- [2] Q. Zhao, Z. Lu, Y. Wu and W. Zhao, Compo. Sci. Technol., 2022, 217, 109090.
- [3] X. Fan, H. Yan, M. Cai, S. Song, Y. Huang and M. Zhu, *Composites Part B*, 2022, 231, 109581.
- [4] C. S. Lu and Y. W. Mai, *Phys. Rev. Lett.*, 2005, 95, 088303.
- [5] C. Liu, H. Zhao, P. Hou, B. Qian, X. Wang, C. Guo and L. Wang, ACS Appl. Mater. Interfaces, 2018, 10, 36229.