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Supporting Information

Magnetic-Field-Free Separation and Recovery of MXene-Doped GO Composites via Dye-Bridged Reassembly in Dye Adsorption

Jin Yang^{a,b}, Xiaoxia Yan^{a,d}, Yifei Qin^{a,b}, Dazhi Zhang^a, Jingyi Zhao^a, Yi Yu^{a,b,c},
Qingjun Liu^{a,b *}, Wenshuai Jiang^{a,b,c *}

^a School of Medical Engineering, Henan Medical University, Xinxiang, Henan 453003, People's Republic of China

^b Xinxiang Key Laboratory of Neurobiosensor, Xinxiang, Henan 453003, People's Republic of China

^c Henan Engineering Technology Research Center of Neural Sensing and Control, Henan 453003, People's Republic of China

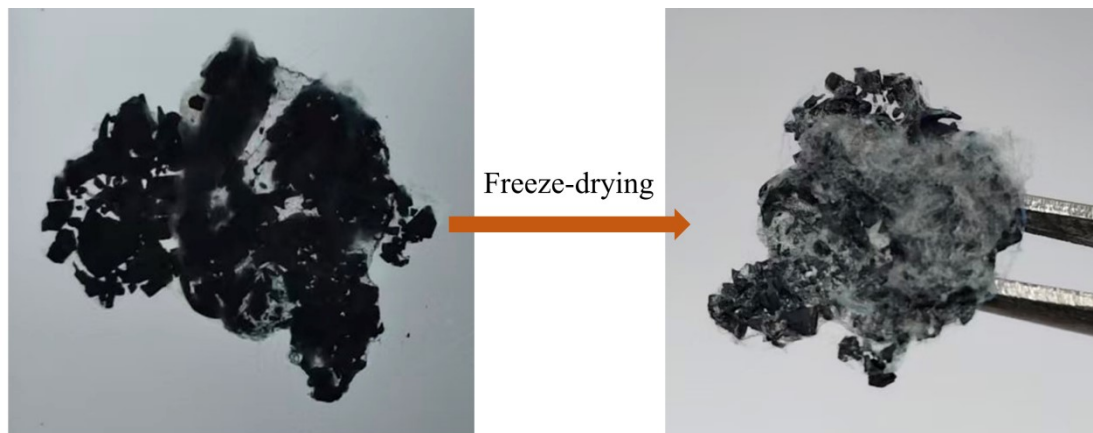
^d School of Digital Technology Industry, Zhengzhou Sias University, Henan 451150, People's Republic of China

* Corresponding author. E-mail addresses: 171024@xxmu.edu.cn (Wenshuai Jiang), qjliu@zju.edu.cn (Qingjun Liu).

21 Supplemental Figures

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25 Fig. S1. Dye-bridged reassembly of the GO/MXene composite accompanied by white

26 flocculent

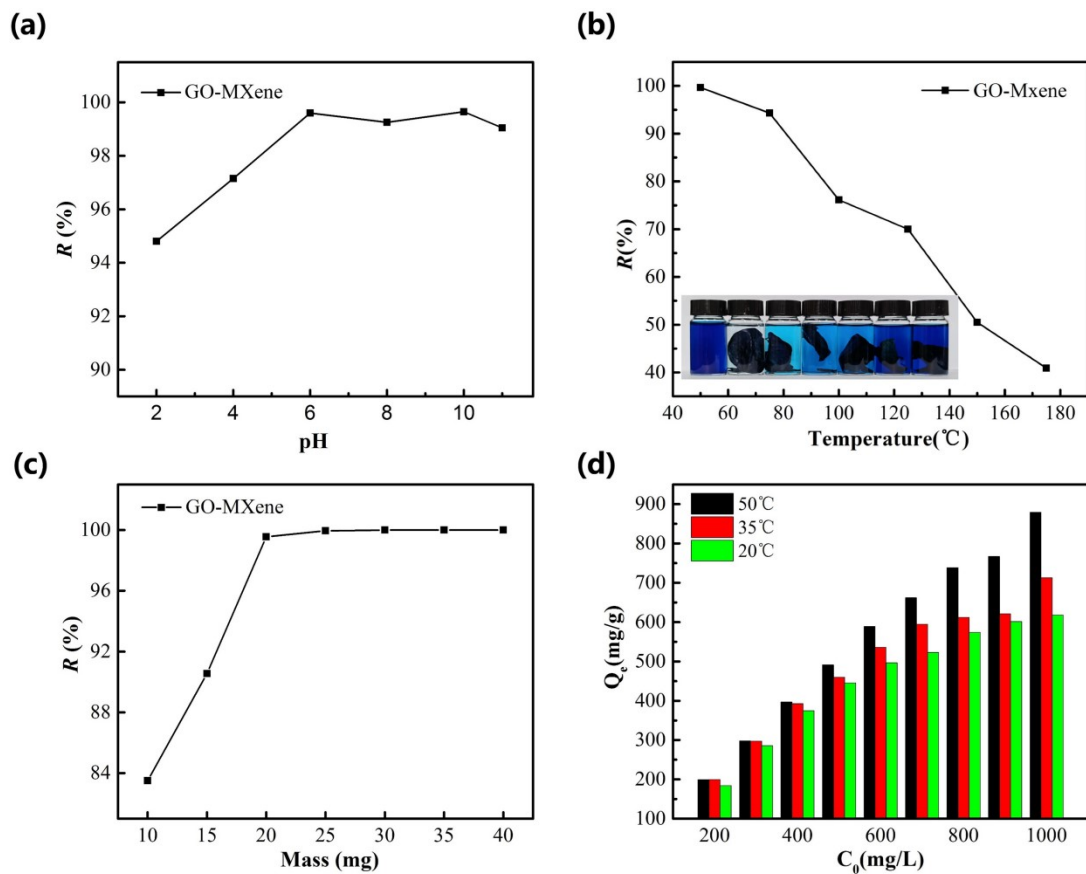
connections.

27 In this study, 20 mg of an intact GO/MXene composite membrane was added to
28 20 mL of MB solution at an concentration of 200 mg/L. After the adsorption
29 equilibrium was reached, the removal effect was measured, and the results are shown
30 in Fig. S2a. According to Fig. S2a, the GO/MXene membrane showed a good
31 removal effect (removal rate > 94%) under acidic, neutral and alkaline conditions,
32 indicating that the pH of the solution did not significantly affect on the MB removal
33 rate. However, at $\text{pH} \geq 6$, the GO/MXene composite membrane was able to adsorb
34 more MB, and the MB removal rate reached more than 99%. Considering that the pH
35 of the 200 mg/L MB solution used in this study was approximately 6, we did not
36 adjust the pH of the solution further in subsequent experiments.

37 In addition, we found that the MB removal efficiency of the GO/MXene
38 membrane decreased with increasing membrane preparation temperatures. In
39 this study, samples of 20 mg of GO/MXene membranes prepared at different
40 temperatures were taken separately and placed into 20 mL of an MB solution with a
41 concentration of 200 mg/L. The discoloration effect is shown in the inset of Fig. S2b,
42 which indicates that the GO/MXene membranes prepared at 50 °C obtain faster MB
43 discoloration. As the temperature of the membrane treatment increases, the
44 discoloration effect becomes weaker. Furthermore, we measured and calculated the
45 MB removal rates, and the results are shown in Fig. S2b. According to Fig. S2b, the
46 removal rate of MB by the GO/MXene membrane prepared at 50 °C can reach
47 ~99.7%, and the removal rate gradually decreases with increasing membrane
48 treatment temperature, which is consistent with the discoloration results of MB in the
49 inset of Fig. S2b. These results indicate that the treatment temperature of the
50 GO/MXene membrane affects the adsorption of MB, and the higher the membrane
51 treatment temperature, the worse the removal rate and the removal effect.

52 The effect of the adsorbent dose on the MB removal rate was also investigated.
53 In this study, by placing GO/MXene membranes with different masses (10–40 mg)
54 into 20 mL of an MB solution at a concentration of 200 mg/L, we investigated the
55 effect of the adsorbent dose on the removal of MB. Based on the results presented in
56 Figs. S2c, the removal of MB was approximately 100% when the adsorbent dosage
57 was 20 mg; thereafter, the removal rate hardly changed as the adsorbent dose
58 increased, which may be attributed to the overlapping of the active surfaces at high
59 doses. Therefore, we used 20 mg of the GO/MXene membrane as an adsorbent in all
60 subsequent studies on the adsorption of MB.

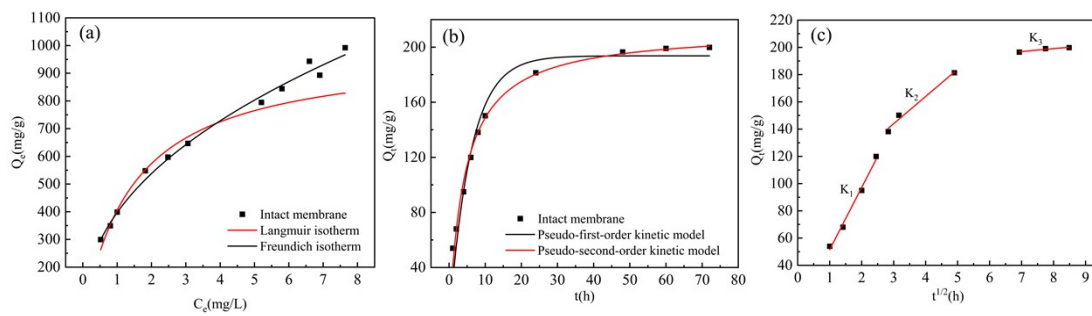
61 Temperature is an important factor that affects the adsorption process. In
 62 practical applications, large temperature changes and seasonal alternations occur in
 63 the process of dye wastewater discharge that often affect the adsorption of dyes.
 64 Therefore, it was necessary to investigate the effect of temperature on the adsorption
 65 process. To determine the effect of temperature on the adsorption process of MB, we
 66 completed adsorption experiments of MB at 20, 35, and 50 °C, and the results are
 67 shown in Fig. S2d. In this study, the adsorption capacity and removal effect of MB
 68 increased with increasing temperature, suggesting that the adsorption of MB on
 69 GO/MXene is endothermic.



70

71 Fig. S2. (a) Effects of initial solution pH, (b) GO/MXene membrane preparation
 72 temperature, (c) adsorbent dose; (d) temperature on MB adsorption.

73 Fig. S3a shows the adsorption isotherms of the GO/MXene membranes and the
 74 fitting results obtained using the Langmuir and Freundlich isotherm equations. Table
 75 S1 summarizes the Langmuir and Freundlich isotherm model constants and
 76 correlation coefficients for the adsorption of MB onto intact GO/MXene membranes.
 77 According to Fig. S3a, the maximum adsorption capacity of the intact GO/MXene
 78 membrane is 994.5 mg/g. Combined with the fitted parameters in Table S1, when the
 79 adsorbent is an intact GO/MXene membrane, the Freundlich isotherm model ($R^2 =$
 80 0.992) provides a better description.



81

82 Fig. S3. Intact membrane. (a) Langmuir and Freundlich adsorption isotherms, (b)
 83 pseudo-first-order and pseudo-second-order kinetic models, (c) intraparticle diffusion
 84 model.

85 Table S1. Fitting parameters of the Langmuir and Freundlich models for the intact
 86 membrane

Langmuir			Freundlich		
Q_m (mg/g)	K_L (L/mg)	R^2	$1/n$	K_F $((L/mg)^{1/n} \cdot mg/g)$	R^2
982.97	0.701	0.972	0.43	397.17	0.992

87

88 Based on the results presented in Fig. S3b, c, and Table S2, the pseudo-second-
 89 order model ($R_2^2 = 0.992$) better fitted the kinetic adsorption behavior of MB on
 90 GO/MXene. The relatively low k_2 indicates slower adsorption kinetics, which may be
 91 attributed to the complex surface morphology of GO/MXene. During the adsorption
 92 process, MB was transferred and diffused in various ways. These results suggest that
 93 the adsorption process of MB on GO/MXene membranes is mainly influenced by
 94 chemisorption, which may be accompanied by ion exchange or electron sharing.

95

96 Table S2. Kinetics parameters for the adsorption of MB on intact GO/MXene
 97 membrane

pseudo-first-order model			pseudo-second-order model		
k_1	Q_{e1}	R_1^2	k_2	Q_{e2}	R_2^2
h^{-1}	mg/g		$g/(mg \cdot h)$	mg/g	
0.169	193.6	0.963	0.001	212.7	0.990

98

99 The fitting results shown in Fig. S3d suggest that MB adsorption on the
 100 GO/MXene membrane can be divided into three successive stages. In addition, as can
 101 be observed in Fig. S3d and Table S3, none of the linear relationships exceeded the
 102 origin, and the linear correlation coefficients were greater than 0.9, indicating that
 103 intraparticle diffusion was not the only speed-control step.

104 Table S3. Parameters of the intraparticle diffusion model of intact membrane

Stage I			Stage II			Stage III		
K_1	C_1	R_1^2	K_2	C_2	R_2^2	K_3	C_3	R_3^2
$mg \cdot g^{-1} \cdot h^{-0.5}$	mg/g		$mg \cdot g^{-1} \cdot h^{-0.5}$	mg/g		$mg \cdot g^{-1} \cdot h^{-0.5}$	mg/g	
45.7	5.87	0.991	19.9	84.3	0.985	2.1	182.0	0.917

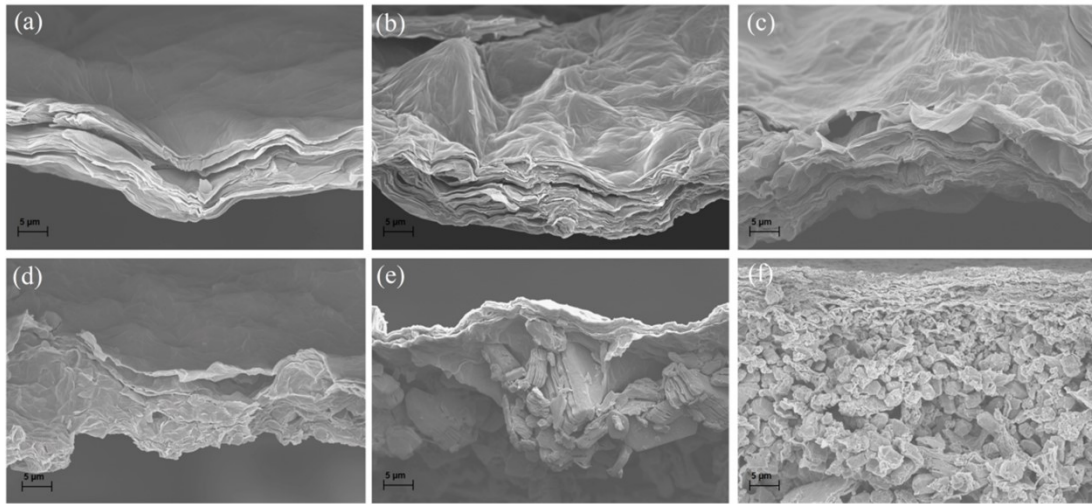
106 Furthermore, we investigated the MB adsorption effect of the GO/MXene
107 composite membranes prepared by doping different proportions of MXene, and the
108 results are shown in Fig. S4. According to Fig. S4, the GO/MXene membranes
109 prepared by doping with 20 wt% MXene exhibit faster discoloration, indicating a
110 faster adsorption rate and adsorption effect. In addition, the results presented in Fig.
111 S4 indicate that with an increase in the MXene doping ratio, the adsorption effect of
112 the GO/MXene composite membrane on MB worsens. This is partly explained by the
113 poorer MB adsorption effect of MXene, and may be related to the decrease in
114 agglomeration and adsorption sites caused by the increase in MXene content in the
115 membrane formation process. Considering that the GO/MXene composite membrane
116 prepared by doping with 20 wt% MXene has a faster MB removal rate, we focused on
117 the removal of MB by the GO/MXene composite membrane prepared with this
118 doping proportion in the following study. Without any special indication, all the
119 GO/MXene membranes described below are GO/MXene composite membranes
120 prepared by doping with 20 wt% MXene.



121
122 Fig. S4. Effect of different MXene content in intact membranes on MB adsorption.
123 From left to right: MB without membrane, MB with GO membrane, MB with
124 GO/MXene-10% membrane, MB with GO/MXene-20% membrane, MB with
125 GO/MXene-40% membrane, MB with GO/MXene-60% membrane and MB with
126 GO/MXene-80% membrane.

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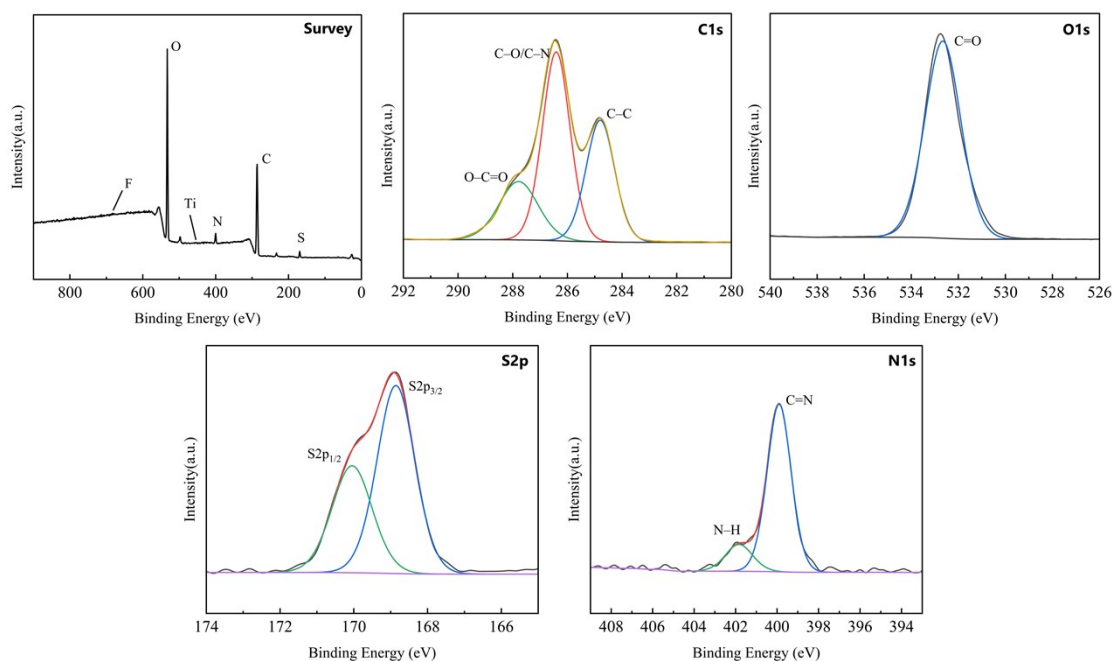
128 Fig. S5 shows the SEM results of the cross-section of the GO/MXene
129 membranes prepared by doping with different proportions of MXene. These results
130 indicate that an increase in the doped MXene content led to the agglomeration of
131 MXene during the membrane formation process.



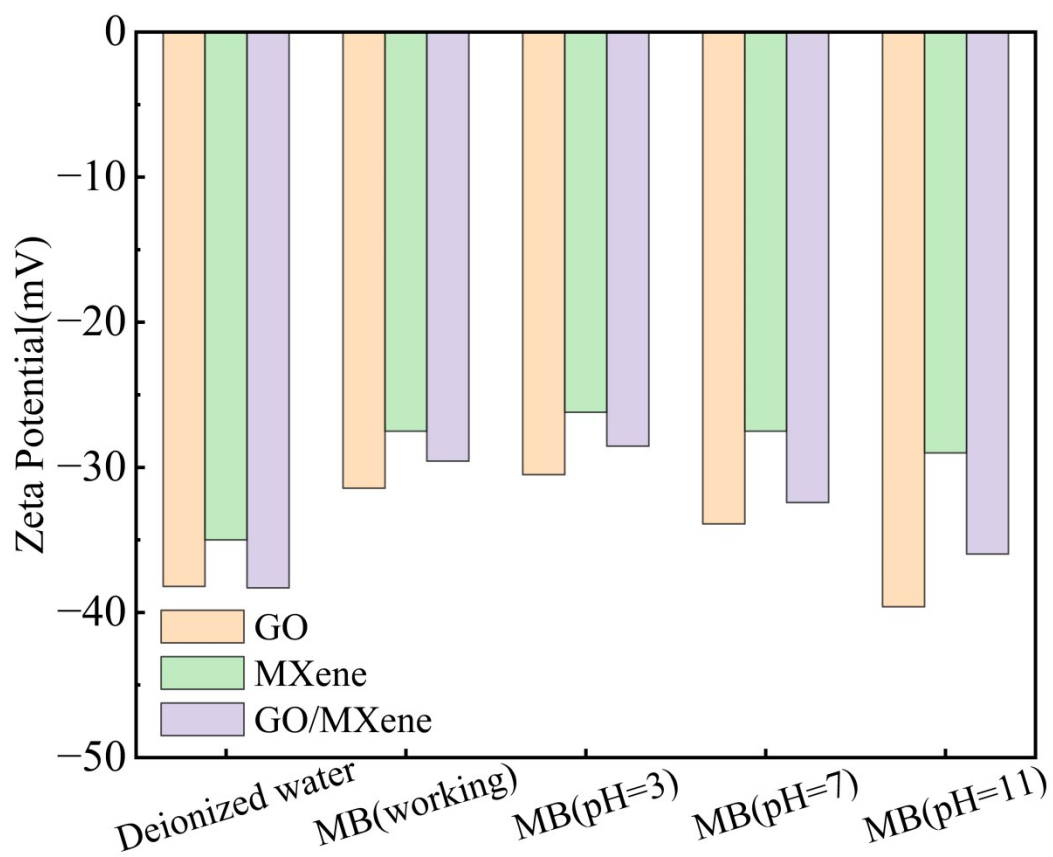
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133 Fig. S5. SEM results of GO/MXene membranes with different doping ratios.

134 XPS was used to investigate the flocculent connections in the dye-bridged
135 reassembly. As shown in Fig. S6, the survey spectrum reveals the coexistence of C, O,
136 Ti, F, N, and S in the flocculent connections. Although the intensities of Ti and F are
137 relatively weak, these signals confirm the presence of both GO/MXene fragments and
138 MB molecules in the flocculants. In the high-resolution C1s spectrum, the peaks at
139 284.8, 286.41, and 287.79 eV were attributed to C–C, C–O/C–N, and O–C=O bonds,
140 respectively. The high-resolution O1s spectrum exhibited a peak at 532.66 eV
141 corresponding to C–O bonds. The high-resolution S 2p spectrum showed peaks at
142 170.05 and 168.86 eV, corresponding to S 2p_{1/2} and S 2p_{3/2}, respectively. The high-
143 resolution N1s spectrum showed peaks at 399.90 and 401.84 eV, corresponding to
144 C=N and N–H, respectively.

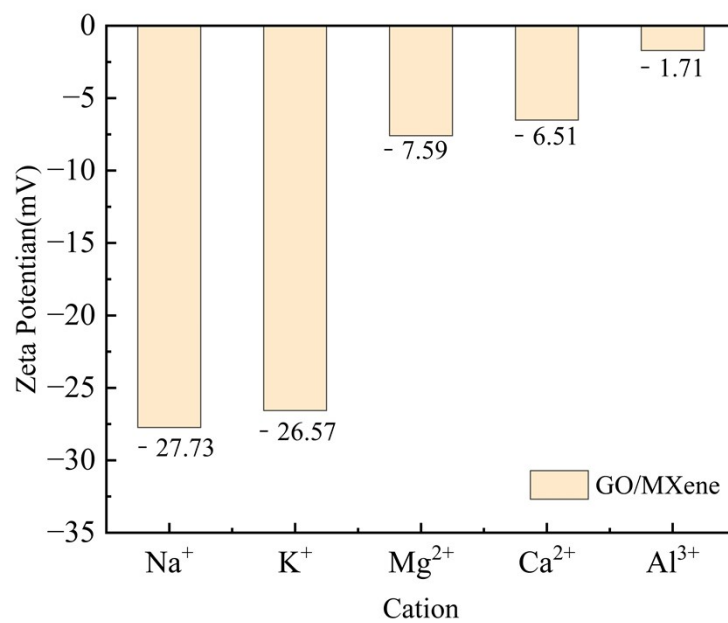


145
146 Fig. S6. XPS survey and high-resolution spectra (C1s, O1s, S2p, N1s) of the
147 flocculent connections.



148

149 Fig. S7. Zeta potentials of GO, MXene, and GO/MXene composite fragments in
 150 deionized water, working-concentration MB solution, and MB solutions at pH 3, 7,
 151 and 11.



152

153 Fig. S8. Zeta potential of GO/MXene composites in MB solutions containing different

154 cations