ACS OMEGA

http://pubs.acs.org/journal/acsodf

Mini-Review

Recent Advances in MXene/Polyaniline-Based Composites for Electrochemical Devices and Electromagnetic Interference Shielding Applications

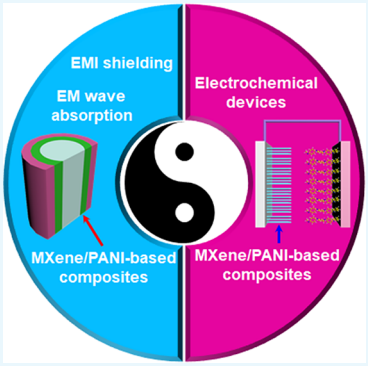
Zhiwei He,* Hangming Xie, Hanqing Wu, Jiahao Chen, Shiyu Ma, Xing Duan, Aqing Chen, and Zhe Kong

Cite This: *ACS Omega* 2021, 6, 22468−22477

Read Online

ACCESS | Metrics & More | Article Recommendations

ABSTRACT: Due to serious global warming and environmental issues, the demand for clean and sustainable energy storage devices is significantly increased. Often accompanied by rapid growth of portable electronic vehicles and devices, massive electromagnetic wave pollution becomes unavoidable. To mitigate the above two issues, this mini-review summaries preparation methods and recent developments of MXene/polyaniline-based composites for their applications in electrochemical devices and electromagnetic interference shielding. Based on excellent synergistic effects between single compounds and designed hierarchical structures, MXene/polyaniline-based composites usually exhibit enhanced physical and chemical properties, showing great potentials in sustainable electrochemical properties and electromagnetic wave protections for human health as well as normal operation of precise electronic devices.



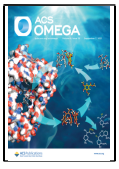
INTRODUCTION

Along with environmental pollution and the energy crisis, the demand for clean and sustainable energy is significantly increased.[1] Energy storage devices are environmentally friendly, cost-effective, and powerful, which promote the development of electronic vehicles and portable electronics.[2] Often accompanied by rapid growth of portable electronic vehicles and devices, massive electromagnetic wave pollution becomes unavoidable and thus has detrimental impacts on public health and the surrounding environment.[3] To mitigate electromagnetic (EM) wave pollution, electromagnetic interference (EMI) shielding and EM wave absorption materials are usually used such that human health can be protected as well as normal operation of precise electronic devices.[4]

MXenes, an emerging class of 2D transition metal carbides, nitrides, and carbonitrides as first published by Gogosti et al. in 2011,[5] are usually prepared by selective etching of the A layers (Group III A or IV A) from MAX phases with a general formula of $M_{n+1}X_nT_x$, where M is an early transition metal, X is C and/or N, T_x represents surface functional groups (e.g., −OH, −F, and/or −O), and n = 1, 2, or 3 (see Figure 1a).[5,6] Thus, MXenes possess unique properties,[7] such as high specific surface area, high electrical conductivity, surface hydrophilicity, and high electrochemical activity (see Table 1). It shows great potential in the applications of electrochemical devices, EMI shielding, and EM wave absorption. Due to the rich surface functional groups (such as −O, −OH, and/or −F), MXene can provide nucleation sites to obtain composites by the combination with other nanostructured polymers.[6c,8] Moreover, polyaniline (PANI), a typical pseudocapacitive conducting polymer, has been widely investigated because of its ease of synthesis, excellent thermal and environmental stability, low cost, and attractive redox and simple acid/base doping/dedoping (see Figure 1b and Table 1).[9] Nanostructured PANI can be easily introduced among layered structures of MXene to form hierarchical MXene/PANI-based composites when MXene acts as an active material with a large surface area and a framework (see Figure 1c).[7b]

Recently, MXene/PANI-based composites have aroused considerable interest for applications in supercapacitors,[6a,b,7d,11b] sensors,[7c,10c,d,14] sodium ion batteries,[12a] absorbers,[15] EMI shielding,[4] and EM wave absorption.[3a] To satisfy the demand for sustainable electrochemical devices and mitigate EM wave pollutions, this mini-review discusses preparation methods and applications of MXene/PANI-based composites in electrochemical performance, EMI shielding, and EM wave absorption. What's more, the analysis of electrochemical performance and EM performance of MXene/PANI-based

Received: June 8, 2021
Accepted: August 12, 2021
Published: August 25, 2021



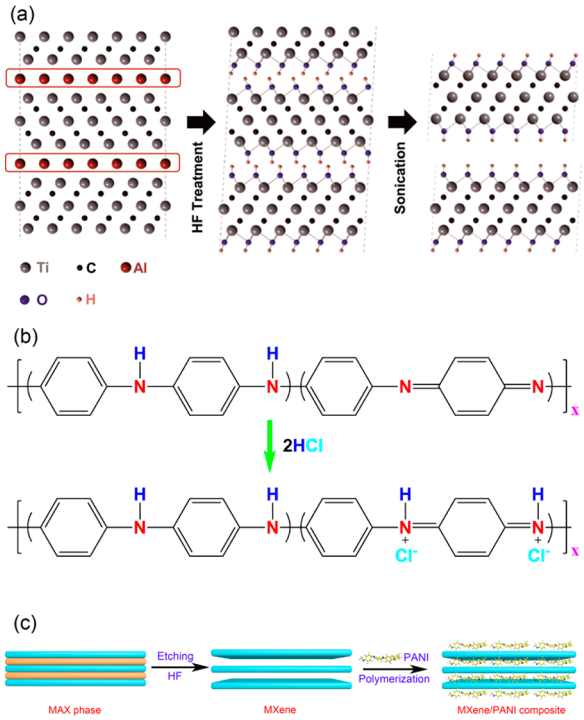


Figure 1. (a) Schematic of the exfoliation process for Ti_3AlC_2. (b) Doped PANI. (c) Schematic overview of preparation of MXene/PANI-based composites. Panel (a) is adapted with permission from ref 5. Copyright 2011 John Wiley & Sons, Inc.

composites is based on rational designed hierarchical structures and excellent synergistic effects between single compounds of the composites. Hence, this mini-review provides new insights into the preparation of MXene/PANI-based composites as well as the practical applications.

PREPARATION STRATEGIES OF MXENE/PANI-BASED COMPOSITES

MXene/PANI-based composites can be obtained by many approaches, such as in situ polymerization, interfacial polymerization, electropolymerization, self-assembly, layer-by-layer assembly, vacuum-assisted filtration, mechanical blending, dip-coating, spray coating, and hydrothermal reaction and so on (see Table 2). In this section, the above preparation strategies of MXene/PANI-based composites will be discussed.

In situ polymerization is one widely used method to prepare MXene/polyaniline-based composites. A general procedure is usually as follows (see Figure 2):[6c,10c,15,16] the delaminated $Ti_3C_2T_x$ powders are dispersed in a certain HCl solution, followed by addition of aniline monomers to obtain a mixed solution as stirred in an ice−water bath (0−5 °C). Then, ammonium persulfate (APS)-dispersed HCl solution is gradually added into the above mixed solution. The resulting dispersed solution is magnetically stirred for several hours at 0−5 °C. Finally, the MXene/PANI-based composite is obtained with the treatments of centrifuging, washing and vacuum-drying. In a polymerization process, positively charged aniline and negatively charged functional groups (e.g., Ti−OH^- and Ti−F^-) on the surface of MXene would attract each other.[16a] The electrostatic interactions promote nanostructured PANI anchored on the surface of MXene, and then the formed PANI nanostructures prevent MXene layers from stacking and collapsing. Moreover, VahidMohammadi et al. demonstrated

Table 1. Advantages and Disadvantages of MXene and PANI

materials	advantages	disadvantages
MXene	high specific surface area,[2a,6c,7a−c] high electrical conductivity,[2b,6c,7a−c,10] high electrochemical activity,[2a,7a,b] rich surface chemistry and hydrophilicity,[2b,7a,c,d,10b−d] mechanical rigidity,[7b,d,10b,c] special layered structure,[2a,7c] good thermal property,[10c] high pseudocapacity,[7c,10b,d] fascinating optical property,[10c] biocompatibility[7c]	low current density,[7a] restack and agglomerate during processing,[6c,11] narrow operating voltage windows,[7d,10b] poor specific capacitance,[7d] low theory capacitance[2a]
PANI	ease of synthesis[7b,10a] good electrical conductivity,[7b,10a] low cost,[2a,7b,12] low environmental impact,[2a,12] chemical stability,[10a] easy doping/dedoping,[2a,7a] high theoretical specific capacitance,[2a,7a,12b,13] nitrogen-containing functional groups,[13] good solubility[12b]	expansion/contraction of conjugated polymer chains,[7a] poor cycle stability,[6a,7a] self-polymerization[7d]

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477

Table 2. Preparation Methods of MXene/PANI-Based Composites

preparation methods	composites	features	ref
in situ polymerization	$Ti_3C_2T_x$/PANI	PANI was well-dispersed onto MXene surfaces	6c,10c,15,16
	Ti_3C_2/Fe_3O_4/PANI		7a,22
	Ti_3C_2/PANI-NTs		1
	$Ti_3C_2T_x$/TiO_2/PANI		7b,23
	graphene-$Ti_3C_2T_x$/PANI		7d
	PANI/Ti_3C_2		13
	graphene-Ti_2CT_x@PANI		2b
oxidant-free in situ polymerization	$Ti_3C_2T_x$/PANI	no oxidants required	11b
interfacial polymerization	$Ti_3C_2T_x$/CNZFO/PANI	two immiscible solvents	3b
electropolymerization	Pt/PANI/MXene	well-controlled	7c
	N-Ti_3C_2/PANI		2a
self-assembly	MXene-DLTA/PANI	electrostatic interactions induced, ordered structures	7a
	$Ti_3C_2T_x$/PANINFs		18
in situ assembly	PANI/$Ti_3C_2T_x$	easy to form on charged substrate	10b,14
	PNF/$Ti_3C_2T_x$		11a
layer-by-layer assembly	PANI/$Ti_3C_2T_x$/CF	controllable thickness, time-consuming and poor stability	24
	$Ti_3C_2T_x$/PANIF		10d
	PANI/$Ti_3C_2T_x$		10a
	$Ti_3C_2T_x$/Fe_3O_4@PANI		19a
vacuum-assisted filtration	$Ti_3C_2T_x$/c-PANI	easy to obtain free-standing films	4
	$Ti_3C_2T_x$-CNT/PANI		19b
	PANI@M-$Ti_3C_2T_x$		19c
mechanical blending	PANI/$Ti_3C_2T_x$	no solvents and additives required, but the product quality is not good	3a
dip-coating	$Ti_3C_2T_x$@PANI/mPP	easy to operate and control MXene content	20
spray coating	$Ti_3C_2T_x$-PAT-PANI-PpAP	cost-effective	21
hydrothermal reaction	$Ti_3C_2T_x$/PANI	no oxidants required but easy to oxidize MXene	6b

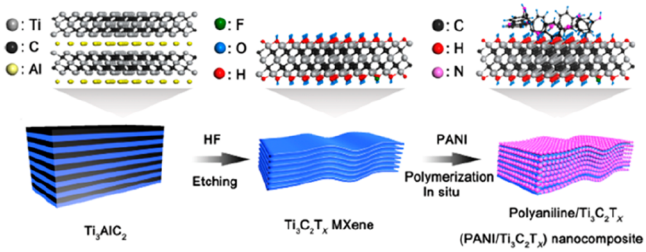


Figure 2. General procedure of preparing MXene/PANI-based composites. Adapted with permission from ref 10c. Copyright 2019 John Wiley & Sons, Inc.

that the MXene/PANI hybrid can be fabricated through an oxidant-free in situ polymerization process.[11b] Usually, ammonium persulfate is needed to initiate a polymerization process of conducting polymers, whereas aniline monomers can polymerize without an oxidant on the surface of MXene as long as a charge transfer occurs between aniline and MXene.[17] Using in situ polymerization, PANI can be well-dispersed onto the surface of MXene. Nanostructured PANI can easily anchor on the surface of MXene, and thus a large number of the resulting PANI can prevent the MXene nanosheets from stacking together.[10c]

Layer-by-layer assembly method is another facile method to synthesize MXene/PANI-based composite using electrostatic interactions between PANI and the surface of MXene.[11a] The hydrophilicity and negative charges on the surface of MXene provide the polymerization conditions for the combination with positively charged PANI.[10d] For example, Yin et al. developed a multifunctional MXene/PANI composite textile by a combination of 1D PANI nanowires and 2D MXene nanosheets onto a carbon fiber fabric substrate based on the layer-by-layer assembly method (see Figure 3).[10a] Moreover, self-assembly and in situ assembly methods are also used in the fabrication of

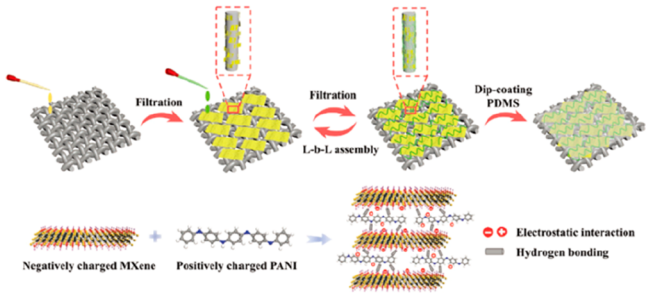


Figure 3. Schematic illustrating of the as-prepared conductive, flexible, and multifunctional PDMS/PMCFF-n fabric by vacuum-filter-assisted layer-by-layer assembly. Adapted with permission from ref 10a. Copyright 2021 Wiley-VCH GmbH.

MXene/PANI-based composites.[7a,10b,14,18] Wang et al. took solvent-assisted self-assembly and a redispersion strategy to prepare PANI/MXene inks and then obtained PANI/MXene composite films via an evaporation-induced assembly process.[10b] Chen et al. used DL-tartaric acid assembled on the surface of MXene to achieve supramolecular self-assembly and then obtained a PANI/MXene composite using the electronegative oxygen groups that can induce the polymerization of PANI.[7a]

Vacuum-assisted filtration is usually used to obtain free-standing MXene/PANI composite films.[4,19] In a typical vacuum-assisted filtration process, the PANI dispersion and the colloidal solution of MXene are mixed and stirred evenly, and then the MXene/PANI composite is dried and obtained by using vacuum-assisted filtration.[19b] For example, Zhang et al. dissolved a certain amount of c-PANI, $Ti_3C_2T_x$, and CNF-P in DMF,

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477

sonicated the above solution for 30 min, and obtained MXene/PANI composite films by vacuum-assisted filtration.[4] Wang et al. added different amounts of Fe_3O_4@PANI powder to $Ti_3C_2T_x$ solution, achieved a well-mixed solution after continuous stirring, and obtained a composite film by vacuum-assisted filtration (see Figure 4).[19a] In short, high-quality free-standing

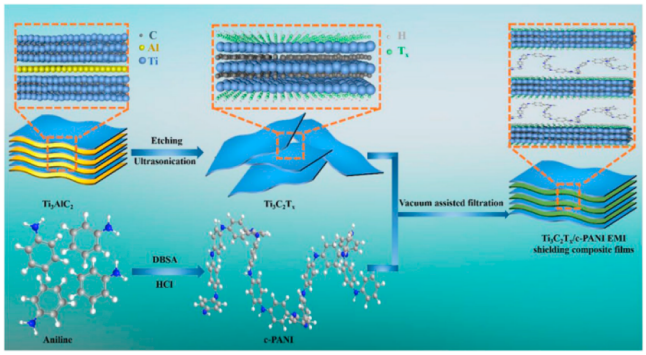


Figure 4. Fabrication diagram of the $Ti_3C_2T_x$/c-PANI EMI shielding composite films. Adapted with permission from ref 4. Copyright 2019 Elsevier Ltd.

MXene/PANI composite films can be prepared by vacuum-assisted filtration, but the whole process usually needs a long time (e.g., overnight).

In addition, other preparation strategies of MXene/PANI-based composites are also developed. For example, Lei et al. synthesized PANI onto $Ti_3C_2T_x$/CNZFO powder via a interfacial polymerization.[3b] Neampet et al. produced nanostructured PANI on the MXene surface by electropolymerization.[7c] Kumar et al. prepared the MXene/PANI composite by mechanical blending of PANI powder with MXene powder in a 1:1 weight ratio.[3a] Jia et al. fabricated MXene@PANI/mPP foam beads via a dip-coating approach and obtained foam beads with different MXene content by repeating the dip-coating process several times.[20] Raagulan et al. fabricated the $Ti_3C_2T_x$-PAT-PANI-PpAP composite by a cost-effective spray-coating technique.[21] Li et al. added aniline solution into the Ti_3AlC_2 solution under ultrasonic treatment for 6 h and obtained a composite via a hydrothermal reaction at different temperatures.[6b]

ELECTROCHEMICAL PROPERTIES OF SUPERCAPACITORS

Supercapacitors (SCs) have drawn substantial interest in portable electronic devices and electric vehicles because of high power density, long cycle life, fast charging and discharging rates, safe operation, and low maintenance costs.[1,2,6a,c,7a] The electrochemical properties of electrode materials in SCs can be evaluated by a potential window, specific capacity, energy density, power density, and cyclic stability and so on (see Table 3). In this section, the electrochemical properties of MXene, PANI, and MXene/PANI-based composites used as electrode materials of pseudocapacitive SCs are discussed in terms of a three-electrode system, a symmetric device, and an asymmetric device (Figure 5), respectively.

The three-electrode system is a conventional approach to evaluate the electrochemical performance of electrode materials

Table 3. Electrochemical Properties of MXene, PANI, and MXene/PANI-Based Composites Used as Electrode Materials for Supercapacitors

materials	electrolytes	potential window	specific capacity	energy density	power density	cyclic stability	ref
$Ti_3C_2T_x$//PANI@rGO	3 M H_2SO_4	1.45 V	57 F/g (at 5 mV/s)	~17 Wh/kg	~0.207 kW/kg	88% after 20000 cycles (at 100 mV/s)	6a
$Ti_3C_2T_x$//PANI@$Ti_3C_2T_x$	3 M H_2SO_4	1.2 V	87.3 F/g (at 10 mV/s)	50.6 Wh/L	1.7 kW/L		19c
graphene/Ti_2CT_x@PANI//graphene	1 M H_2SO_4	1.8 V	94.5 F/g (at 1 A/g)	42.3 Wh/kg	0.95 kW/kg	94.25% after 10000 cycles (at 10 A/g)	2b
$Ti_3C_2T_x$//rGO/CNT/PANI	3 M H_2SO_4	1.45 V	116.9 F/g (at 10 mV/s)	70 Wh/L	111 kW/L	80% after 10000 cycles (at 100 mV/s)	12b
PANI/$Ti_3C_2T_x$//$Ti_3C_2T_x$	1 M H_2SO_4	1.4 V	82.6 F/g (at 10 mV/s)	65.6 Wh/L	1.687 kW/L	87.5% after 5000 cycles (at 20 mA/cm^2)	10b
graphene/$Ti_3C_2T_x$/PANI//$Ti_3C_2T_x$	1 M H_2SO_4	1.5 V		15.6 Wh/kg	0.711 kW/kg	72.8% after 5000 cycles (at 10 A/g)	7d
$Ti_3C_2T_x$/PANI//active carbon	7 M KOH	1.2 V	563 F/g (at 0.5 A/g)	22.67 Wh/kg	0.217 kW/kg	90.82% after 10000 cycles (at 5 A/g)	6b
Ti_3C_2/PANI-NTs//Ti_3C_2/PANI-NTs	1 M H_2SO_4	1.8 V	300.8 F/g (at 0.1 A/g)	13.2 Wh/kg	1.61 kW/kg	81.8% after 4000 cycles (at 1 A/g)	1
$Ti_3C_2T_x$/PANI//$Ti_3C_2T_x$/PANI	3 M H_2SO_4	1 V	130 F/g (at 2 mV/s)	79.8 Wh/L	0.575 kW/L		11b
$Ti_3C_2T_x$/DLTA/PANI//$Ti_3C_2T_x$/DLTA/PANI	PVA-H_2SO_4 gel	0.8 V	710 mF/cm^2 (at 1 mA/cm^2)	0.063 mWh/cm^2	0.398 W/cm^2	61.5% after 10000 cycles (at 5 mA/cm^2)	7a
N-Ti_3C_2/PANI-420	0.5 M H_2SO_4	0.5 V (vs Hg/Hg_2Cl_2)	228 mF/cm^2 (at 5 mV/s)			85% after 1000 cycles (at 1 mA/cm^2)	2a
$Ti_3C_2T_x$/PANI	1 M H_2SO_4	0.8 V (vs Ag/AgCl)	556.2 F/g (at 0.5 A/g)			91.6% after 5000 cycles (at 5 A/g)	6c
PANI-Ti_3C_2	1 M Na_2SO_4	0.6 V (vs Ag/AgCl)	164 F/g (at 2 mV/s)			96% after 3000 cycles (at 3 A/g)	13
PANI@TiO_2/$Ti_3C_2T_x$	1 M KOH	0.7 V (vs Ag/AgCl)	188.3 F/g (at 10 mV/s)			94% after 8000 cycles (at 1 A/g)	7b
$Ti_3C_2T_x$/PANINFs	3 M H_2SO_4	0.95 V (vs Ag/AgCl)	645.7 F/g (at 10 mV/s)			98% after 5000 cycles (at 10 mV/s)	18
$Ti_3C_2T_x$-CNT/PANI	1 M H_2SO_4	1 V (vs Hg/Hg_2SO_4)	429.4 F/g (at 1 A/g)			10000 cycles (at 20 A/g)	19b

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477

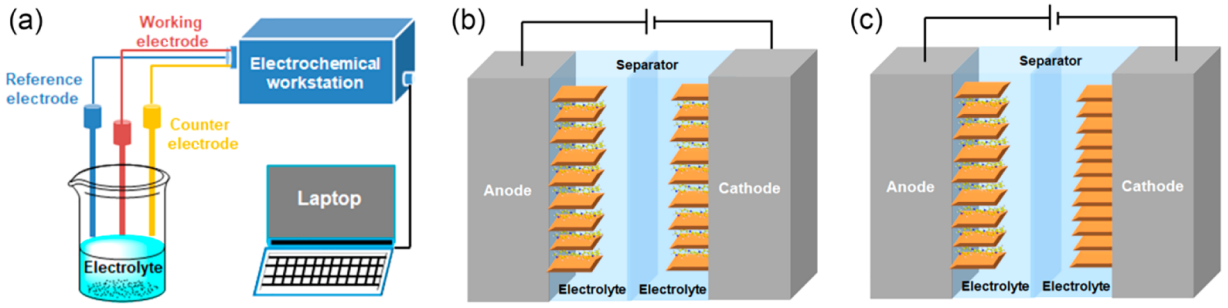


Figure 5. Schematic overview of evaluating electrochemical properties of SCs via (a) three-electrode system, (b) symmetric device, and (c) asymmetric device.

of SCs, consisting of a working electrode, a reference electrode, and a counter electrode. Usually, a MXene/PANI-based composite is used in the working electrode, Ag/AgCl (or Hg/Hg_2Cl_2, Hg/Hg_2SO_4) is used as the reference electrode, and platinum electrode (or glass carbon) is used in the counter electrode. Compared with the two-electrode system, the three-electrode system can maintain the steady potential of the working electrode and thus facilitate the evaluation of the electrochemical properties of the working electrode. For example, PANI-Ti_3C_2 composites were synthesized via in situ polymerization and can be used as a high-performance supercapacitor electrode material as tested by a three-electrode system.[13] The results showed that the introduction of amino groups in PANI to Ti_3C_2 improved not only electric conductivity but also surface wettability and thus improved the electrochemical properties because of the synergistic effect between PANI and Ti_3C_2.[13] To increase amino groups, Wu et al. used the amino group of amino-Ti_3C_2 as the active site to react with the amine nitrogen of PANI chains.[2a] As a result, the electrochemical activity sites and charge transport can be further increased because of the synergistic effect between N-Ti_3C_2 and PANI, and thus the electrochemical properties of N-Ti_3C_2/PANI for SCs improved.[2a]

For PANI-Ti_3C_2 composites, hierarchical structures can also influence electrochemical properties. Wu et al. introduced PANI nanotubes onto the interlamination of ultrathin graphene-like Ti_3C_2 nanosheets and found that the dispersed PANI nanotubes showed an open and interlocking structure preventing Ti_3C_2 nanosheets from stacking and collapsing.[1] Another type of MXene $Ti_3C_2T_x$ is used to combine with PANI to fabricate $Ti_3C_2T_x$/PANI composites.[6c,7b,11b] The experimental results indicate that hierarchical structures and the synergistic effect between PANI and $Ti_3C_2T_x$ are attributed to the high performance of $Ti_3C_2T_x$/PANI composites used as electrode materials of SCs as tested in a three-electrode system.[7b]

To evaluate the feasibility of electrode materials used in practical SCs,[19c] electrode materials of SCs are usually assembled into symmetric and asymmetric devices. Compared with asymmetric devices, symmetric devices usually show relatively narrow operating voltage window.[10b] In a symmetric device, anode and cathode materials are the same and require chemical stability as used in electrolytes. The conducting polymers usually show the best electrochemical performance in acidic electrolytes under positive potentials.[9a] Therefore, the target materials used in both anode and cathode need to be stable under negative or positive potentials. For example, Wu et al. fabricated Ti_3C_2/PANI-NTs-1 composites by decorating 2D Ti_3C_2 nanosheets with 1D hollow PANI-NTs, and these unique structures as combined from Ti_3C_2 and PANI-NTs-1 enlarge ion diffusion kinetics during the redox reaction.[1] When the composite is used as advanced electrode materials for symmetric supercapacitors (SSCs), it can light up a 1.8 V LED, indicating its great potential in practical energy storage.[1]

For asymmetric supercapacitors (ASCs), matching cathode materials with anode materials is still a challenge to improve the integral electrochemical performance of SCs, such as operating voltage window and energy density. Take MXene/PANI-based composites as cathode materials, for instance; Li et al. prepared a hierarchical PANI@MXene cathode with a PANI layer uniformly coated on a 3D porous MXene $Ti_3C_2T_x$ network.[19c] Compared with pure MXene, the integrated PANI@MXene heterostructure can enhance the electrochemical stability toward anodic oxidation, as demonstrated by first-principles calculations.[16b,19c] Similarly, Wang et al. designed a scalable PANI/MXene cathode material decorated with PANI nanodots.[10b] In this cathode material, the PANI nanoparticles not only work as high pseudocapacitive materials but also act as pillaring components for reducing MXene stacking and enabling electron and ion transport, thereby obtaining an excellent synergistic effect in the electrochemical performance of ASCs.[10b]

In order to enhance electrochemical properties, many researchers introduced a third component into MXene/PANI-based composites to form ternary composites as used in ASCs. For example, Fu et al. reported that a graphene-encapsulated MXene Ti_2CT_x@PANI composite used as a cathode material can exhibit improved cycling stability and better electrochemical performance of ASCs owing to the robust hierarchical nanostructures and complementary synergistic effect between graphene-encapsulated Ti_2CT_x and PANI.[2b] Wang et al. successfully prepared a layered graphene-decorated 2D $Ti_3C_2T_x$/polyaniline (GTP) nanocomposite, and GTP showed a superior capacitive performance, large voltage window, and good cycling stability based on the synergistic effect among three components when served as the cathode materials in ASCs.[7d] In short, when used as cathode materials, MXene/PANI-based composites with hierarchical structures usually exhibit synergistic effects between MXene and PANI and thus improve the electrochemical performance of ASCs.

ELECTROCHEMICAL PROPERTIES OF OTHER DEVICES

Recently, MXene/PANI-based composites have also been used as electrode materials for other electrochemical energy storage devices[11a,12a] and sensors.[7c,10c,14] For electrochemical energy storage, the introduction of conducting PANI into MXene/PANI-based composites can prevent MXene nanosheets from restacking and agglomerating and thus improve electrochemical

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477

properties because of potential synergistic effects. For example, first-principles calculations were utilized to systematically investigate electrochemical properties, and the results showed the introduction of PANI into MXene/PANI composites can improve electric conductivity and maintain strong Na adsorption and high Na diffusion kinetics when used as anode materials for sodium ion batteries.[12a] Yun et al. prepared a 2 μm thick thin film PNF/MXene electrode via a layer-by-layer assembly process and applied it for thin-film electrochemical energy storage.[11a]

For sensors, MXene/PANI-based composites can be used as sensitive films to detect low content substances, such as NH_3,[14] lactate,[7c] or ethanol gas.[10c] Generally, hierarchical MXene/PANI-based composites have abundant absorption sites and large specific surface area that facilitate a fast response and high sensitivity toward low content substances. For example, Li et al. prepared a flexible PANI/$Ti_3C_2T_x$ hybrid-sensitive film-based gas sensor that exhibited excellent NH_3-sensing properties to 20−80% relative humidity environments at a temperature range of 10−40 °C.[14] Neampet et al. used a Pt/PANI/MXene nanocomposite for amperometric sensing of hydrogen peroxide and lactate, and this nanocomposite can provide a low detection limit of 1.0 μM.[7c] Zhao et al. demonstrated high electrocatalytic sensitivity of PANI/$Ti_3C_2T_x$ nanocomposites for ethanol gas via an integration of density functional theory simulations and bulk electrosensitive measurements.[10c]

ELECTROMAGNETIC INTERFERENCE SHIELDING PROPERTIES

In the development of electronic devices, EM waves are unavoidable during the normal performance in daily life.[25] EMI and EM wave pollutions become critical issues for public health and malfunctioning of electronic devices and thus restrict the wide use of portable electronics.[16a,19a] EMI shielding materials and EM wave absorption materials are usually chosen to mitigate the influence of EM waves. To date, various types of materials have been utilized in preventing EM wave pollution.[26] For metals, copper, aluminum, silver, nickel, and stainless steel have been used as a shield, while these metals have certain limitations, such as high density, difficult machining, and high corrosion susceptibility.[26c,d] Many heterogeneous composites with conducting fillers (e.g., carbon nanotubes (CNTs), graphene, reduced graphene oxide (rGO), and MoS_2),[27] magnetic fillers (e.g., Fe_3O_4, Fe_2O_3, and barium ferrite),[19a,26c] and dielectric fillers (e.g., $BaTiO_3$, TiO_2, and $PbTiO_3$)[23,26c,28] have been applied in the EMI shielding application instead of metals because of their advantages (e.g., low weight, better environmental stability, and excellent corrosion-resistant properties).[26c−f]

Generally, for EMI shielding, there are three kinds of mechanisms for EMI shielding: (1) to reflect radiation; (2) to absorb EM radiation; (3) have multiple internal reflections (see Figure 6).[25] Usually, EM reflection is due to mobile charge carriers in EMI shielding materials, whereas EM absorption is because of electric and/or magnetic dipoles.[3a] Thus, absorption, reflection, and multiple reflections are three key factors for the EMI shielding behavior of materials.[21] High electrical conductivity usually determines the reflection and absorption performance of EMI shielding materials. Moreover, the structural design of EMI materials is also important to minimize the secondary EMI pollution.[20]

Since 2011, Gogotsi et al. have reported a novel 2D material named MXene[5] and found that 2D $Ti_3C_2T_x$ MXene can be used

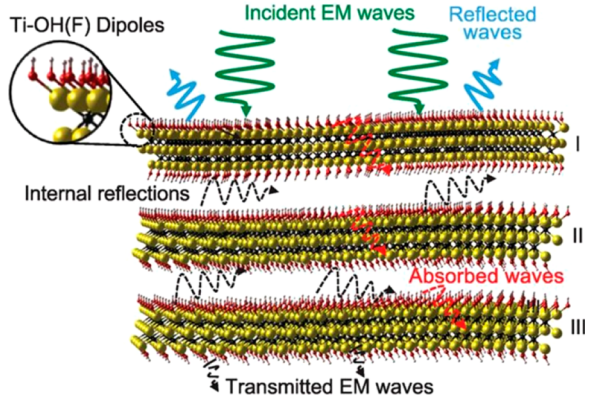


Figure 6. EMI shielding mechanism of MXene $Ti_3C_2T_x$. Adapted with permission from ref 25. Copyright 2016 the American Association for the Advancement of Science (AAAS).

in the EM field because of its unique layered structures, providing many conductive paths.[25] Although the $Ti_3C_2T_x$ film (a 45 μm thickness) can exhibit EMI SE of 92 dB among synthetic materials with comparable thickness,[25] the high dielectric constant of $Ti_3C_2T_x$ will cause impedance mismatch, limiting its further improvement in the electromagnetic performance.[16b] PANI is also used as EMI shielding material because of its low cost, easy doping/dedoping, low environmental impact, and easy synthesis. Synergies between $Ti_3C_2T_x$ and PANI, such as interface polarization and dipole-oriented polarization, multiple conductive paths, improved impedance matching, and increased dielectric loss are conductive to attenuation of EM waves.[3b,16a,22,23] Therefore, many researchers show great interest in the MXene/PANI-based nanocomposites as well as their application in EMI shielding (see Table 4).[3a,4,10a,19a,20,21,24,25] Generally, EMI shielding effectiveness (SE) describes the ability of materials to shield EM waves, when the reflection and absorption coefficient are quantitative descriptions of EM waves reflection and absorption.[4] For example, binary MXene/PANI-based composites have been used in the application of EMI shielding. Kumar et al. reported that a lightweight Ti_3C_2/PANI composite had a good enhanced EMI SE of ≈23 dB with the sample thickness of 1.5 mm and found that the laminated MXene with a superior specific surface area plays a dominate role in the enhanced microwave absorbing performance of Ti_3C_2/PANI composite.[3a] Based on optimized organic/inorganic (1D/2D) continuous conductive network, Yin et al. developed a flexible, lightweight, and multifunctional PANI/MXene-based composite textile possessing an outstanding electrical conductivity (325 S/m) and an excellent EMI shielding performance (SE of ≈35.3 dB) at the thickness of only 0.376 mm.[10a] Zhang et al. fabricated ultrathin and flexible $Ti_3C_2T_x$/c-PANI composite films with EMI SE of 36 dB at the thickness of 40 μm and showed that the shielding mechanism changed from the equal shielding mechanism to the reflection-dominated shielding mechanism as the content of $Ti_3C_2T_x$ in $Ti_3C_2T_x$/c-PANI composite increased.[4]

To further improve the EMI shielding properties, several researchers show interest in the fabrication of ternary or quaternary MXene/PANI-based composites. For example, Jia et al. constructed compressible MXene@PANI/mPP composite foams, and these composite foams showed an absorption-dominated shielding mechanism with effective EMI SE of ∼23.5−39.8 dB and low R coefficient of ∼0.20−0.31 after encapsulating with PDMS.[20] Raagulan et al. reported that a

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477

Table 4. EMI Shielding Properties of MXene/PANI-Based Composites

materials	electrical conductivity (S/m)	EMI SE (dB)	thickness (mm)	band frequency (GHz)	ref
MXene@PANI/mPP		~23.5−39.8	12	5.38−8.17	20
$Ti_3C_2T_x/Fe_3O_4$@PANI	59900	58.8	0.0167	8.2−12.4	19a
PANI/MXene/CF	24.57	26.0	0.55	8.2−12.4	24
MXene/PAT/PANI-PpAP	781.3	45.18	0.8	8.2	21
Ti_3C_2/PANI		22.7	1.5	10.8	3a
$Ti_3C_2T_x$/c-PANI	2440	36	0.04	8.2−12.4	4
PANI/MXene	325	35.3	0.376	8.2−12.4	10a

MXene-PAT-PANI-PpAP composite had an EMI SE of 45.18 dB at 8.2 GHz.[21] They found that reduced form of MXene increased EMI SE, and the absorption was enhanced by the ant-farm-like structures.[21] Wang et al. prepared a flexible and lightweight $Ti_3C_2T_x/Fe_3O_4$@PANI composite film by vacuum-assisted filtration, and this ternary composite achieved an EMI SE of 62 dB with the film thickness of 16.7 μm, which exhibited great potential for applications in packaging, wearable electronic equipment, and military fields.[19a] Yin et al. prepared multilayer structured PANI/MXene/CF fabric via a layer-by-layer strategy, and this fabric realized a high EMI SE (26.0 dB) and specific EMI SE (135.5 dB cm^3/g).[24] Moreoever, this fabric maintained flexibility, air permeability and washability of textile substrates, showed an absorption-dominated shielding mechanism and thus effectively decreased secondary EM wave pollution.[24]

ELECTROMAGNETIC WAVE ABSORPTION PROPERTIES

To alleviate EM pollution, one alternative approach is to use EM wave absorption materials. Ideal EM wave absorption materials need to exhibit excellent properties, such as lightweight,[23] strong absorption peaks,[3b] broad effective absorption bandwidths,[3b] good interfacial polarization,[22] EM wave attenuation capability,[3b] proper impedance matching,[3b,22] and so on.

Recently, MXene has been used in EM wave absorption because of its excellent electrical conductivity, dielectric loss, high specific surface area, mechanical rigidity, and stable chemical properties (see Table 1).[3b,7a−c,23] Meanwhile, PANI as a conducting polymer is also a EM wave absorption material with electrical loss.[23] To enhance EM wave absorption properties, MXene/PANI-based composites have been studied by many researchers (see Table 5).[3b,16a,22,23] For example, Wei et al. fabricated a sandwich-like $Ti_3C_2T_x$/PANI composites by the in situ polyreaction of aniline monomers onto $Ti_3C_2T_x$, showing an effective absorption bandwidth (>90%) ranging from X-band (8−12.4 GHz) to Ku-band (12.4−18 GHz) because of synergistic effect between $Ti_3C_2T_x$ and PANI and exhibiting great potential in serving as microwave absorption materials.[16a] Wang et al. prepared a hierarchical Ti_3C_2/Fe_3O_4/PANI ternary composite by HF etching, coprecipitation, and in situ polymerization route, and this composite realized an efficient absorption bandwidth (<−10 dB) of 5.2 GHz (from 12.8 to 18 GHz) at only 1.9 mm.[22] This improved microwave absorption performance may be attributed to hierarchical structures and synergistic effect of combining Ti_3C_2, Fe_3O_4, and PANI.[22] Lei et al. synthesized a $Ti_3C_2T_x$/CNZFO/PANI composite via a facile three-step procedure in terms of exfoliation, hydrothermal process, and interfacial polymerization and investigated EM wave absorption mechanisms for the $Ti_3C_2T_x$/CNZFO/PANI composite, as shown in Figure 7.[3b] They found that the $Ti_3C_2T_x$/CNZFO/PANI composite showed a relatively better impedance matching and attenuation constant and thus exhibited a good microwave absorption.[3b] Gao et al. fabricated $Ti_3C_2T_x/TiO_2$/PANI multilayer composites by HCl and LiF etching, a one-step hydrothermal method, and in situ polymerization, exhibiting an effective absorption bandwidth (R_L < −10 dB) of 5.92 GHz (11.84 to 17.76 GHz) at 2.1 mm.[23] This $Ti_3C_2T_x/TiO_2$/PANI composite also possessed excellent EM wave absorption properties, probably because of excellent dielectric properties and synergistic effect.[23] In a word, unique hierarchical structures and synergistic effects of combining MXene, PANI, and other components play a vital role in the improvement of dielectric loss and optimization of impedance matching and thus facilitate the absorption of EM waves.

Table 5. Electromagnetic Wave Absorption Properties of MXene/PANI-Based Composites

materials	reflection loss (dB)	effective absorption bandwidth (GHz)	thickness (mm)	ref
$Ti_3C_2T_x$/CNZFO/PANI	−37.1 (at 10.2 GHz)	4.1 (8.2−12.3 GHz)	2.2	3b
$Ti_3C_2T_x$/PANI	−56.3 (at 13.8 GHz)	4.28 (11.99−16.27 GHz)	1.8	16a
$Ti_3C_2T_x/TiO_2$/PANI	−65.61 (at 13.92 GHz)	5.92 (11.84−17.76 GHz)	2.18	23
Ti_3C_2/Fe_3O_4/PANI	−40.3 (at 15.3 GHz)	5.2 (12.8−18 GHz)	1.9	22

CONCLUSIONS AND OUTLOOK

In summary, this mini-review introduces preparation methods and recent developments of MXene/polyaniline-based composites for their applications in electrochemical devices and EMI shielding. When used as electrode materials, MXene/polyaniline-based composites show excellent electrochemical properties in supercapacitors, lithium ion battery, sodium ion battery and sensors because of hierarchical structures and synergistic effects by the combination of MXene and PANI. Moreover, MXene/polyaniline-based composites can be also applied in the field of preventing EM wave pollution based on unique properties of MXene and PANI (e.g., good electrical conductivity and dielectric loss).

In the future, the widespread use of portable electronic vehicles and devices is a general trend, and it is necessary to design electrochemical energy storage devices with massive power density and life span. Thus, construction of electrode materials with stable hierarchical structures and synergistic effects during an electrochemical process is crucial for energy storage devices. Of course, EM wave pollution is also a huge challenge when abundant portable electronic vehicles and devices are used. By optimizing hierarchical structures and introducing more components, excellent composites can be achieved to mitigate EM wave pollutions. Moreover, MXene has

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477

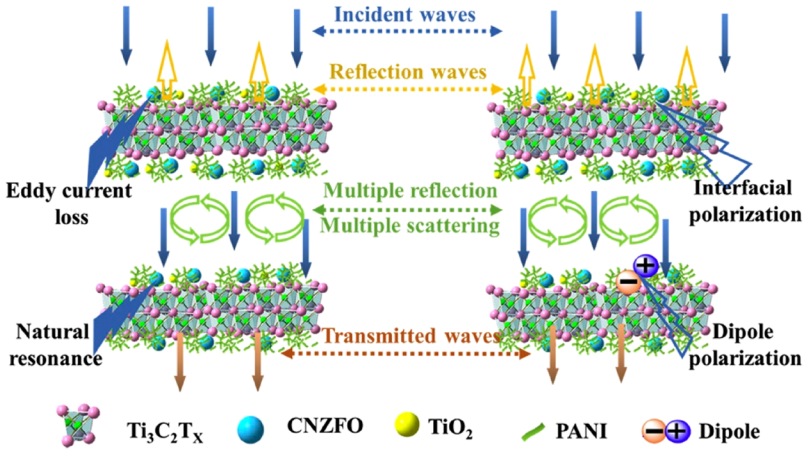


Figure 7. EM wave absorption mechanisms for $Ti_3C_2T_x$/CNZFO/PANI composite. Adapted with permission from ref 3b. Copyright 2020 Elsevier.

good environmental stability, and PANI is also an environmentally friendly polymer. Thus, a MXene/PANI-based composite has less environmental impact during its various applications. Based on controllable hierarchical structures and synergistic effects between MXene and PANI, a MXene/PANI-based composite shows great commercial potential in the future. All in all, rationally designed hierarchical MXene/polyaniline-based composites are promising candidates for electrochemical devices, EMI shielding, and EM wave absorption and also provide great potential in the practical applications.

AUTHOR INFORMATION

Corresponding Author

Zhiwei He − *Center for Advanced Optoelectronic Materials, Key Laboratory of Novel Materials for Sensor of Zhejiang Province, College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, China;* orcid.org/0000-0002-3251-8731; Email: zhiwei.he@hdu.edu.cn

Authors

Hangming Xie − *School of Electronics Information, Hangzhou Dianzi University, Hangzhou 310018, China*

Hanqing Wu − *School of Mechanical Engineering, Hangzhou Dianzi University, Hangzhou 310018, China*

Jiahao Chen − *Center for Advanced Optoelectronic Materials, Key Laboratory of Novel Materials for Sensor of Zhejiang Province, College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, China*

Shiyu Ma − *Center for Advanced Optoelectronic Materials, Key Laboratory of Novel Materials for Sensor of Zhejiang Province, College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, China*

Xing Duan − *Center for Advanced Optoelectronic Materials, Key Laboratory of Novel Materials for Sensor of Zhejiang Province, College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, China;* orcid.org/0000-0002-3968-7650

Aqing Chen − *Center for Advanced Optoelectronic Materials, Key Laboratory of Novel Materials for Sensor of Zhejiang Province, College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, China;* orcid.org/0000-0003-2442-5727

Zhe Kong − *Center for Advanced Optoelectronic Materials, Key Laboratory of Novel Materials for Sensor of Zhejiang Province, College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, China;* orcid.org/0000-0001-8622-0175

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.1c02996

Notes

The authors declare no competing financial interest.

Biographies

Zhiwei He received his Ph.D. degree in Nanomechanics from NTNU Nanomechanical Lab (Prof. Zhiliang Zhang) in 2016. He continued to work as a postdoctoral researcher at NTNU Nanomechanical Lab in 2017. He joined the College of Materials and Environmental Engineering at Hangzhou Dianzi University in 2018 and was appointed as an associate professor. His research interest focuses on superhydrophobic and icephobic surfaces and conducting polymer-based nanomaterials.

Hangming Xie received his M.S. degree in Electronic Science and Technology from Hunan University of Science and Technology in 2020. He continues to work as a postgraduate at Hangzhou Dianzi University (Prof. Zhiwei He). His research interest focuses on conductive polymers and supercapacitors.

Hanqing Wu received his M.S. degree in Mechanical Engineering from Shandong Jianzhu University in 2019. He continues to work as a postgraduate at Hangzhou Dianzi University (Prof. Zhiwei He). His research interest focuses on superhydrophobic and icephobic surfaces.

Jiahao Chen is an undergraduate at College of Materials and Environmental Engineering in Hangzhou Dianzi University. His research interest focuses on sonochemistry and current collectors.

Shiyu Ma received her Ph.D. degree in Materials Science and Engineering from Shandong University (Prof. Jianxin Zhang) in 2018. She joined the College of Materials and Environmental Engineering at Hangzhou Dianzi University in 2018. Her research interest focuses on first-principles calculations.

Xing Duan received his Ph.D. degree in Materials Science and Engineering from Zhejiang University (Prof. Guodong Qian) in 2015. He joined the College of Materials and Environmental Engineering at Hangzhou Dianzi University in 2015 and was appointed as an associate professor in 2019. His research interest focuses on metal−organic frameworks and gas adsorption separation.

Aqing Chen received his Ph.D. degree in Condensed Matter Physics from Beihang University in 2015. He joined the College of Materials

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477

and Environmental Engineering at Hangzhou Dianzi University in 2015 and was appointed as an associate professor. His research interest focuses on electrochemistry, coatings, nanomaterials, and thin films.

Zhe Kong received her Ph.D. (2011) from Zhejiang University. She is an associate professor in the College of Materials and Environmental Engineering at Hangzhou Dianzi University in China. Her research is focused on the properties and applications of two-dimensional nanomaterials.

■ ACKNOWLEDGMENTS

The National Natural Science Foundation of China (Grant No. 51803043) and the Science Foundation of Hangzhou Dianzi University (KYS205618119) are acknowledged for the financial support. Z.H. appreciates the kind help from Dr. Boota and Prof. Dr. Yury Gogotsi during collecting literature data, and thanks Prof. Dr. Qiu-Feng Lü for her useful comments on this mini-review.

■ REFERENCES

(1) Wu, W.; Wang, C.; Zhao, C.; Wei, D.; Zhu, J.; Xu, Y. Facile strategy of hollow polyaniline nanotubes supported on Ti_3C_2-MXene nanosheets for High-performance symmetric supercapacitors. *J. Colloid Interface Sci.* **2020**, *580*, 601−13.

(2) (a) Wu, W.; Niu, D.; Zhu, J.; Gao, Y.; Wei, D.; Liu, X.; Wang, F.; Wang, L.; Yang, L. Organ-like Ti_3C_2 Mxenes/polyaniline composites by chemical grafting as high-performance supercapacitors. *J. Electroanal. Chem.* **2019**, *847*, 113203. (b) Fu, J.; Yun, J.; Wu, S.; Li, L.; Yu, L.; Kim, K. H. Architecturally robust graphene-encapsulated MXene Ti_2CT_x@ polyaniline composite for high-performance pouch-type asymmetric supercapacitor. *ACS Appl. Mater. Interfaces* **2018**, *10* (40), 34212−21.

(3) (a) Kumar, S.; Arti; Kumar, P.; Singh, N.; Verma, V. Steady microwave absorption behavior of two-dimensional metal carbide MXene and Polyaniline composite in X-band. *J. Magn. Magn. Mater.* **2019**, *488*, 165364. (b) Lei, Y.; Yao, Z.; Li, S.; Zhou, J.; Haidry, A. A.; Liu, P. Broadband high-performance electromagnetic wave absorption of Co-doped NiZn ferrite/polyaniline on MXenes. *Ceram. Int.* **2020**, *46* (8), 10006−10015.

(4) Zhang, Y.; Wang, L.; Zhang, J.; Song, P.; Xiao, Z.; Liang, C.; Qiu, H.; Kong, J.; Gu, J. Fabrication and investigation on the ultra-thin and flexible $Ti_3C_2T_x$/co-doped polyaniline electromagnetic interference shielding composite films. *Compos. Sci. Technol.* **2019**, *183*, 107833.

(5) Naguib, M.; Kurtoglu, M.; Presser, V.; Lu, J.; Niu, J.; Heon, M.; Hultman, L.; Gogotsi, Y.; Barsoum, M. W. Two-dimensional nanocrystals produced by exfoliation of Ti_3AlC_2. *Adv. Mater.* **2011**, *23* (37), 4248−53.

(6) (a) Boota, M.; Gogotsi, Y. MXene—Conducting polymer asymmetric pseudocapacitors. *Adv. Energy Mater.* **2019**, *9* (7), 1802917. (b) Li, Y.; Kamdem, P.; Jin, X.-J. Hierarchical architecture of MXene/PANI hybrid electrode for advanced asymmetric supercapacitors. *J. Alloys Compd.* **2021**, *850*, 156608. (c) Xu, H.; Zheng, D.; Liu, F.; Li, W.; Lin, J. Synthesis of an MXene/polyaniline composite with excellent electrochemical properties. *J. Mater. Chem. A* **2020**, *8* (12), 5853−58. (d) Naguib, M.; Mochalin, V. N.; Barsoum, M. W.; Gogotsi, Y. 25th anniversary article: MXenes: A new family of two-dimensional materials. *Adv. Mater.* **2014**, *26* (7), 992−1005.

(7) (a) Chen, Z.; Wang, Y.; Han, J.; Wang, T.; Leng, Y.; Wang, Y.; Li, T.; Han, Y. Preparation of polyaniline onto $_{DL}$-tartaric acid assembled MXene surface as an electrode material for supercapacitors. *ACS Appl. Energy Mater.* **2020**, *3* (9), 9326−36. (b) Lu, X.; Zhu, J.; Wu, W.; Zhang, B. Hierarchical architecture of PANI@TiO_2/$Ti_3C_2T_x$ ternary composite electrode for enhanced electrochemical performance. *Electrochim. Acta* **2017**, *228*, 282−89. (c) Neampet, S.; Ruecha, N.; Qin, J.; Wonsawat, W.; Chailapakul, O.; Rodthongkum, N. A nanocomposite prepared from platinum particles, polyaniline and a Ti_3C_2 MXene for amperometric sensing of hydrogen peroxide and lactate. *Microchim. Acta* **2019**, *186* (12), 752. (d) Wang, S.; Ma, Z.; Lü, Q.-F.; Yang, H. Two-dimensional $Ti_3C_2T_x$/polyaniline nanocomposite from the decoration of small-sized graphene nanosheets: Promoted pseudocapacitive electrode performance for supercapacitors. *ChemElectroChem* **2019**, *6* (10), 2748−54. (e) Zhang, C. J.; Pinilla, S.; McEvoy, N.; Cullen, C. P.; Anasori, B.; Long, E.; Park, S.-H.; Seral-Ascaso, A.; Shmeliov, A.; Krishnan, D.; Morant, C.; Liu, X.; Duesberg, G. S.; Gogotsi, Y.; Nicolosi, V. Oxidation stability of colloidal two-dimensional titanium carbides (MXenes). *Chem. Mater.* **2017**, *29* (11), 4848−56.

(8) (a) Jimmy, J.; Kandasubramanian, B. Mxene functionalized polymer composites: Synthesis and applications. *Eur. Polym. J.* **2020**, *122*, 109367. (b) He, Z.-W.; Lü, Q.-F.; Lin, Q. Fabrication, characterization and application of nitrogen-containing carbon nanospheres obtained by pyrolysis of lignosulfonate/poly(2-ethylaniline). *Bioresour. Technol.* **2013**, *127*, 66−71.

(9) (a) Vonlanthen, D.; Lazarev, P.; See, K. A.; Wudl, F.; Heeger, A. J. A stable polyaniline-benzoquinone-hydroquinone supercapacitor. *Adv. Mater.* **2014**, *26*, 5095−100. (b) He, Z.-W.; Yang, J.; Lü, Q.-F.; Lin, Q. Effect of structure on the electrochemical performance of nitrogen- and oxygen-containing carbon micro/nanospheres prepared from lignin-based composites. *ACS Sustainable Chem. Eng.* **2013**, *1* (3), 334−40. (c) He, Z.-W.; Lü, Q.-F.; Lin, Q. Fabrication of poly(N-ethylaniline)/lignosulfonate composites and their carbon microspheres. *Int. J. Biol. Macromol.* **2012**, *51* (5), 946−52.

(10) (a) Yin, G.; Wang, Y.; Wang, W.; Qu, Z.; Yu, D. A flexible electromagnetic interference shielding fabric prepared by construction of PANI/MXene conductive network via layer-by-layer assembly. *Adv. Mater. Interfaces* **2021**, *8* (6), 2001893. (b) Wang, Y.; Wang, X.; Li, X.; Bai, Y.; Xiao, H.; Liu, Y.; Yuan, G. Scalable fabrication of polyaniline nanodots decorated MXene film electrodes enabled by viscous functional inks for high-energy-density asymmetric supercapacitors. *Chem. Eng. J.* **2021**, *405*, 126664. (c) Zhao, L.; Wang, K.; Wei, W.; Wang, L.; Han, W. High-performance flexible sensing devices based on polyaniline/MXene nanocomposites. *InfoMat* **2019**, *1* (3), 407−16. (d) Chao, M.; Wang, Y.; Ma, D.; Wu, X.; Zhang, W.; Zhang, L.; Wan, P. Wearable MXene nanocomposites-based strain sensor with tile-like stacked hierarchical microstructure for broad-range ultrasensitive sensing. *Nano Energy* **2020**, *78*, 105187.

(11) (a) Yun, J.; Echols, I.; Flouda, P.; Wang, S.; Easley, A.; Zhao, X.; Tan, Z.; Prehn, E.; Zi, G.; Radovic, M.; Green, M. J.; Lutkenhaus, J. L. Layer-by-layer assembly of polyaniline nanofibers and MXene thin-film electrodes for electrochemical energy storage. *ACS Appl. Mater. Interfaces* **2019**, *11* (51), 47929−38. (b) VahidMohammadi, A.; Moncada, J.; Chen, H.; Kayali, E.; Orangi, J.; Carrero, C. A.; Beidaghi, M. Thick and freestanding MXene/PANI pseudocapacitive electrodes with ultrahigh specific capacitance. *J. Mater. Chem. A* **2018**, *6* (44), 22123−33.

(12) (a) Zhou, W.; Kuang, W.; Liang, X.; Zhou, W.; Guo, J.; Gan, L.; Huang, D. Exploration of MXene/polyaniline composites as promising anode materials for sodium ion batteries. *J. Phys. D: Appl. Phys.* **2021**, *54* (6), 064001. (b) Li, K.; Wang, X.; Wang, X.; Liang, M.; Nicolosi, V.; Xu, Y.; Gogotsi, Y. All-pseudocapacitive asymmetric MXene-carbon-conducting polymer supercapacitors. *Nano Energy* **2020**, *75*, 104971.

(13) Ren, Y.; Zhu, J.; Wang, L.; Liu, H.; Liu, Y.; Wu, W.; Wang, F. Synthesis of polyaniline nanoparticles deposited on two-dimensional titanium carbide for high-performance supercapacitors. *Mater. Lett.* **2018**, *214*, 84−87.

(14) Li, X.; Xu, J.; Jiang, Y.; He, Z.; Liu, B.; Xie, H.; Li, H.; Li, Z.; Wang, Y.; Tai, H. Toward agricultural ammonia volatilization monitoring: A flexible polyaniline/$Ti_3C_2T_x$ hybrid sensitive films based gas sensor. *Sens. Actuators, B* **2020**, *316*, 128144.

(15) Gu, P.; Song, S.; Zhang, S.; Wei, B.; Wen, T.; Wang, X. Enrichment of U(VI) on polyaniline modified Mxene composites studied by batch experiment and mechanism investigation. *Huaxue Xuebao* **2018**, *76* (9), 701−08.

(16) (a) Wei, H.; Dong, J.; Fang, X.; Zheng, W.; Sun, Y.; Qian, Y.; Jiang, Z.; Huang, Y. $Ti_3C_2T_x$ MXene/polyaniline (PANI) sandwich intercalation structure composites constructed for microwave absorption. *Compos. Sci. Technol.* **2019**, *169*, 52−59. (b) Cao, M.-S.; Cai, Y.-Z.;

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477

He, P.; Shu, J.-C.; Cao, W.-Q.; Yuan, J. 2D MXenes: Electromagnetic property for microwave absorption and electromagnetic interference shielding. *Chem. Eng. J.* **2019**, *359*, 1265−302.
(17) Boota, M.; Anasori, B.; Voigt, C.; Zhao, M.-Q.; Barsoum, M. W.; Gogotsi, Y. Pseudocapacitive electrodes produced by oxidant-free polymerization of pyrrole between the layers of 2D titanium carbide (MXene). *Adv. Mater.* **2016**, *28* (7), 1517−22.
(18) Chen, B.; Song, Q.; Zhou, Z.; Lu, C. A novel sandwiched porous MXene/polyaniline nanofibers composite film for high capacitance supercapacitor electrode. *Adv. Mater. Interfaces* **2021**, *8* (12), 2002168.
(19) (a) Wang, Z.; Cheng, Z.; Xie, L.; Hou, X.; Fang, C. Flexible and lightweight $Ti_3C_2T_x$ MXene/Fe_3O_4@PANI composite films for high-performance electromagnetic interference shielding. *Ceram. Int.* **2021**, *47* (4), 5747−57. (b) Cai, Y.-Z.; Fang, Y.-S.; Cao, W.-Q.; He, P.; Cao, M.-S. MXene-CNT/PANI ternary material with excellent super-capacitive performance driven by synergy. *J. Alloys Compd.* **2021**, *868*, 159159. (c) Li, K.; Wang, X.; Li, S.; Urbankowski, P.; Li, J.; Xu, Y.; Gogotsi, Y. An ultrafast conducting polymer@MXene positive electrode with high volumetric capacitance for advanced asymmetric supercapacitors. *Small* **2020**, *16* (4), 1906851.
(20) Jia, X.; Shen, B.; Zhang, L.; Zheng, W. Construction of compressible polymer/MXene composite foams for high-performance absorption-dominated electromagnetic shielding with ultra-low reflectivity. *Carbon* **2021**, *173*, 932−40.
(21) Raagulan, K.; Braveenth, R.; Kim, B. M.; Lim, K. J.; Lee, S. B.; Kim, M.; Chai, K. Y. An effective utilization of MXene and its effect on electromagnetic interference shielding: flexible, free-standing and thermally conductive composite from MXene−PAT−poly(p-amino-phenol)−polyaniline co-polymer. *RSC Adv.* **2020**, *10* (3), 1613−33.
(22) Wang, Y.; Gao, X.; Zhang, L.; Wu, X.; Wang, Q.; Luo, C.; Wu, G. Synthesis of Ti_3C_2/Fe_3O_4/PANI hierarchical architecture composite as an efficient wide-band electromagnetic absorber. *Appl. Surf. Sci.* **2019**, *480*, 830−38.
(23) Gao, X.; Wang, B.; Wang, K.; Xu, S.; Liu, S.; Liu, X.; Jia, Z.; Wu, G. Design of $Ti_3C_2T_x$/TiO_2/PANI multi-layer composites for excellent electromagnetic wave absorption performance. *J. Colloid Interface Sci.* **2021**, *583*, 510−21.
(24) Yin, G.; Wang, Y.; Wang, W.; Yu, D. Multilayer structured PANI/MXene/CF fabric for electromagnetic interference shielding constructed by layer-by-layer strategy. *Colloids Surf., A* **2020**, *601*, 125047.
(25) Shahzad, F.; Alhabeb, M.; Hatter, C. B.; Anasori, B.; Man Hong, S.; Koo, C. M.; Gogotsi, Y. Electromagnetic interference shielding with 2D transition metal carbides (MXenes). *Science* **2016**, *353* (6304), 1137−40.
(26) (a) Ma, G.; Sun, J.; Aslani, F.; Huang, Y.; Jiao, F. Review on electromagnetic wave absorbing capacity improvement of cementitious material. *Constr. Build. Mater.* **2020**, *262*, 120907. (b) Yao, Y.; Jin, S.; Zou, H.; Li, L.; Ma, X.; Lv, G.; Gao, F.; Lv, X.; Shu, Q. Polymer-based lightweight materials for electromagnetic interference shielding: a review. *J. Mater. Sci.* **2021**, *56* (11), 6549−80. (c) Iqbal, A.; Sambyal, P.; Koo, C. M. 2D MXenes for Electromagnetic Shielding: A Review. *Adv. Funct. Mater.* **2020**, *30* (47), 2000883. (d) Geetha, S.; Satheesh Kumar, K. K.; Rao, C. R. K.; Vijayan, M.; Trivedi, D. C. EMI shielding: Methods and materials—A review. *J. Appl. Polym. Sci.* **2009**, *112* (4), 2073−86. (e) Liu, J.; Zhang, L.; Wu, H. Electromagnetic wave-absorbing performance of carbons, carbides, oxides, ferrites and sulfides: review and perspective. *J. Phys. D: Appl. Phys.* **2021**, *54* (20), 203001. (f) Wang, Z.; Cheng, Z.; Fang, C.; Hou, X.; Xie, L. Recent advances in MXenes composites for electromagnetic interference shielding and microwave absorption. *Composites, Part A* **2020**, *136*, 105956.
(27) (a) Zhu, L.; Zeng, X.; Chen, M.; Yu, R. Controllable permittivity in 3D Fe_3O_4/CNTs network for remarkable microwave absorption performances. *RSC Adv.* **2017**, *7* (43), 26801−08. (b) Liang, J.; Wang, Y.; Huang, Y.; Ma, Y.; Liu, Z.; Cai, J.; Zhang, C.; Gao, H.; Chen, Y. Electromagnetic interference shielding of graphene/epoxy composites. *Carbon* **2009**, *47* (3), 922−25. (c) Wang, S.; Zhao, Y.; Gao, M.; Xue, H.; Xu, Y.; Feng, C.; Shi, D.; Liu, K.; Jiao, Q. Green synthesis of porous cocoon-like rGO for enhanced microwave-absorbing performances. *ACS Appl. Mater. Interfaces* **2018**, *10* (49), 42865−74. (d) Xie, H.; Wu, H.; He, Z.; Kong, Z. Research progress in MoS_2/MXene nano-composites. *Acta Mater. Compos. Sin.* **2021**.
(28) (a) Cui, L.; Tian, C.; Tang, L.; Han, X.; Wang, Y.; Liu, D.; Xu, P.; Li, C.; Du, Y. Space-confined synthesis of core−shell $BaTiO_3$@carbon microspheres as a high-performance binary dielectric system for microwave absorption. *ACS Appl. Mater. Interfaces* **2019**, *11* (34), 31182−90. (b) Quan, B.; Liang, X.; Ji, G.; Cheng, Y.; Liu, W.; Ma, J.; Zhang, Y.; Li, D.; Xu, G. Dielectric polarization in electromagnetic wave absorption: Review and perspective. *J. Alloys Compd.* **2017**, *728*, 1065−75.

https://doi.org/10.1021/acsomega.1c02996
ACS Omega 2021, 6, 22468−22477