

Bioinspired, Ultra-Light and Sandwich structured MXene-AgNWs/cellulose nanofiber porous film for excellent electromagnetic interference shielding with Joule Heating Performance

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1 **Bioinspired, Ultra-Light and Sandwich structured MXene-AgNWs/cellulose**
2 **nanofiber porous film for excellent electromagnetic interference shielding with**
3 **Joule Heating Performance**

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12 **Abstract:** With the rapid development and popularization of intelligent, portable, and wearable
13 flexible electronic devices, urgently required a new generation electromagnetic interference (EMI)
14 shielding materials to manage the increasing serious radiation pollution. In this work, ultrathin,
15 lightweight, and flexible porous films with reasonable strength were fabricated via vacuum filtered
16 the cellulose nanofiber (CNF) dispersion on both sides of the MXene-AgNWs film and followed
17 by a freeze-drying process. The prepared porous composite film presents to be a typical sandwich-
18 structured with dense in surface and porous inside. This novel and unique structure endows the
19 sandwich-structured porous film with greatly improved EMI performance to 67.5 from 40 dB,
20 enhanced absorption coefficient from 0.1 to 0.4, and satisfactory mechanical properties compared
21 to the conventional sandwich-structured films. Furthermore, the prepared films present the
22 remarkable low-voltage-driven Joule heating performance. Therefore, ultrathin, lightweight,

23 flexible, and versatile properties CNF-MXene-AgNWs composite porous film with an excellent
24 EMI-shielding performance is hold great potential in the fields of aerospace, portable and wearable
25 electronics.

26 **Keywords:** MXene, AgNWs, Cellulose nanofiber (CNF), Sandwich-structured porous film,
27 Electromagnetic interference (EMI) shielding, Joule heating performance

28 **Introduction**

29 The rapid development of telecommunication technology and wearable electronic devices has
30 made electromagnetic radiation pollution problems, seriously affecting the normal operation of
31 adjacent equipment and even human health (Guo, Ren, Lu, et al., 2022; Iqbal, Shahzad, et al., 2020;
32 J. Liu et al., 2022; J. Wang, Ma, Zhou, Du, & Teng, 2022; Wei et al., 2020; Y. Zhang, Ruan, & Gu,
33 2021). Thus, it is very necessary to reduce the intensity of electromagnetic radiation to an
34 acceptable range. Electromagnetic interference (EMI)shielding materials are capable of solving
35 electromagnetic pollution (Guo et al., 2023; Guo, Ren, Wang, et al., 2022; Y. Zhang & Gu, 2022).
36 Very recently, MXene, as newly emerged two-dimensional transition metal carbide/nitride,
37 presents exceptional metal-like conductivity, which is widely exploited in the field of
38 electromagnetic shielding (M.-S. Cao et al., 2019; Iqbal, Sambyal, & Koo, 2020; H. Liu et al.,
39 2022; Oliveira & Gusmao, 2020; Z. Wang, Cheng, Fang, Hou, & Xie, 2020; Yun et al., 2020). For
40 example, Gogotsi and his co-workers firstly fabricated a free-standing Ti_3C_2Tx -based film (45 μm)
41 with excellent conductivity ($4665 S cm^{-1}$) and superior EMI shielding effectiveness (SE)
42 of $>92 dB$ (Shahzad et al., 2016). Since the conductivity of pure MXene film is too high to meet
43 the impedance matching, which is easy to cause a large number of surface reflection. Therefore, it
44 is necessary to improve the attenuation performance of Ti_3C_2Tx MXene composite film for

45 electromagnetic waves by carefully designing the morphology through interface engineering.

46 In order to alleviate the above predicament, more explorations are attempted to optimize the
47 preparation process by foam methods (Deng, Tang, Wu, Zhang, & Yu, 2021; X. Wu et al., 2020;
48 Zhu et al., 2021). After foaming, the composite not only exhibits low density and lightweight by
49 introducing a large number of holes, but also enhances the internal absorption and dissipation
50 efficiency of electromagnetic waves by creating multiple interfaces (Q. Chen, Zhang, Huang, Li,
51 & Yuan, 2022; Z. Chen, Xu, Ma, Ren, & Cheng, 2013; Han et al., 2019). However, the traditional
52 formation strategies of foamed/porous structure, such as freeze-drying, supercritical CO₂ foaming,
53 and the template method, inevitably interrupt the construction of the conductive network by
54 introduced air bubbles and reduce the overall shielding performance (Fan et al., 2020; H.-B. Zhang,
55 Yan, Zheng, He, & Yu, 2011). Thus, to achieve ideal EMI SE, a large thickness is usually needed,
56 which will result in poor flexibility and mechanical strength.

57 The developed foamed composites with efficient EMI shielding performance in recent years
58 were of ultrathin, light weight, flexible, and high absorption characteristics (Liu et al., 2017; Tang
59 et al., 2022; Y. Zhang, Ruan, Shi, et al., 2021). Zhang et al. successfully prepared Ti₃C₂T_x-based
60 graphene porous composite films (Ti₃C₂T_x-rGO) by decomposition of NH₄HCO₃ as foaming agent
61 during heat treatment (350 °C, 2h and hydrogen-argon atmosphere). Due to the synergic effect of
62 the high electrical conductivity and porous structure, the obtained Ti₃C₂T_x-rGO with a thickness of
63 60 μm achieved the maximum value of 59 dB. In addition, Yu and co-workers fabricated
64 lightweight and flexible MXene foams with desirable strength and superior EMI Shielding via the
65 hydrazine-induced foaming process. A greatly enhanced EMI-shielding performance was achieved
66 for the MXene foam (~70 dB) compared to its unfoamed film (~50 dB) because of its favorable

67 porous structure (Liu et al., 2017). Nevertheless, MXene foam are confronted with the issue of
68 poor mechanical properties due to loosing pore structure. Besides, the heating treatment requires
69 harsh conditions and hydrazine-induced foaming process does harm to human health and
70 environment, which limit the large-scale application of the foam (Tang et al., 2022). Therefore,
71 how to realize the rational construction of ultrathin, lightweight, flexible, and high mechanical
72 strength MXene porous film with excellent EMI SE becomes a crucial challenge.

73 Cellulose nanofibers (CNF) produced from plant-derived renewable resources display
74 outstanding mechanical properties (e.g., Young's modulus up to 250 GPa, intrinsic flexibility, etc.),
75 high aspect ratio, low density, and smooth surface (Lapka et al., 2023). Recently quite a few
76 researchers have chosen CNF as flexible substrate and reinforcing fillers to support MXene (Bai et
77 al., 2022; W.-T. Cao et al., 2018; Yuehu Li, Chen, Liu, Zhang, & Qi, 2021; Wan et al., 2021). For
78 instance, Chen et al used CNFs to fabricate MXene-based composite film, which has great
79 mechanical property (Zhan, Song, Zhou, & Lu, 2019). However, the insulated CNF blocks the
80 electrons transmission between MXene layers, thus, decreasing the EMI performance property.
81 Therefore, it is still a great challenge to fabricate MXene/CNF composites with excellent
82 mechanical strength and EMI shielding property.

83 Skeleton, one of the hardest tissue of a person or animal, has a complex internal with a
84 honeycomb-like three-dimensional structure and a dense external structure, thus exhibiting high
85 mechanical strength while reducing weight. As exemplified by the case of skeleton, the
86 construction of skeletal structures (sandwich bionic porous structure) can effectively improve the
87 mechanical properties of materials, electromagnetic shielding properties and meet the requirements
88 of light weight. Therefore, in this work, we demonstrate a facile approach to prepare the sandwich

89 bionic porous MXene-AgNWs/CNF by vacuum filtering the CNF dispersion on both sides of the
90 porous MXene-AgNWs film. The outer CNF layers not only endow the composite film with
91 excellent mechanical strength, but effectively improve the oxidation resistance. More importantly,
92 the porous internal structure of MXene-AgNWs greatly enhanced EMI-shielding performance and
93 strong absorption capacity. In addition, the prepared composite porous films present high Joule
94 heating temperature at relatively low applied voltages and rapid Joule heating response. This work
95 provides novel insights for the designing multifunctional EMI shielding materials for aerospace,
96 artificial intelligence, and next-generation flexible wearable electronic devices.

97 **1. Experimental**

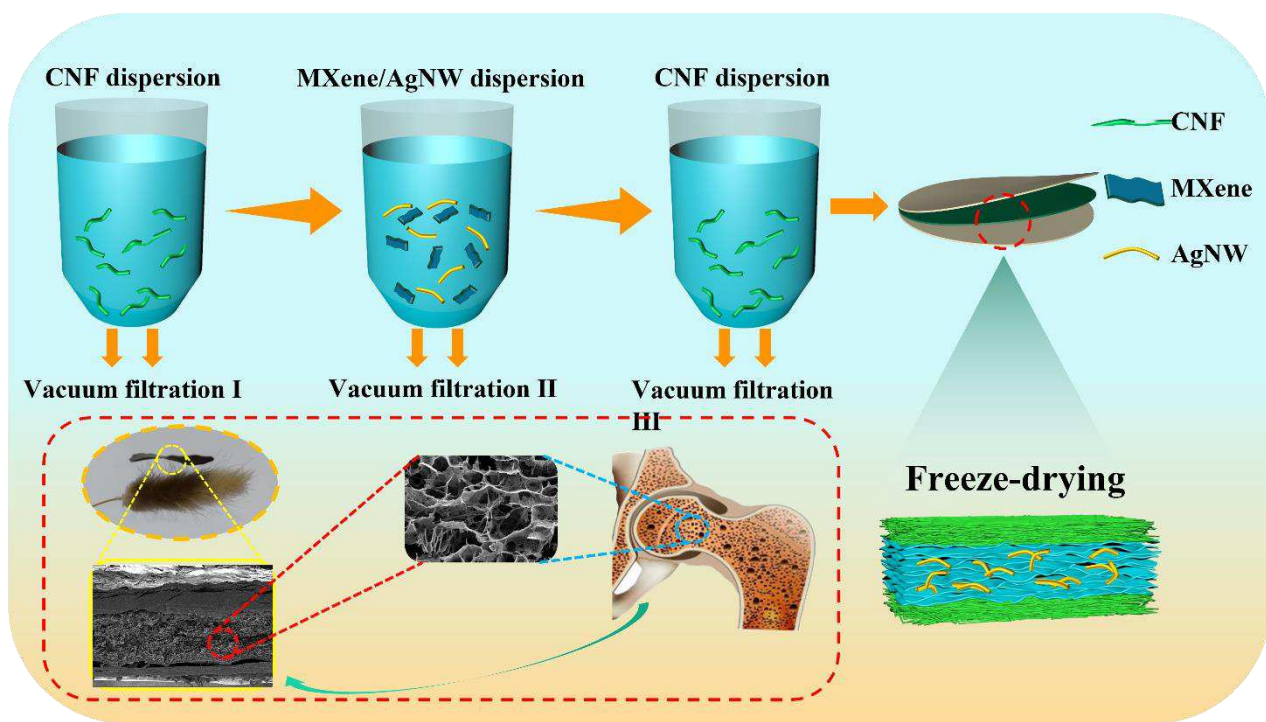
98 *2.1 Materials*

99 MAX (Ti_3AlC_2 powder, 400 meshes) were bought from Jilin 11 Technology Co., Ltd. Silver
100 nanowires (AgNWs) suspension (10 mg/mL, 20-30 nm in diameter, 10-15 μm in length), was
101 supplied by Zhejiang Kechuang Advanced Materials Co., Ltd. Cellulose nanofiber (CNF) aqueous
102 solution (1 wt%, 0.8-2.0 μm in length) was obtained from Tianjin Woodelf biotechnology Co., Ltd.
103 Lithium fluoride (LiF , $\geq 99.0\%$, AR) and hydrochloric acid (HCl , 35–37 wt% in H_2O) were
104 provided from Shanghai Macklin Biochemical Co., Ltd., China. All chemicals were used as
105 received without further purification.

106 *2.2 Fabrication of the sandwich bionic MXene-AgNWs/CNF porous film*

107 $\text{Ti}_3\text{C}_2\text{T}_x$ MXene was synthesized according to our previous works (F. Zhang et al., 2022;
108 Zong et al., 2022). As illustrated in Figure 1, the sandwich bionic MXene-AgNWs/CNF porous
109 films were prepared via a controlled vacuum-assisted filtration and freeze-drying methods. Firstly,
110 12.5 mL of CNF dispersion with a concentration of 2 mg/mL was vacuum filtered (vacuum

111 filtration I) onto the poly(ethersulfone) membrane until the formation of a stable CNF hydrogel.
112 Then, a homogeneous mixed dispersion of 50 mg of $Ti_3C_2T_x$ MXene and AgNWs in different
113 ratios of 10:0, 9:1, 7:3, 5:5, and 3:7, respectively, was stirred ultrasonically and deposited on the
114 above of the CNF hydrogel (vacuum filtration II). Thereafter, 12.5 mL of CNF dispersion with a
115 concentration of 2 mg/mL was deposited on the topmost layer (vacuum filtration III) in the same
116 way. Finally, the obtained sandwich-structured hydrogels were frozen longitudinally under liquid
117 nitrogen until the films were completely frozen and formed, and then the films were vacuum
118 freeze-dried to obtain sandwich bionic MXene-AgNWs/CNF composite porous films. For
119 simplicity, the sandwich bionic MXene-AgNWs/CNF porous films were labeled as CM_xA_yC ,
120 where x and y represent the mass ratio of the $Ti_3C_2T_x$ MXene and AgNWs, respectively. The
121 prepared composite film showed a honeycomb-like three-dimensional structure inside, exhibiting
122 lightweight characteristics.



123

124 Figure. 1. Schematic diagram of the manufacturing process of laminated bionic composite films.

125 *2.4 Characterization*

126 The structural morphologies and microstructures of the samples were observed by the field
127 scanning electron microscopy (SEM, FEI Inspect-F, Finland) and transmission electron
128 microscopy (TEM, JEM 2100, JEOL). X-ray photoelectron spectra (XPS, XSAM800, Japan) were
129 employed to analyze the surface chemical compositions and chemical bond of Ti₃C₂T_x MXene.
130 The phase compositions of the samples were characterized by X-ray diffraction (XRD, TTRIII,
131 Rigaku, Japan). The resistance of samples was measured by a Keithley electrometer Model 4200-
132 SCS (USA) according to a two-point method. The electrochemical workstation (RST 5200,
133 Suzhou Resitest Electronic) was used to record the voltammetric current signals. The electrical
134 conductivity of the sandwich bionic CNF-MXene/AgNWs composite porous films was calculated
135 as Eq.1:

$$136 \quad \sigma = L/RA \quad (1)$$

137 where σ is the electrical conductivity (S cm⁻¹), R is the resistance (Ω), L is the length of the
138 test samples (cm), and A is the cross-sectional area of the measured samples (cm²). The
139 mechanical tests of samples were performed at RT by a universal testing machine (UTM2103)
140 with a sample rectangle size of 25 × 5 mm² and loading rate of 0.5 mm/min. The surface
141 temperature of the composite film was recorded using an infrared thermal imager (Fluke Ti300).

142 The EMI SE of the as-prepared films was measured by an Agilent N5247A vector network
143 analyzer. All samples were cut into cylindrical shape with the diameter of 13 mm for
144 measurements. The scattering parameters (S₁₁ and S₂₁) in the frequency range of 8.2-12.4 GHz
145 were recorded, and the coefficients of reflectance (R), absorption (A) and transmittance (T), EMI
146 SE (SE_T), microwave reflection (SE_R) and microwave absorption (SE_A) were calculated using the

147 following equations:

$$148 \quad R = |S_{11}|^2, T = |S_{21}|^2 \quad (2)$$

$$149 \quad A = 1 - R - T \quad (3)$$

$$150 \quad SE_R(\text{dB}) = -10 \log(1 - R), SE_A(\text{dB}) = -10 \log(T / (1 - R)) \quad (4)$$

$$151 \quad SE_T(\text{dB}) = SE_R + SE_A + SE_M \quad (5)$$

152 Where SE_A is the absorption value, SE_R is the reflection value, and SE_M is the microwave
153 multiple internal reflections, which can be negligible when SE_T is higher than 10 dB (Abbasi,
154 Antunes, & Ignacio Velasco, 2019; Kim, Lee, Kim, & Lee, 2010). In order to fairly compare the
155 effectiveness of the shielding materials, the density and thickness of the shielding materials are
156 also considered. The related equations were described as:

$$157 \quad SSE(\text{dB cm}^3 \text{g}^{-1}) = \text{EMI SE} / \text{density} \quad (6)$$

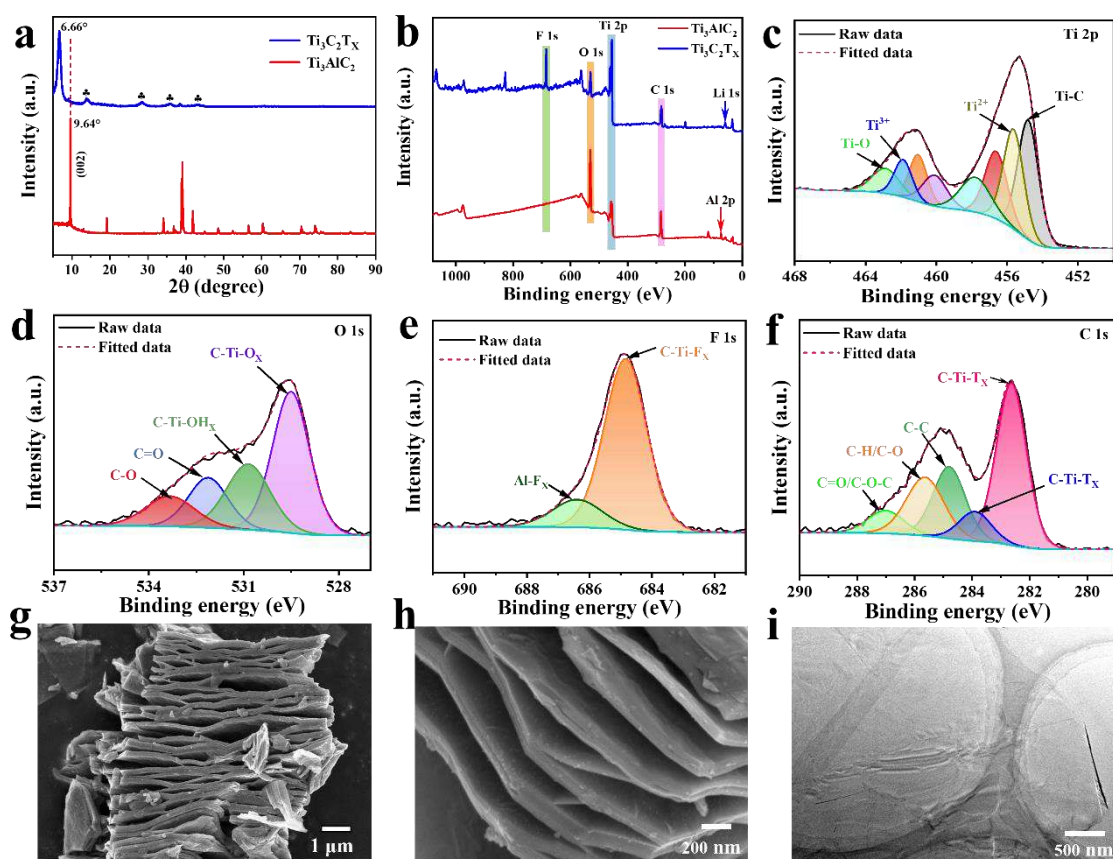
$$158 \quad SSE/t(\text{dB cm}^2 \text{g}^{-1}) = \text{SSE} / \text{thickness} \quad (7)$$

159 **2. Results and discussion**

160 *3.1 Morphology and structure characterization*

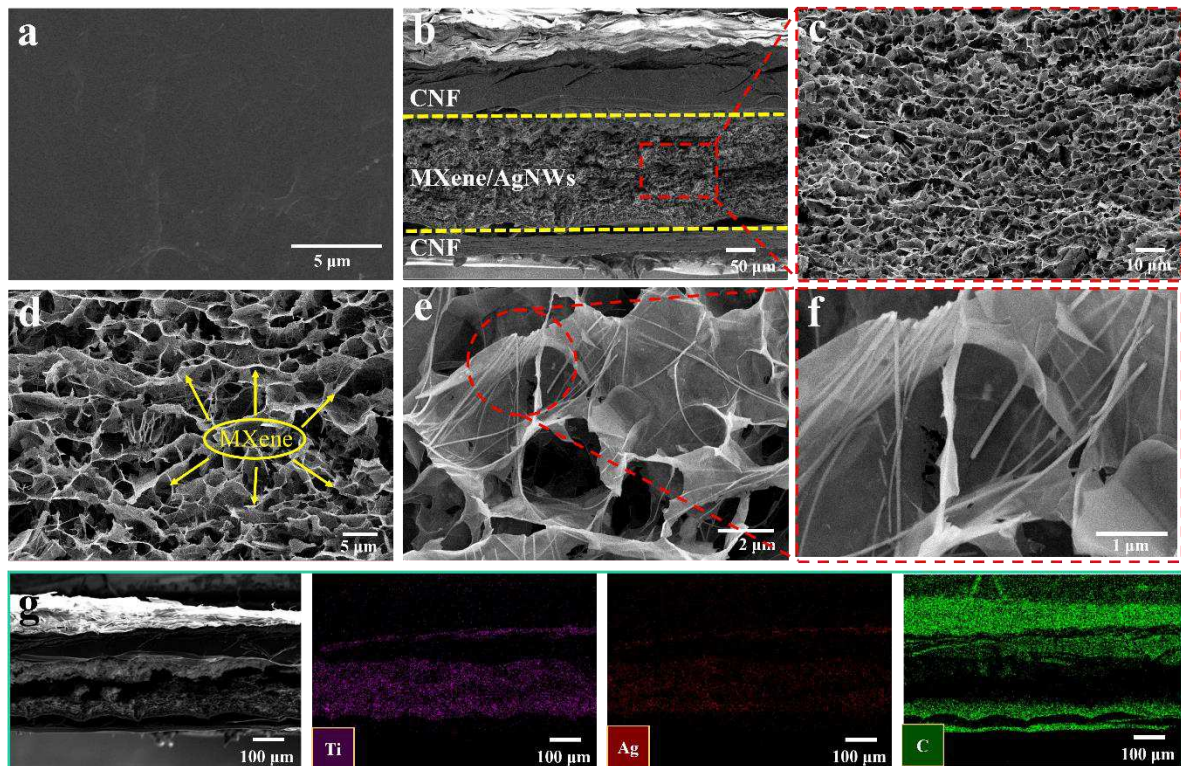
161 Figure 2a shows the XRD patterns of $\text{Ti}_3\text{C}_2\text{T}_x$ MXene and Ti_3AlC_2 MAX. After etching, the
162 absence of the main peak (104) located at 39° and the peak (002) shifting from 9.64° to 6.66° ,
163 indicate the successful synthesis of MXene (Zhu et al., 2021). The XPS wide-scan spectra of the
164 Ti_3AlC_2 MAX and $\text{Ti}_3\text{C}_2\text{T}_x$ MXene, as well as the high-resolution spectra of Ti 2p, O 1s, F 1s and
165 C 1s are displayed in Fig. 2b–f. Al 2p at around 74 eV for Ti_3AlC_2 MAX disappears completely,
166 indicating the Al layer is selectively etched. The Ti 2p spectrum shows two peaks of Ti 2p_{1/2} and Ti
167 2p_{3/2}, containing Ti–C (460.2 and 455.1 eV), Ti²⁺ (460.9 and 456.0 eV), Ti³⁺ (462.1 and 457.1 eV)
168 and Ti–O (463.2 and 458.3 eV). The four peaks located at 530.1, 530.6, 532.5, and 533.8 eV,

169 assigned to C–Ti–O_x, C–Ti–OH_x, C=O and C–O, are detected in O1s, indicating the existence of
 170 oxidized region. The F 1s spectrum exhibits two main peaks (684.8 eV and 686.4 eV), which
 171 belong to C–Ti–F_x and Al–F_x, respectively. Five peaks appear in C 1s spectrum: C–Ti–T_x bond at
 172 284.1 and 282.5 eV, C–C bond at 284.8 eV, C–H/C–O bond at 285.6 eV, and C–O/C–O–C bond at
 173 287.0 eV. The morphology and structure of the prepared Ti₃C₂T_x was characterized by SEM and
 174 TEM, and the corresponding results are presented in Fig. 2g-i, respectively. The prepared Ti₃C₂T_x
 175 exhibits a typical accordion-like microstructure (Fig.2g-h), which further demonstrated that Al
 176 layer has been successfully etched. The TEM image of Ti₃C₂T_x MXene dispersion shows clear
 177 transparent feature and few-layer 2D nanosheets (shown in Fig.2h).



178
 179 Figure. 2. (a) X-ray diffraction (XRD) patterns and (b) X-ray photoelectron spectroscopy (XPS)
 180 survey spectra of Ti₃AlC₂ MAX and delaminated Ti₃C₂T_x MXene. High resolution XPS spectra of
 181 (c) Ti 2p; (d) O 1s; (e) F 1s and (f) C 1s of Ti₃C₂T_x MXene. (g, h) Scanning electron microscopy
 182 (SEM) images and (i) Transmission electron microscopy (TEM) image of Ti₃C₂T_x MXene

183 Figure 3 presents the surface and cross-sectional SEM images of the CM₇A₃C. As shown in
184 Fig. 3a, the composite film exhibits a relatively smooth and dense structure over the entire surface.
185 From the Fig. 3b-f, the CM₇A₃C exhibits obvious morphological difference between outer-layer
186 and core-layer. The cellulose nanofibers tightly interwoven together on both sides of the CM₇A₃C,
187 exhibiting a smooth cross-sectional morphology, while the core-layer of MXene-AgNWs shows a
188 honeycomb-like three-dimensional structure (Fig. 3c-d), which provides the material with
189 lightweight. Note that the MXene sheets are not separated individually, but partially glued to each
190 other to form a continuous cellular structure due to the sublimation of the ice during freeze-drying
191 (Fig.3d). It can be clearly observed in Fig.3e-f that AgNWs are randomly cross-distributed
192 between MXene lamellae as interlayer "linkers" and "hardeners", which are conducive to the
193 formation of a dense multi-channel conductive network. The cross-sectional SEM images of
194 CM₃A₇C are depicted in Fig. S1. As can be seen, a large number of AgNWs interwove with each
195 other, and the internal pores decreased significantly due to the reduction of MXene. In addition,
196 the distribution of each component in CM₇A₃C was investigated by element mapping, and Fig. 3g
197 shows the element selective distribution of Ti, C, and Ag in the composite film. In particular, the
198 distribution of Ti and Ag elements is in perfect agreement with the distribution of the intermediate
199 layer MXene and AgNWs observed in the SEM images, further demonstrating the composite film's
200 layered porous structure.



201

202 Figure 3. SEM images of the CM₇A₃C (a) surface SEM image, (b-g) Cross-sectional SEM images,
 203 and the elemental mapping (g) of the CM₇A₃C sample.

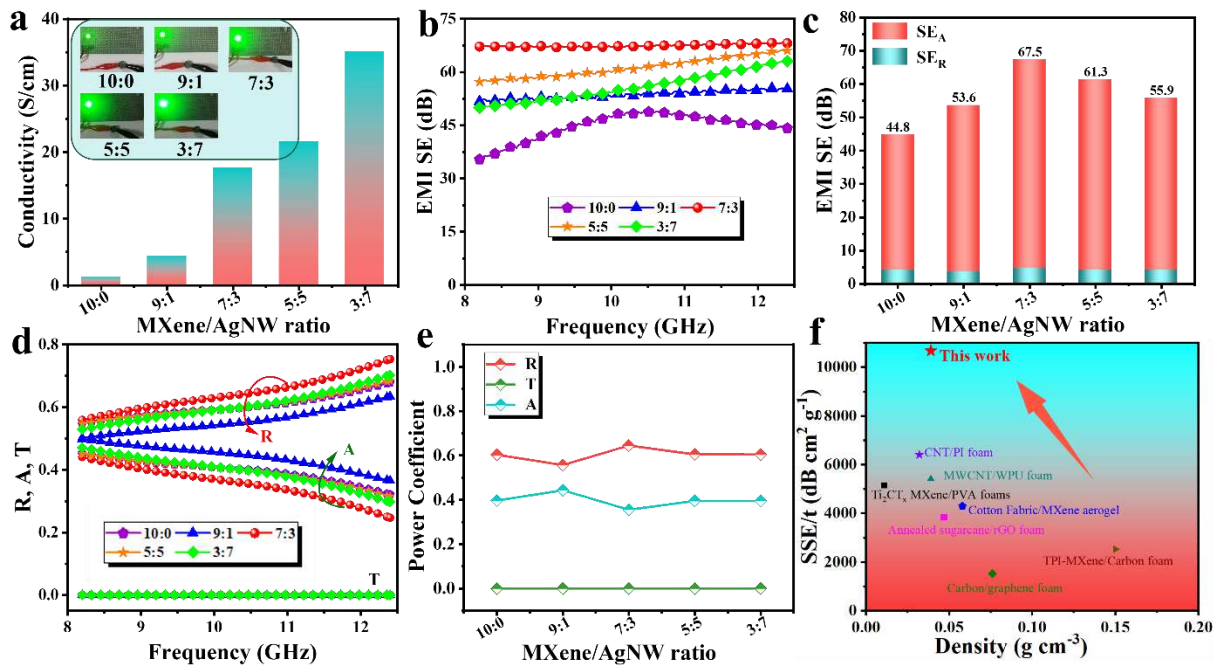
204 *3.2. Electrical and EMI shielding performance of the sandwich bionic MXene-AgNWs/CNF*
 205 *composite porous films*

206 The electrical conductivity (σ) of a material is an important influence on its EMI SE. High
 207 electrical conductivity is often conducive to excellent electromagnetic shielding performance (Tan,
 208 Gou, Zhang, Ding, & Wang, 2023). The σ of the obtained CM_xA_yC is shown in Fig. 4a. The
 209 conductivity of the composite films showed an upward trend, increasing from 1.22 S/cm to 35.09
 210 S/cm as the ratio of MXene and AgNWs changed from 10:0 to 3:7. This is mainly due to the
 211 tightly connected conductive network as the content of AgNWs increases. The conductivity
 212 evolution was more clearly from the brightness of the LED lamp, which further reflects the change
 213 of the σ at different ratios of MXene and AgNWs.

214 Figures 4b and c display the EMI shielding performance of the sandwich bionic MXene-

215 AgNWs/CNF porous films in X-band (8.2-12.4 GHz). As expected, the fabricated composite
216 porous films exhibit high EMI SE due to its outstanding electrical conductivity and porous
217 structure. Besides, the EMI SE enhanced dramatically first and then decreased with increasing
218 AgNWs ratios (Fig. 4b). Specifically, the electromagnetic shielding values of the composite films
219 with MXene and AgNWs ratios of 10:0, 9:1, 7:3, 5:5 and 3:7, reached 44.8 dB, 53.6 dB, 67.5 dB,
220 61.3 dB, and 55.9 dB, respectively. The best electromagnetic shielding performance of the
221 prepared films was achieved at the MXene and AgNWs ratio of 7:3. This phenomenon is due to
222 the perfect porous structure inside the composite film when the MXene/AgNWs ratio is 7:3. The
223 reflection efficiency (SE_R) and absorption efficiency (SE_A) derived from the SE_T are employed to
224 evaluate the EMI shielding mechanism (Fig. 4c). As the ratio of MXene and AgNWs changes, the
225 SE_T and SE_A of the composite porous films exhibit a clear variation. At the same time, SE_A
226 contributes more to EMI SE than SE_R . However, all of the conductive films have SE_R values in
227 excess of 3 dB, manifesting that over 50% of the EMWs are reflected. Thus, SE_R cannot estimate
228 the EMW reflected from the surface of the material due to the impedance mismatch. Based on it,
229 power coefficients such as reflection coefficient (R), absorption coefficient (A) and transmission
230 coefficient (T) are calculated to evaluate the real EMI shielding mode. Fig. 4d-e show the R-A
231 coefficients of all the composite porous films. As illustrated, the R values are always a little bit
232 higher than A values, which implies a reflection-dominated shielding mechanism. Despite this,
233 compared to conventional composite films prepared by vacuum-assisted filtration in which more
234 than 90% of the EMWs are reflected at the surface, the composite porous films prepared in this
235 paper effectively improve the absorption loss of incident EMWs. Furthermore, Fig. S2 plots the
236 power coefficients of the CM_7A_3C at X-band compared with the films with 7:3 ratio of MXene to

237 AgNWs (named C-F) obtained by the conventional vacuum-assisted filtration method. The A
 238 coefficient of CM₇A₃C can reach 0.4, which is much higher than 0.1 for C-F. In summary, the
 239 prepared sandwich-structured porous composite film exhibits improved absorption loss of incident
 240 electromagnetic waves. Meanwhile, Fig. S2 compares the EMI SE of the sample C-F and CM₇A₃C.
 241 As can be seen, CM₇A₃C is substantially improved to 67.5 dB from 40 dB of C-F. In practical
 242 application, in order to fairly compare the effectiveness of the shielding materials and eliminate the
 243 effect of density and thickness on the electromagnetic shielding performance, the specific
 244 shielding performance (SSE) and SSE/t of the CNF-MXene/AgNWs composite porous film is
 245 introduced to evaluate and compare with the reported literature. As shown in Figure 4f and table 1,
 246 the Mxene-AgNWs/CNF composite films prepared in this paper exhibit excellent SSE/t values,
 247 with the highest SSE/t value reaching 10675 dB·cm²·g⁻¹.



248
 249 Figure. 4. (a) Conductivity, (b-c) EMI SE and (d-e) Power coefficients composite films with
 250 different ratios of MXene and AgNWs (10:0, 9:1, 7:3, 5:5 and 3:7, respectively). (f) Comparison
 251 of SSE/t as a function of thickness with previous reports.

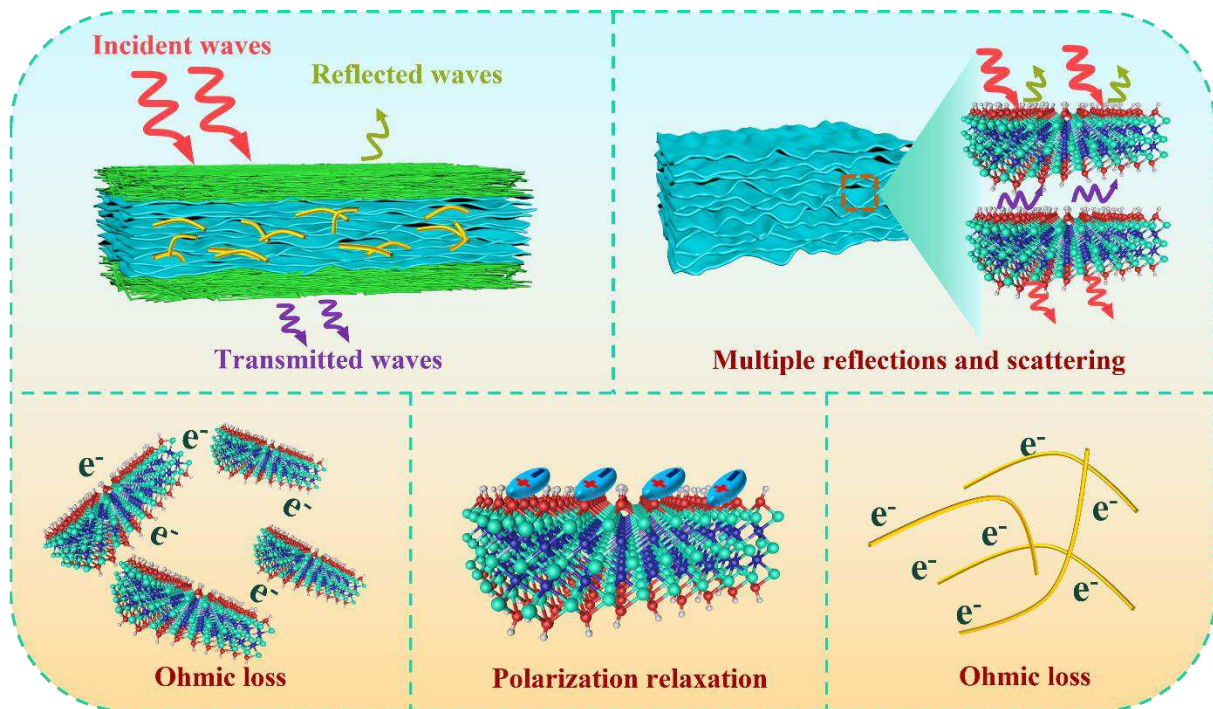
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253 Table 1. Comparison of EMI shielding performance of the sandwich bionic Mxene-AgNWs/CNF
 254 porous film and other porous materials.

Samples	EMI SE (dB)	Density (g cm ⁻³)	SSE/t (dB cm ² g ⁻¹)	Ref.
TPI-MXene/Carbon foam	44.7	0.15	298	(Jia, Shen, Zhang, & Zheng, 2021)
Graphene/PI aerogel	28.8	0.076	1518	(Yu, Dai, Yuan, Zou, & Liu, 2020)
Carbon/graphene foam	51	0.72	970	(Yang Li et al., 2016)
CNT/PI foam	41.1	0.0321	6402	(Y.-Y. Wang et al., 2020)
Cotton Fabric/MXene aerogel	42.1	0.058	4299	(Zhai et al., 2022)
Annealed sugarcane/rGO foam	53	0.047	3830	(L. Wang, Shi, Zhang, Zhang, & Gu, 2020)
MWCNT/WPU foam	21.1	0.039	5410	(Zeng et al., 2016)
Ti ₂ CT _x MXene/PVA foam	28	0.0109	5136	(Xu et al., 2019)
MXene/silver nanowire/PU foam	50	0.15	2525	(Cheng et al., 2021)
CNF/Mxene-AgNWs porous film	67.5	0.158	10675	This work

255 To better elucidate the EMI shielding mechanism, the speculated EMWs transmission path is
 256 depicted in Fig.5. When the incident EMWs strike the CNF layers, nearly all of the EMWs can
 257 readily enter the porous films due to the good impedance matching. When the incident EMWs
 258 reach the porous MXene-AgNWs conductive layer, the EMWs will undergo reflection, absorption,
 259 and multiple reflection in porous structured layer. Specifically, as EMWs pass through the MXene-
 260 AgNWs porous layer, the sheet-like pores acting as shielding walls, effectively extend the internal
 261 electromagnetic wave transmission path, and the multiple internal reflections and scatterings allow
 262 the remaining electromagnetic waves to continue to be attenuated by conductive losses. This
 263 "absorption-reflection-reabsorption" process continues until the electromagnetic waves are
 264 completely depleted.

265



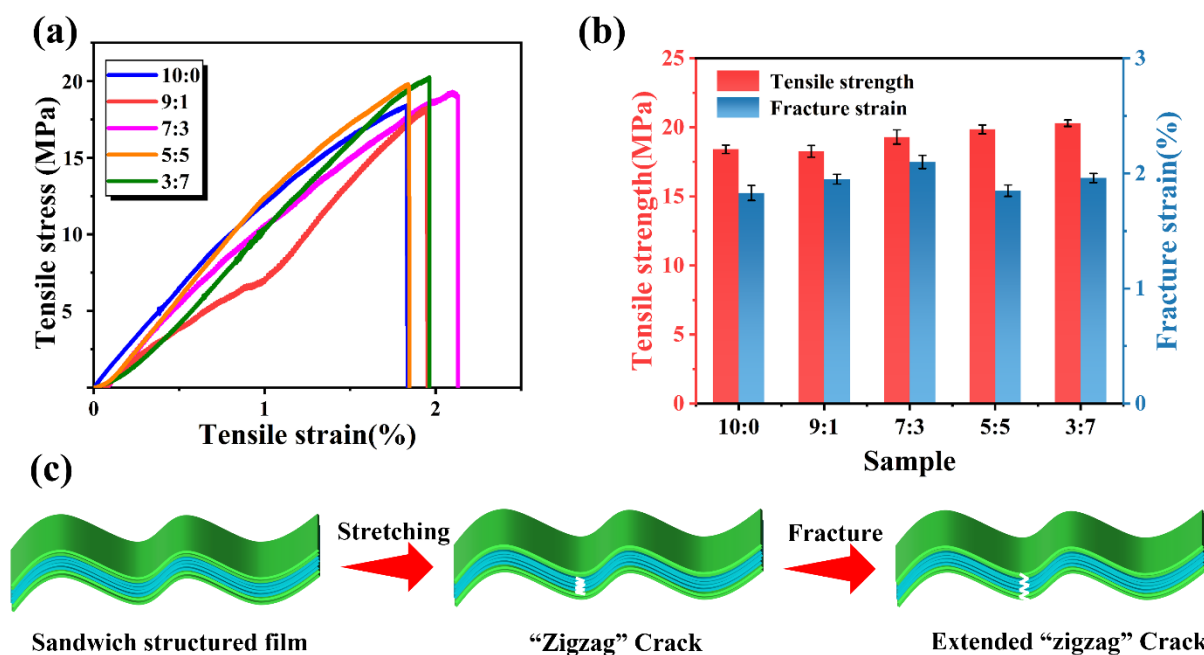
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267 Figure 5. Schematic diagram of electromagnetic wave transmission on a composite CNF-
 268 Mxene/AgNWs film with a sandwich bionic structure.

269 *3.3. Mechanical properties of the sandwich bionic MXene-AgNWs/CNF composite porous films*

270 Apart from remarkable EMI shielding property, excellent mechanical property is also an
 271 important parameter for the practical application of next-generation EMI shielding materials. Thus,
 272 tensile test was used to evaluate the mechanical property of the composite porous films, and the
 273 relevant results are shown in Fig. 6a-b. Pure $Ti_3C_2T_x$ MXene films are brittle fracture due to poor
 274 interfacial interactions, which makes them unsatisfactory for practical application requirements
 275 (Quero & Rosenkranz, 2021). The prepared sandwich bionic CNF/Mxene-AgNWs porous film can
 276 improve the mechanical property to the maximum extent because of the presence of bilateral CNF
 277 lamellar structures. Figures 6a shows representative stress-strain curves for MXene-
 278 AgNWs/CNFporous film. In the range of $Ti_3C_2T_x$ MXene and AgNWs ratios from 10:0 to 3:7, the
 279 tensile strength and elongation at break of the sandwich bionic composite porous films reached
 280 considerably high levels of 18.2-20.3 MPa and 1.8-2.1% (Figures 6b). Besides, the abundant

281 functional groups of the CNF (seen in Fig.S3) are also beneficial for chemical crosslinking of the
 282 MXene, significantly improving mechanical property and oxidation stability (N. Wu et al., 2023) .
 283 In order to more clearly illustrate the mechanism of strengthening toughness, the fracture process
 284 of MXene-AgNWs/CNF with sandwich structure under tension is depicted in Figure 6c. The
 285 fracture mechanism can be elucidated as follows: the MXene-AgNWs layer ruptures firstly due to
 286 weak interlayer interactions. Then, the robust CNF layer prevents the initial crack in the rigid but
 287 brittle MXene-AgNWs layer from spreading throughout the film. When CNF reaches the ultimate
 288 load, the cracks propagate to the interface and spread to the CNF layer in nanoscale "zigzag" crack
 289 paths, while micrometer-scale "zigzag" crack paths are formed between the layers.

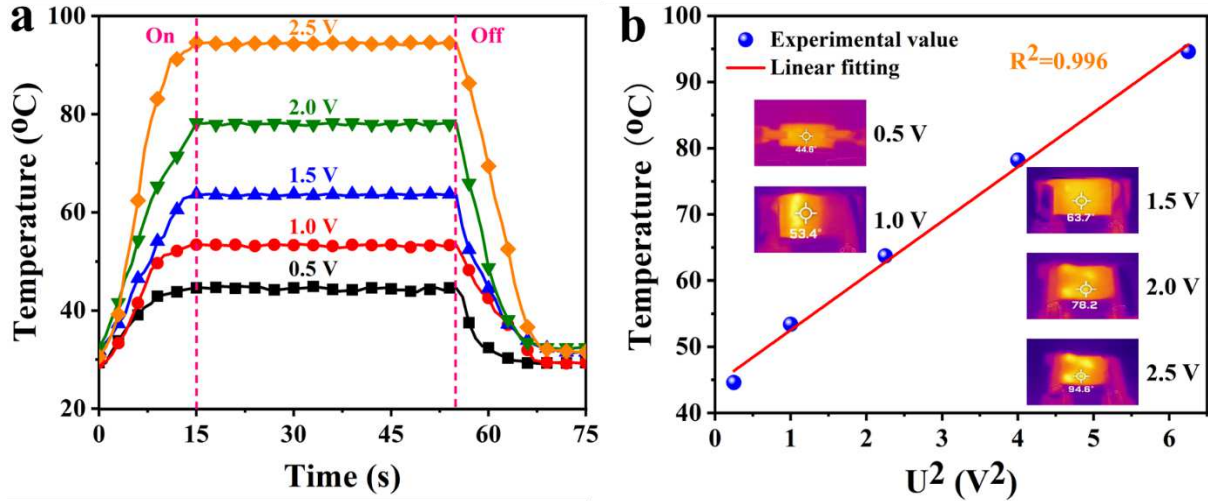


290
 291 Figure. 6. Mechanical properties of the composite films with MXene and AgNWs ratios of 10:0,
 292 9:1, 7:3, 5:5 and 3:7: (a) Tensile stress-strain curves; (b) Tensile strength and fracture strain; (c)
 293 Schematic diagram of the fracture mechanism.

294 3.3. Joule heating performance

295 According to Joule's law ($Q = \frac{U^2 t}{R}$, where Q, U, R and t are the generated heat, applied voltage,
 296 resistance of the material and working time, respectively), the high conductivity confers a

297 satisfactory Joule heat capacity to the prepared MXene-AgNWs/CNT films, ensuring the proper
298 operation of the electrons in the device under extremely cold conditions (Hu et al., 2022; L. Zhang
299 et al., 2022). Figure 7a represents the surface temperature of the prepared MXene-AgNWs/CNT
300 films as heaters recorded by the infrared imager at low applied voltages of 0.5-2.5 V. The surface
301 temperature of all the heaters rapidly reaches a maximum and then remains stable, indicating a
302 state of equilibrium between the photothermally induced and dissipated heat. After the voltage
303 turned off, the temperature drops rapidly to the initial value. Obviously, the greater the voltage
304 applied, the higher the equilibrium temperature. The equilibrium temperature is 44.5 °C when the
305 voltage is 0.5 V, and when the applied voltage rises to 1.5 V, it greatly rises to 63.5 °C, and further
306 increases to 94.6 °C at the voltage of 2.5 V. After unplugging the voltage, the temperature drops
307 below 30 °C within 13 s. In addition, the plateau saturation temperature of the MXene-
308 AgNWs/CNT film was linear to the square of the applied voltage (Figure 7b), in accordance with
309 Joule's law. The strong dependence of the surface temperature on the applied voltage indicates that
310 the prepared composite films have a highly controllable joule heating capacity. The temperature
311 variation of the MXene-AgNWs/CNT film surface when the applied voltage was varied from 0.5
312 to 2.5 V was further shown by infrared thermal images (see the insets in Fig. 7b). The obtained
313 thermal infrared images show a uniform temperature distribution, which is an important criterion
314 as an electric heater. These results indicate that the prepared MXene-AgNWs/CNT exhibit low
315 trigger voltage and outstanding Joule heating performance, which has potential applications in the
316 field of electric heating clothing and thermal therapy.



317

318 Fig. 7 (a) Temperature variation of MXene-AgNWs/CNT films with time at different operating
 319 voltages; (b) Experimental data and linear fitting of saturation temperature versus U^2 and infrared
 320 images at different voltages.

321 4. Conclusion

322 In summary, we have developed a multifunctional sandwich bionic CNF/Mxene-AgNWs
 323 porous film with excellent EMI shielding and Joule heating performance. The sandwich bionic
 324 porous films were constructed through vacuum-assisted filtration and freeze-drying method using
 325 cellulose nanofibers as the substrate, MXene-AgNWs as the porous internal, revealing high EMI
 326 SE (67.5 dB), excellent SSE/t ($10675 \text{ dB cm}^2\text{g}^{-1}$), enhanced absorption coefficient up to 0.4 at a
 327 thickness of only 0.4 mm and excellent mechanical properties with a tensile strength of 20.3 MPa
 328 and a fracture strain of 2.1%. In addition, the prepared sandwich bionic MXene-AgNWs/CNF
 329 porous films exhibited excellent electrical heating performance and fast temperature response,
 330 which ensured the normal function of the prepared composite films under cold condition and
 331 facilitated the adaptation to complex application environments. Overall, the sandwich bionic
 332 porous films with excellent EMI shielding properties, high mechanical performance, and
 333 outstanding electrical heating performance, are potentially applied in the field of high-performance

334 multifunctional EMI shielding materials and thermal management.

335 **Data availability statement**

336 The raw/processed data required to reproduce these findings cannot be shared at this time as
337 the data also forms part of the ongoing study.

338 **Ethics declarations**

339 **Conflict of interest**

340 The authors declare that they have no competing interests as defined by Springer, or other
341 interests that might be perceived to influence the results and/or discussion reported in this paper.

342 **Consent for publication**

343 The manuscript is approved by all authors for publication.

344 **Ethical approval and consent to participate**

345 According to the guide for authors, I would like to declare on behalf of my co-author that this
346 work described was an original comment that has not been published previously. All the authors
347 listed have approved the manuscript that is enclosed.

348 **CRedit author statement**

349 **Fang Ren:** Writing-original draft, Data curation, Software. **Jiale Zhang:** Methodology,
350 Formal analysis, Resources. **Tong Wu:** Validation, Software. **Fudong Zhang:** Data curation,
351 Formal analysis. **Zhengzheng Guo:** Software. **Yanling Jin:** Supervision, Resources. **Penggang**
352 **Ren:** Conceptualization, Resources.

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358

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