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# Heteronanoarchitecture of $Ti_3C_2T_x$ MXene and Amorphous MOF for Exceptional Durability in Electro-Ionic Soft Actuator

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The assembly of 2D nanosheets with other functional nanomaterials enables the creation of materials with unique property combinations that cannot be achieved in single-phase materials. In particular, a combination of inorganic and organic components provides a pathway to structures offering highly durable ionic and electronic conductivity simultaneously. Here, a controlled growth of amorphous metal-organic framework (aMOF) in the interlayer spaces of  $Ti_3C_2T_4$  MXene for enhancing oxidation stability and accelerating fast ion transport is reported. The hydrophilic terminations of MXene provide support for the continuous growth of iron-based aMOF in the available interlayer 2D slits. Effective electronic interactions involving hydrogen bonding, coordination, and esterification in-between the open surfaces of MXene and nanoporous aMOF enhance the electrochemical strength of MXene-aMOF hybrid electrodes and allow the design of extremely durable electro-ionic soft actuators. The MXene-aMOF exhibits a fivefold increment in electroactuation compared to a conventional poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) soft actuator, with robust stability up to 50 000 cycles in open air. Using the MXene-aMOF soft actuator, a deformable morphing surface with reversibly adjustable shapes and patterns is demonstrated. research. Over 100 distinct MXenes can be formed by altering the metal atoms in both in-plane and out-of-plane configurations, enabling the variation of properties for specific applications.<sup>[1]</sup> Surface terminations increase the number of possible stoichiometric compositions to over 1 000. This does not include solid solutions on M and X sites and MXenes with mixed surface terminations. To date, the versatile physical and chemical properties of MXenes have played a significant role in a wide range of advanced technologies, including energy storage and harvesting,<sup>[2,3]</sup> smart electronics and textiles,<sup>[4]</sup> electromagnetic interference shielding and antennas, [5-7] smart sensors and actuators,<sup>[8-10]</sup> selective adsorption and filtration,<sup>[11,12]</sup> optoelectronics,<sup>[13]</sup> biomedicine,<sup>[14]</sup> and catalysis,<sup>[15,16]</sup> to name a few. Considering superior electrical conductivity, tunable optoelectronic, and high mechanical properties, MXenes launched a new field of electronics - MXetronics. Adjustable dispersion viscosity and colloidal

## 1. Introduction

Soon after the discovery of  $Ti_3C_2T_x$  in 2011, 2D transition metal carbides, defined as MXenes, significantly impacted materials

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stability in aqueous solution allow MXene inks to contribute to eco-friendly printing for flexible electronics.<sup>[17,18]</sup> Progress in manufacturing high-performance MXene ink-based printable electronic devices has been reported.<sup>[8]</sup> At this stage, it is

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necessary to consider the long-term stability of MXene/MXene ink for scale-up to real-world MXetronics.

MXenes are typically obtained by the selective etching aluminum layers from their MAX phase precursors. Removing the aluminum causes the available moieties in the nearby environment to terminate the surface, forming O, OH, F, Cl, and other terminations. Although this surface termination stabilizes the MXene in a closed system, the corresponding substitutional defects make the titanium or other M metal atoms susceptible to oxidation. Therefore, it may react gradually with water and either dissolved oxygen in the colloidal state or atmospheric oxygen in the solid state, losing its important structural properties by forming titanium dioxide. MXenes would benefit from complete protection of the surface terminals for each layer of MXene immediately after the synthesis to preserve the structural identity for long-term open-air and/or extreme environment applications.

Many approaches to protect the surface oxidation of MXene have been reported.<sup>[19-21]</sup> The most common approach involves engaging surface terminals with suitable guest species, such as metal ions,<sup>[22,23]</sup> functional organic linkers,<sup>[23-25]</sup> and other functional carbon materials.<sup>[26–28]</sup> Protection by metal ions or discrete organic linkers can successfully make the MXene a better candidate for electrocatalysis or smart electronic skin, but its stability under air in humid environments may limit some applications. In addition, these protections do not add value to the storage capacity of electrochemical charges and ions and, thus, are unsuitable for high-performance energy storage devices. Meanwhile, efforts have been made to combine suitable functional covalent-organic frameworks (COFs) and metal-organic frameworks (MOFs) with MXene to enhance their electrochemical performance.<sup>[29-33]</sup> However, the crystalline macro/micro-scale structural configurations of COFs and MOFs render them incapable of penetrating the nanoscale interlayer spacing of 2D MXene, resulting in surface deposition.<sup>[29-32]</sup> Therefore, these combinations of MXene and MOFs/COFs augmented the electrochemical ion storage capacity by enlarging the overall surface area of active electrodes, but did not stabilize the MXene interlayers. Consequently, while an enhancement in the capacity for electrochemical ion storage was observed, the lifetime was reduced due to the instability of MXene.<sup>[33]</sup>

Herein, we decorated the interlayer spaces of  $Ti_3C_2T_r$  MXene by controlling the growth of nanoporous amorphous metalorganic framework (aMOF) to develop highly durable electroionic soft actuators. The host-guest interaction mechanism observed in parasitic plants has been emulated in the growth of iron-based *a*MOF via the in situ method between the interlayers of MXene (Figure 1a). It is postulated that MXene exhibited host-like behavior, manifesting discrete and regular structural configurations. The growth of aMOF as a guest with irregular sizes and shapes was directed and controlled by the surface terminations of MXene layers via physicochemical interactions (Figure 1b). The amorphous structure of MOF enables it to effectively occupy and electronically stabilize the limited subnanometer or few-nanometer wide interlayer spaces of MXene, owing to the absence of long-range periodic order in MOF.<sup>[34]</sup> This phenomenon contrasts with conventional crystalline MOFs, which are characterized by the presence of long-range periodic order of basic building blocks within their structural frameworks.<sup>[34]</sup> The physicochemical interactions, including H-bonding,

coordination, and esterification, prevent oxidation of MXene and simultaneously provide an augmented electrochemical interactive surface for ion transportation. This resulted in a notable enhancement in the ion storage capacity of MXene, exhibiting a specific capacitance of 305.4 F g<sup>-1</sup> at a scan rate of 5 mV s<sup>-1</sup> and retaining  $\geq$ 235.2 F g<sup>-1</sup> even at 100 mV s<sup>-1</sup> in a non-aqueous 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIM-BF<sub>4</sub>) electrolyte. With regard to electrochemical stability in the electro-ionic actuators, the aMOF-decorated MXene interface, employed as the active electrode material, exhibited a retention of  $\geq$ 98.77% even after 50 000 actuation cycles in the open air. Additionally, it substantially improved the actuation performance, displaying a mechanical deflection of  $\geq$  20.30 mm at an ultralow voltage of  $\pm$  0.5 V and a rapid response time of ≤1.3 s without back relaxation. This notable advancement in electrochemical stability and actuation performance paves the way for constructing an active morphing surface with reconfigurable dimensions and patterns, offering a potential avenue for application in smart frequency-selective surfaces (FSS).

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#### 2. Results and Discussion

# 2.1. Synthesis of MXene–*a*MOF Electrode for Electro-Ionic Soft Actuators

To prevent any structural damage, the charged ampoule containing the  $Ti_2C_2T_x$  MXene ink, ferric chloride, benzophenone-3,3',4,4'-tetracarboxylic dianhydride (BPTC), and N.Ndimethylformamide (DMF) was subjected to complete degassing under vacuum through a series of freeze-pump-thaw cycles. Subsequently, the glass ampoule was flame-sealed in a degassed state and placed within a well-maintained, temperature-controlled oven for solvothermal treatment at 120 °C for 72 h. The initial light gray solution mixture transformed into orange-brown sediment of MXene-aMOF, settling at the bottom of the ampoule after the complete growth of *a*MOF within the available interlayer spaces of MXene (Figure 1c). Figure S1 (Supporting Information) depicts the real-time images of the glass ampule both before and after the solvothermal process. It was postulated that during the freeze-pump-thaw cycles, the BPTC interacted with the surface terminations of each MXene layer, subsequently coordinating with the iron ions during the thermal treatment (Figure S2, Supporting Information). The unreacted iron salts and BPTC were removed through a series of washes with DMF and deionized water. The specific details regarding the experimental parameters were discussed in depth within the Experimental Section. The amorphous nature of the MOF (Figure S3, Supporting Information) was observed to be successfully grown on the interlayer spaces of MXene, as evidenced by the comparative few nanometer scale transmission electron microscopic (TEM) images of MXene and MXene-aMOF (Figure 1d). This distinctive stabilization technique endows the MXene-aMOF with remarkable chemical stability, establishing a metallic pathway for electron conduction and facilitating the expansion of active surfaces. These attributes have the potential to usher in a new era of MXene and MOF application in advanced electrochemical capacitive devices. Figure 1e schematically illustrates the potential of MXene-aMOF active electrode material to enhance the air-working actuation performances of an

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Natural parasitism

0

Cl

Ti₃C₂T<sub>×</sub>

host

guest

H Surf. O/F/OH

N2-

a

С

FeCl<sub>3</sub>·6H<sub>2</sub>C

Fe С

Ti







Figure 1. Inspiration, synthesis, and hypothesis of MXene-aMOF electrode for electro-ionic soft actuators. Conceptual illustration for host-guest interaction mechanism of a) natural parasitism and b) analogous MXene-aMOF: parasitism in-between MXene and aMOF. c) Air-free synthesis of MXeneaMOF: growth of aMOF at the available interlayer spaces of MXene. d) Comparative nanostructural interlayer configuration of MXene and MXene-aMOF: High-resolution nanoscale scanning TEM and corresponding schematic illustrations. e) Schematic illustration for the potential of MXene-aMOF active electrode material to enhance the electrochemical actuation performance of an air-working soft actuator device.

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**Figure 2.** Morphological analysis of *a*MOF and MXene–*a*MOF nanostructural surfaces. High-resolution SEM images of a–c) *a*MOF and d–f) MXene–*a*MOF active materials. g–i) Nanometer-scale TEM images of MXene–*a*MOF.

electrochemical soft actuator. The protection in MXene–*a*MOF can provide exceptional durability in oxidizing environments. The metallic conducting pathway enables rapid ion intercalation and deintercalation during electrical signal switching, enhancing actuation speed. The enlarged surface area also provides a highly active platform capable of accommodating a substantial volume of electrolyte ions, thereby maximizing the mechanical bending and actuation force.

# 2.2. Morphological Analysis of *a*MOF and MXene–*a*MOF Materials

The high-resolution scanning electron microscope (SEM) images revealed a highly porous surface of *a*MOF, which comprised an interconnected irregular nanonetwork of iron-based MOF particles (**Figure 2**a–c). This irregular nanostructure provided preliminary evidence that the particles could undergo growth between the narrow interlayer spaces of  $Ti_3C_2T_x$  MXene. This hypothesis was further confirmed by the SEM and TEM analyses of MXene-aMOF. The SEM images revealed that the interlayer spaces of MXene were entirely occupied by the amorphous carbon structures of aMOF (Figure 2d-f; Figure S4, Supporting Information). The high-resolution TEM images confirmed the successful growth of aMOF, as evidenced by the well-defined nanostructural surface configurations observed in the MXene-aMOF (Figure 2g-i; Figure S5, Supporting Information). Figure 2g illustrates the vertical and horizontal alignment of MXene-aMOF layers, while the decoration of *a*MOF at the interlayers of MXene is reflected in the scanning TEM images (Figure 2h; Figure S5, Supporting Information). At the sub-nanometer scale in TEM, the integration of amorphous MOF structures at the interlayers of crystalline  $Ti_3C_2T_x$  MXene was discernible (Figure 2i). The typical structural configuration of MXene-aMOF was corroborated by the elemental mapping images generated from the scanning TEM high-angle annular dark-field (HAADF) tomography (Figure S6a, Supporting Information). The distribution of characteristic atoms, including titanium (Figure S6b,

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**Figure 3.** Structural analysis of *a*MOF and MXene–*a*MOF active electrode material. a) Powder XRD patterns of  $Ti_3C_2T_x$  MXene and MXene–*a*MOF materials. b) Comparative nitrogen-physisorption and pore-size distribution characteristics of *a*MOF and MXene–*a*MOF. c) FTIR spectral patterns of *a*MOF and MXene–*a*MOF materials in transmission mode. XPS spectral analysis of d)  $Ti_2p$ , e) O1s, and f) C1s atomic configurations of MXene–*a*MOF active material.

Supporting Information), iron (Figure S6c, Supporting Information), carbon (Figure S6d, Supporting Information), oxygen (Figure S6e, Supporting Information), and fluorine (Figure S6f, Supporting Information), substantiates the presence of *a*MOF and MXene mixed at the nanoscale level. Furthermore, elemental maps of iron from *a*MOF and titanium from MXene show their homogeneous distribution at the nanometer scale, in accordance with the tomography image of MXene–*a*MOF (Figure S7a,b, Supporting Information). Therefore, it can be concluded that the surface terminations of MXene consistently facilitated the growth of *a*MOF, even within interlayer spaces. This stabilizes MXene electronically and expands the active surfaces with nanoporous framework structures to maximize the electrochemical interactions with electrolyte ions.

#### 2.3. Structural Analysis of aMOF and MXene-aMOF Materials

A comprehensive spectroscopic analysis was conducted to assess the structural configuration of the synthesized MXene–aMOF hybrid. The powder X-ray diffraction (XRD) analysis demonstrated that the regular structure of Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene layers was maintained in the MXene–aMOF (**Figure 3**a). The characteristic (002) lattice plane of MXene exhibited a distinctive shift

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in the diffraction angle (2 $\theta$ ) from 7.4° to 6.6°, which was attributed to the expansion of *d*-spacing as a consequence of *a*MOF growth at interlayers.<sup>[35]</sup> The predominantly amorphous character of the aMOF, exhibiting an average structural correlation length of  $\approx$ 1.05–1.25 nm, was ascertained through small-angle X-ray scattering (SAXS) analysis (Figure S8, Supporting Information). The lower intensity of other MXene peaks may have resulted in their complete submergence within the MXene-aMOF due to the dominant amorphous XRD signal of the iron-based MOF (Figure S3, Supporting Information). The integration of nanoporous MOF at the interspaces resulted in an increase in the specific surface area of  $Ti_3C_2T_y$  MXene up to 260 m<sup>2</sup> g<sup>-1</sup>, which was slightly lower than that of pristine *a*MOF (282 m<sup>2</sup> g<sup>-1</sup>) (Figure 3b). The pore-size distribution of MXene-aMOF exhibited a distinctive nanoporous structure at the micro- and mesoporous regions, with the pore widths of 1.4, 2.7, 3.8, 5.9, and 7.7 nm (inset of Figure 3b). This phenomenon was attributed to the growth control of aMOF induced by the MXene surface terminations as the host. Without a host, the aMOF exhibited an irregular pore-size distribution, spanning a wide range from 2 to 100 nm, as illustrated in Figure S9a (Supporting Information). In contrast, pristine Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene displays microporosity, with a dominant pore size centered around 1.5 nm (Figure S10, Supporting Information). The integration with MXene limits the maximum pore size to 7.7 nm, suggesting spatial confinement within the MXene interlayers. Detailed analysis indicates minimal variation in the pore size distribution within the 2-8 nm range between the pristine aMOF and the MXene-aMOF, confirming that this porosity primarily originates from the aMOF (inset of Figure 3b). This structural evolution is attributed to the templating and growth-directing influence of the MXene surface terminations, which act as nucleation and confinement sites during *a*MOF formation. Another crucial physicochemical property that renders the MXene-aMOF an optimal candidate for electrochemical ion storage and discharge is the surface polarity of aMOF. The instantaneous carbon dioxide ( $CO_2$ ) uptake capacity of 53 mg g<sup>-1</sup> at atmospheric pressure indicates a robust electrostatic interaction with the guest CO<sub>2</sub> molecules, which can serve as capacitive charge and discharge pockets during electrochemical switching (Figure S9b, Supporting Information). The atomic-level structural characterization of aMOF, which supports its plausible chemical structure (Figure S2, Supporting Information), is reported in Figure S11a-d (Supporting Information), and a brief discussion is provided in the Supporting Section. Additionally, the formation of similarly disordered framework structures in both aMOF and MXene-aMOF was confirmed by analyzing the atomic configuration of iron in each material (Figure S12, Supporting Information). The surface functionalities associated with the chemical conformations of aMOF and MXene-aMOF were verified by the transmission Fourier-transform infrared (FTIR) spectra (Figure 3c; Figure S13, Supporting Information). The retention of polar functionalities, including O-H, C-H, aromatic (Ar-region) C=C, C=O, C-O, C-H, along with Fe-O at aMOF, and additional Ti-O, and Ti-C at MXene-aMOF, is illustrated in the corresponding FTIR spectra (Figure 3c). It was observed that there was a notable shifting of characteristics in FTIR transmission peaks at the MXene-aMOF, due to the strong electronic interaction of the aMOF with the MXene surface terminations, as compared with the pristine MXene spectra

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(Figure S13, Supporting Information). The transmission peaks for the O-H and Ti-C/O stretching of the MXene exhibited a shift from 3436 cm<sup>-1</sup> to 3422 cm<sup>-1</sup> and 551 cm<sup>-1</sup> to 567 cm<sup>-1</sup>, respectively, in the MXene-aMOF. In contrast, the aMOF displayed a broader peak at 3409 cm<sup>-1</sup> for the O–H stretching mode of vibration. As illustrated in Figure 3c and Figure S13 (Supporting Information), the FTIR transmission patterns at Ar-region for the MXene-aMOF were substantially different from both MXene and aMOF due to the plausible formation of H-bonds, coordination, and esterification interactions. The magnified FTIR transmission signals of aMOF and MXene-aMOF in the spectral range of 1 760-1 200 cm<sup>-1</sup> further substantiate the occurrence of strong interactions at the interface (Figure S14, Supporting Information). To gain deeper insight into these physicochemical interactions, the atomic configurations of the constituent elements were investigated by X-ray photoelectron spectroscopy (XPS). The deconvoluted Ti2p XPS spectra corroborate the atomic-level interaction of titanium and oxygen, as evidenced by the presence of XPS signals at 458.92 and 464.46 eV, which correspond to Ti2 $p_{3/2}$  (Ti–O) and Ti2 $p_{1/2}$  (Ti–O) electronic configurations, respectively (Figure 3d).<sup>[36]</sup> In contrast, these Ti2p (Ti–O) signals were not observed in the deconvoluted XPS spectra of the pristine MXene (Figure S15a, Supporting Information).<sup>[36]</sup> The Ti-O interaction, as displayed in the MXene-aMOF material, indirectly supports the formation of coordination and ester linkages between the carboxylic functional groups of aMOF and the hydroxyl surface terminations of MXene (Figure S2, Supporting Information). Additional distinctive XPS peaks for MXeneaMOF were observed at binding energies of 456.40 and 457.23 eV, which correspond with the electronic states of  $Ti2p_{3/2}$  and that of 461.40 and 462.53 eV for  $\text{Ti}2p_{1/2}$  for the Ti–C and Ti<sup>+2</sup> configurations, respectively (Figure 3d). For  $Ti_3C_2T_2$ , MXene, these two configurations were found to appear at the binding energies of 455.01 and 456.30 eV for  $Ti2p_{3/2}$  and 460.87 and 462.31 eV for Ti $2p_{1/2}$  states (Figure S15a, Supporting Information). The increased binding energy for Ti2p in the MXene-aMOF material was also supported by the fact that the carboxyl functional groups of aMOF were involved in stabilizing the MXene surface terminations. The involvement of the aforementioned coordination and esterification interactions was further corroborated by the emergence of the O-Ti deconvoluted O1s XPS peak at a binding energy of 530.33 eV in the MXene-aMOF (Figure 3e). The deconvoluted O1s XPS spectra of MXene-aMOF revealed the presence of a total of five distinguishable electronic states of oxygen, corresponding to the plausible electronic environments of O=C, OH/Ox-C-Ti, O=C-O, O-Ti, and O=C-O-Fe, at the binding energies of 533.23, 532.03, 531.19, 530.33, and 529.73 eV, respectively. While the pristine MXene showed only two distinct deconvoluted O1s XPS peaks at 531.23 and 532.03 eV, corresponding to the Ox-C-Ti and OH-C-Ti configurations, respectively (Figure S15b, Supporting Information).<sup>[36]</sup> The five plausible carbon configurations of MXene-aMOF, as indicated by C=O, O-C=O, C-C/C-O, C=C, and C-Ti (Figure S2, Supporting Information), were also reflected in the deconvoluted C1s XPS spectra with the characteristic binding energies of 288.78, 288.15, 285.93, 284.97, and 284.43 eV, respectively (Figure 3f). In conclusion, this comprehensive analysis not only validates the structural configuration of MXene-aMOF, but also provides exclusive evidence supporting the hypothesis that the electronic ADVANCED SCIENCE NEWS \_\_\_\_\_\_ www.advancedsciencenews.com

stabilization of MXene surface terminations is achieved through physicochemical interactions. Furthermore, the series of density functional theory (DFT) simulations significantly reinforces the strong electronic coupling between the carboxylic functional groups of the *a*MOF and the surface terminations of  $Ti_3C_2T_x$  MXene (Figures S16–S20, Supporting Information).

## 2.4. Electro-Ionic Actuation Performance of MXene-aMOF Electrodes

To comprehend the technological significance of this distinctive combination of MXene and aMOF, the MXene-aMOF was utilized as an active electrode material for the nascent open-air electrochemical soft actuation under low input voltages. Before this, cyclic voltammetry (CV) measurements were conducted to ascertain the ion storage capacity in a nonaqueous EMIM-BF<sub>4</sub> solution, which is the same electrolyte utilized in fabricating the soft actuator. Figure 4a illustrates the CV response patterns of the MXene-aMOF active electrode in an acetonitrile (ACN) solution of 0.5 M EMIM-BF<sub>4</sub> at varied scan rates (5–100 mV s<sup>-1</sup>) in the presence of Ag/AgCl. The MXene-aMOF exhibited a dominant electric double-layer capacitor (EDLC) nature of response patterns, confirming minimal redox reactions with the electrolyte ions and optimal stabilization by the electronic interactions between MXene and aMOF. Most importantly, aMOF greatly enlarged the accessible surface area due to hierarchically porous nanoarchitectures, substantially improving the electrochemical ion storage capacity at both low and high scan rates (Figure 4b). In the same EMIM-BF<sub>4</sub> electrolyte, the MXene–aMOF exhibited an ionic specific capacitance enhancement of 8.7-fold and 2.5-fold at a scan rate of 5 and 100 mV s<sup>-1</sup>, respectively, in comparison with pristine MXene electrodes. The specific capacitance of the MXene–aMOF electrode material was 306 F g<sup>-1</sup> at a scan rate of 5 mV s<sup>-1</sup> and remained at a high level to 100 mV s<sup>-1</sup>, with a loss of only 23% (236 F g<sup>-1</sup>). In contrast, the MXene electrode material exhibited a loss of 78% in its initial capacitance when the scan rate increased from 5 to 100 mV s<sup>-1</sup> (Figure 4b). The enhancement of capacitive properties in the MXene-aMOF has led to a notable improvement in the conventional open-air actuation parameters of a dry-type electrochemical soft actuator under ultralow voltage. The Ti<sub>3</sub>C<sub>2</sub>T<sub>y</sub> MXene-based soft actuator is known to exhibit rapid actuation, with a response time of approximately one second, due to its electrical conductivity, comparable to that of metals.<sup>[10]</sup> However, due to the limited accessibility of electrolyte ions to the active microporous-dominant surfaces, the pure MXene actuator demonstrated relatively low mechanical bending deflection. Furthermore, the open-air stability of a MXene-only actuator is a concern due to the presence of unprotected surface, which can lead to deterioration with time.<sup>[37]</sup> The MXene-aMOF hybrid can overcome these limitations, with a peak-to-peak bending deflection of 15.8 mm observed at an ultralow voltage of 0.5 V and an excitation frequency of 0.1 Hz (Figure 4c). The device exhibited ultrafast actuation with a response time of 1.3 s under 0.5 V direct-current (DC), without any back relaxation up to 1000 s (Figure 4d). A threshold actuation voltage as low as 0.01 V was observed (Figure 4e), and the device maintained robust airworking stability up to 50 000 cycles, retaining 98.77% of its initial performance (Figure 4f). A comparison of the electrochemical actuation properties of the MXene-aMOF with recently reported analogous materials substantiates it significance for advancing soft actuation technology (Table S1, Supporting Information). The intriguing electron conduction pathways and expanded electrolyte accessible active surfaces of MXene-aMOF have resulted in a significant increase of mechanical deflection, up to 5.4-fold and 2-fold, respectively, when compared to similar actuators based on poly(3,4-ethylenedioxythiophene)poly(styrenesulfonate) (PP) and pristine *a*MOF (Figure 4c). In a DC input potential of 0.5 V, the MXene-aMOF exhibited ultrafast mechanical deflection of 11.60 mm, which is fivefold larger than that of the PP soft actuator (Figure 4d). The distinctive mechanical deflection patterns as observed in the MXene-aMOF actuator under different square wave input potentials (0.01-0.5 V at 0.1 Hz) confirm the controllability of actuation at ultralow voltages (Figure 4e). It is noteworthy that the MXene-aMOF exhibited remarkable operational stability in an open air when stimulated at a frequency of 1.0 Hz and 1.0 V, particularly in comparison to the pristine  $Ti_3C_2T_x$ , as previously studied.<sup>[37]</sup> The MXene actuator exhibited a retention of only 86% after 10 000 cycles of continuous excitation at 1.0 Hz and 1.0 V, with a noticeable decline in actuation performance with each subsequent cycle.<sup>[37]</sup> In contrast, the MXene-aMOF soft actuator demonstrated a remarkable stability, retaining 98.77% even after 50 000 cycles without any significant decline (Figure 4f). Furthermore, under challenging environmental conditions, including elevated temperature and high relative humidity, the MXene-aMOF actuator exhibited excellent resilience and robust performance, underscoring its potential for reliable operation in practical applications (Figure S21a,b, Supporting Information). The stabilization of the MXene surface termination via electronic means using aMOF proved to be a crucial factor in attaining the desired electrochemical actuation stability in open-air conditions. Therefore, the proposed stabilization study regarding parasitic protection represents a novel approach to mitigating unintended side reactions of oxidation or degradation processes that compromise the performance or longevity of materials and devices. In the context of MXene-aMOF hybrids, parasitic protection involves addressing challenges such as oxidation, ion trapping, restacking of layers, and chemical instability. No significant structural or functional changes were observed in the MXene-aMOF active materials even after two years of storage under ambient conditions (Figure S22a,b, Supporting Information).

# 2.5. Demonstration of Actively Morphing Surfaces Based on MXene–*a*MOF Actuator

To gain insight into the electroactive mechanical deformation characteristics of the MXene–aMOF soft actuator as actively morphing surfaces, three sets of patterned surfaces were fabricated in a 2 × 2 matrix configuration (Figure 5a–c). The number of active wings was varied (4, 6, and 8 wings in Figure 5a–c, respectively) in each patterned surface to produce different extents of void volume through low-voltage electrical stimulation (Figure S23a,b, Supporting Information). The MXene–aMOF active metasurfaces exhibited typical out-of-plane mechanical deformation, which resulted in a change of the relative void fraction under the DC stimulation of +3.5 and -3.5 V,

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**Figure 4.** Electrochemical performance of MXene–*a*MOF electrode and actuator. a) The CV response patterns of the MXene–*a*MOF at varying scan rates (5–100 mV s<sup>-1</sup>). b) Comparison of specific capacitance for MXene and MXene–aMOF active electrode materials as a function of increasing scan rates, ranging from 5–100 mV s<sup>-1</sup>. c) The comparative electrochemical actuation responses, measured in terms of mechanical bending deflection over time for the PP, *a*MOF, and MXene–*a*MOF actuators at  $\pm$  0.5 V and 0.1 Hz. d) The typical bending deflections of PP, *a*MOF, and MXene–*a*MOF soft actuators under 0.5 V DC stimulation. Inset: shows deflection responses over 1 000 s under 0.5 V dc stimulation. e) The controlled mechanical bending deflections of the MXene–*a*MOF actuator under ultralow square wave input potentials (0.01–0.5 V) at an excitation frequency of 0.1 Hz. f) Comparative long-cycle air-operating actuation stability of the MXene and MXene–*a*MOF actuator at 1.0 V and 0.1 Hz. Inset: shows initial and final ten cycles for MXene–*a*MOF actuator.

respectively (Figure S24, Supporting Information for a schematic illustration). The mechanical deformations of the active morphing surface are illustrated, including top and side views as shown in Figure 5a–c, and the real-time actuation of different pattern surfaces is recorded in Movies S1, S2, and S3 (Supporting Information). These actuation images, which were presented both schematically and with real-time deformation images (Figure 5a–c), are denoted by T for the top-view and S for the side-view. It was observed that the 4-wing morphing sur-

face generated a relative void fraction of 44.9% of its initial void, followed by 17.8% and 11.5% for the 6-wing and 8-wing configurations, respectively (Figure S23b, Supporting Information). The potential for developing smart and active frequency selective surfaces (FSS) has been demonstrated by metasurfaces with reversible deformable capability.<sup>[38-40]</sup> Active FSSs, particularly those employing mechanical, thermal, optical, electrical, or chemical tuning mechanisms, often suffer from bulky configurations, high power consumption, and limited compatibility

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**Figure 5.** Demonstration of electroactive MXene–aMOF deformable metasurfaces. The schematic and real-time deformation images under DC voltages of +3.5 and -3.5 V, shown from both top and side views for the MXene-aMOF active metasurfaces with a) 4-wing, b) 6-wing, and c) 8-wing patterned configurations. T and S represent top-view and side-view, respectively.

with flexible or curved platforms.<sup>[41]</sup> Mechanically reconfigurable strategies, including origami, kirigami, or MEMS engineeredstretchable elastomers, present fabrication challenges in realizing complex and diverse metasurface patterns, often requiring multi-step and labor-intensive processes.<sup>[42-45]</sup> From this perspective, the metasurface patterned with various geometries on the surface of a MXene–aMOF actuator via laser processing offers simplified fabrication, reduced power consumption, and excellent compatibility with flexible and curved platforms. Moreover, such low-voltage-driven electrochemical soft actuators, which exhibit controlled out-of-plane mechanical deformation and durability in open-air conditions, represent a promising avenue for advancing active FSS. Notably, the out-of-plane mechanical deformation of the MXene–*a*MOF active metasurface can be

harnessed to regulate the electromagnetic waves, with potential applications in electromagnetic wave communication and shielding. The simulations suggest that void fraction differentiation can lead to a target frequency shift of 0.3 GHz for the MXene– MOF active metasurface, while the 4-wing and 6-wing metasurface can be used as band-pass and band-stop FSS, respectively (details are provided in Figure S25, Supporting Information).

## 3. Conclusions

We presented a method to protect MXene surface from oxidation by integrating amorphous MOFs between the MXene nanosheets through coordination, esterification, and hydrogen bonding interactions. The nanoscale hybridization of MXene and *a*MOF ADVANCED SCIENCE NEWS \_\_\_\_\_\_ www.advancedsciencenews.com

yielded remarkably high electrochemical stability and ion storage capacity, due to the synergistic combination of large active surfaces of nanoporous aMOF and the metallic electronic conductivity of MXene. The specific surface area of MXene at the MXene–*a*MOF interface was enhanced from  $\approx 30^{[46]}$  to 260 m<sup>2</sup>  $g^{-1}$ , while the gravimetric capacitance increased to 236 F  $g^{-1}$  in ionic-liquid electrolyte at a high scan rate of 100 mV s<sup>-1</sup> when pure MXene demonstrated the capacitance of only 27 F g<sup>-1</sup>. This improvement in electrochemical performance was supported by the 5.4-fold expansion of mechanical deflection observed in the MXene-aMOF soft actuator device, which exhibited actuation retention of close to 99% after 50 000 cycles in open air. The mechanically deformable MXene-aMOF actuator using low electrical power was successfully demonstrated. We created an active morphing surface for reversibly regulating the void fraction by the out-of-plane bending actuation with different electroactive patterns.

#### 4. Experimental Section

Growth of aMOF at Interlayers of  $Ti_2C_2T_x$  MXene: The  $Ti_2C_2T_x$  MXene ink was synthesized in accordance with the previously reported method.<sup>[37]</sup> In this typical growth process, a solution of ferric chloride hexahydrate (68 mg, 0.25 mmol) and BPTC (81 mg, 0.25 mmol) solution in DMF (7 mL) were added to a glass ampoule (20 mL) containing MXene ink (5 mL, 5 mg mL<sup>-1</sup>). To ensure homogeneity of the solution mixture, the charged ampoule was subjected to ultrasonication for 20 min and then underwent a total of five cycles of freeze-pump-thaw degassing. Subsequently, the ampoule was flame-sealed in a degassed state and subsequently transferred to a heating oven for the solvothermal growth of aMOF. The heating was initiated at 25 °C and increased at a rate of 2° min<sup>-1</sup> until reaching 120 °C, where it was maintained for 72 h before returning to ambient temperature via natural cooling. The growth of the aMOF was confirmed by a change in the color of the reaction mixture, which transitioned from light gray dispersion to an orange-brown sediment (Figure S1, Supporting Information). The resulting sediment was then collected and purified for subsequent characterization and electrochemical applications through washing with anhydrous DMF (100 mL) and acetone (50 mL). The screening process employed for optimizing the growth conditions of the aMOF was briefly described in the supporting information, and the reproducibility of the optimized synthesis was verified through consistent electrochemical CV responses (Figure S26, Supporting Information). To facilitate a comparative analysis, the synthesis of a pristine aMOF was carried out in parallel, employing an identical process but without the inclusion of MXene ink.

Fabrication of Electroactive MXene-aMOF Actuator and Morphing Surface: To validate the hypothesis and ascertain the crucial role played by MXene–aMOF as an electroactive material, a conventional method<sup>[10,37]</sup> was employed in the fabrication of electrochemical soft actuator and metasurface devices. An EMIM-BF₄ ionic liquid embedded Nafion (60 wt.%) solid-state electrolyte membrane with a thickness of 120 µm was initially prepared by solution casting in N,N-Dimethylacetamide (50 mg mL<sup>-1</sup>). The MXene-aMOF based electrode ink (0.5 mg mL<sup>-1</sup>), prepared freshly and comprising aqueous PEDOT-PSS in a weight ratio of 1:2, which serves as both a conductive additive and binder, was then drop cast on the surfaces of the electrolyte membrane. The MXene-aMOF electroactive soft actuator, with a thickness of 180  $\mu$ m, was cut into pieces with dimensions of 20 mm  $\times$  4 mm for the purpose of evaluating the basic actuation parameters. The fabrication methods were kept identical for preparing both the PEDOT-PSS only  $\PP$  actuator and the pristine *a*MOF soft actuators to ascertain the utility of MXene-aMOF in electroactive devices in open-air applications. To construct the active metasurface, a planar sheet MXene*a*MOF soft actuator with dimensions of 40 mm x 40 mm (length x width) was fabricated (Figure S14, Supporting Information). The shapes of the metasurfaces were symmetrically designed with a diameter of 15 mm using AutoCAD software. Each shape was then patterned on the surface of the sheet MXene–*a*MOF actuator in a 2 by 2 matrix configuration, with a distance of 17 mm between the centers of the shapes (i.e., a 2 mm distance between the wings of the shapes). The three shapes were classified according to the number of wings driven in each: 4-wing, 6-wing, and 8-wing. Surface patterning of the soft actuators was conducted using a  $CO_2$  laser engraving machine (C30, Coryart) with a laser power of 9.2 W and a patterning speed of 15 mm s<sup>-1</sup>.

Characterization: The surface morphologies were imaged using a field-emission SEM from JEOL, Japan (JSM-IT800). The structural integration of aMOF at the interlayer spaces of MXene was captured at the sub-nanometer scale using a double C<sub>s</sub> corrected TEM with STEM-HAADF attachment from FEI (Titan cubed G2 60-300). The short-range periodic structural order and crystallinity of the samples were characterized using high-resolution powder XRD with Cu K<sub>a</sub> radiation ( $\lambda = 1.54$  Å), performed on a SmartLab diffractometer (Rigaku), and SAXS measurements conducted on a NANOPIX instrument (Rigaku) with a sample to detector distance of 97 mm. The surface porosities and pore sizes were determined from the nitrogen adsorption-desorption isotherms at 77 K using a surface area and pore size analyzer (3Flex) from Micromeritics. Prior to the physisorption analysis, the samples were completely degassed under vacuum at 110 °C for 24 h. To ensure reproducibility, each measurement was repeated three times. The low-pressure carbon dioxide uptake by aMOF was determined using a static volumetric system at 273 K (ASAP 2020, USA) from Micromeritics. The temperature was maintained constant throughout the adsorption-desorption process using a circulator. In the physisorption figures, filed and blank symbols were used to represent the adsorption and desorption, respectively. The polar functionalities of aMOF and MXene-aMOF were evaluated using a FTIR spectrometer (Nicolet iS50) from Thermo Fisher Scientific. The atomic configurations of aMOF, MXene, and MXene-aMOF were determined by analyzing the XPS profiles, which were recorded using a photoelectron spectrometer (Nexsa G2) from Thermo Scientific.

DFT Simulations: All DFT simulations were performed using QuantumATK software, with subsequent data analysis conducted in Virtual NanoLab (Version 2019.12). The electronic structures of Ti, O, C, F, and H atoms were described using the linear combination of atomic orbitals method within the generalized gradient approximation, employing the BLYP exchange correlation functional. Structure optimization and energy calculations utilized double-zeta polarized basis sets combined with Fritz Haber Institute pseudopotentials. Electron density distributions and density of states were calculated using the same computational framework.

*Electrochemical Characterization*: The electrochemical CV of MXene and MXene–*a*MOF active materials was conducted in a conventional three-electrode configuration (Ag/AgCl as reference and platinum foil as counter electrode) using a multichannel potentiostat/galvanostat (VersaStat) from Princeton Applied Research. The ink of the active material was prepared with acetylene black and polytetrafluoroethylene in a known volume of 1-methyl-2-pyrrolidone (anhydrous, 99.5%, Sigma-Aldrich) at a weight ratio of 8:1:1. Prior to CV measurements, a freshly polished glassy carbon electrode was employed to load the active material in a calculated amount using a non-aqueous acetonitrile solution of 0.5 M EMIM-BF<sub>4</sub>. The specific capacitance ( $C_{sp}$ ) was calculated in F g<sup>-1</sup> using the following Equation (1).

$$C_{sp} = \frac{A}{2 \times \Delta V \times \nu \times S} \tag{1}$$

where A represents the area inside the CV curve,  $\Delta V$  is the potential window,  $\nu$  is the scan rate, and S signifies the weight of the active material on the working electrode.

*Electroactive Actuation Characterization in Open Air*: The ultralow voltage-driven mechanical bending deflection of the electrochemical soft actuators was recorded using a laser displacement sensor (Keyence, LK031) under various amplitudes and oscillation frequencies.

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The bending strain difference ( $\epsilon$ ) between the opposing electrode layers of the MXene–aMOF electrochemical soft actuator was calculated using the following Equation (2).<sup>[37,47]</sup>

$$\epsilon = \frac{2 \,\delta \,d}{l^2 + \delta^2} \tag{2}$$

In this equation,  $\delta$  is defined as the maximum tip displacement from the original position, detected by laser (i.e., half of the peak-to-peak displacement under AC excitation), *d* is given as the total thickness of the actuator, and *l* is specified as the effective active length.

*Void Fraction Characterization of Electroactive Metasurfaces*: To facilitate a precise and quantitative comparison of the change in vacancy area of the patterned metasurfaces, Image J software was employed to quantify the electroactive void fraction from a top-view perspective. Images obtained from the top view were sampled at 1-s intervals, and the images of the three different metasurfaces were extracted at equal time intervals, resulting in a total of 29 samples. The changes in the normalized values of void fraction were plotted against the normalized sampling time data with a total of 29 samples.

Frequency Selective Surface Simulation: Electromagnetic simulations were conducted using the finite-element method (FEM) implemented in COMSOL Multiphysics (RF Module). Floquet periodic boundary conditions were applied at the lateral boundaries of the unit cell to model an infinitely periodic surface. Floquet ports were defined on the top and bottom boundaries to excite and capture the reflection and transmission characteristics, respectively. Perfectly matched layers (PMLs) were positioned beyond these ports to effectively absorb outgoing waves and prevent unwanted reflections. To ensure accuracy, the maximum mesh element size was set smaller than one-fifteenth of the wavelength at the highest simulated frequency. Due to the extremely thin nature of the MXene and Nafion layers, a transition boundary condition (thin-layer approximation) was used, incorporating the effective dielectric and conductive properties of MXene and Nafion into the simulation.

## **Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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## **Conflict of Interest**

The authors declare no conflict of interest.

#### **Author Contributions**

M.M. and J.K. contributed equally to this work. S.J. and G.K. fabricated actively morphing surfaces and took movie files. M.J.L. performed the FSS simulations. S.N., J.S.K., V.H.N., M.G., H.Y., and D.S. discussed all data and helped to interpret experimental results. Z.U. performed the DFT calculation. C.W.A and Y.G. prepared MXene precursors, discussed the results, and wrote the paper. I-K.O. supervised the research at all stages, led all groups, and wrote the paper.

## **Data Availability Statement**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

## Keywords

amorphous MOFs, deformable surfaces, electroactive actuation, MXenes, parasite protection

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- [1] A. VahidMohammadi, J. Rosen, Y. Gogotsi, *Science* 2021, 372, abf1581.
- [2] M. R. Lukatskaya, S. Kota, Z. Lin, M. Q. Zhao, N. Shpigel, M. D. Levi, J. Halim, P. L. Taberna, M. W. Barsoum, P. Simon, Y. Gogotsi, *Nat. Energy* 2017, 2, 17105.
- [3] X. Pan, X. Yang, M. Yu, X. Lu, H. Kang, M. Q. Yang, Q. Qian, X. Zhao, S. Liang, Z. Bian, *Nat. Commun.* **2023**, *14*, 4183.
- [4] W. Eom, H. Shin, R. B. Ambade, S. H. Lee, K. H. Lee, D. J. Kang, T. H. Han, Nat. Commun. 2020, 11, 2825.
- [5] A. Iqbal, F. Shahzad, K. Hantanasirisakul, M. K. Kim, J. Kwon, J. Hong, H. Kim, D. Kim, Y. Gogotsi, C. M. Koo, *Science* 2020, 369, 446.
- [6] F. Shahzad, M. Alhabeb, C. B. Hatter, B. Anasori, S. M. Hong, C. M. Koo, Y. Gogotsi, *Science* **2016**, *353*, 1137.
- [7] A. Sarycheva, A. Polemi, Y. Liu, K. Dandekar, B. Anasori, Y. Gogotsi, Sci. Adv. 2018, 4, aau0920.
- [8] S. Pinilla, J. Coelho, K. Li, J. Liu, V. Nicolosi, Nat. Rev. Mater. 2022, 7, 717.
- [9] Y. Cai, J. Shen, C. W. Yang, Y. Wan, H. L. Tang, A. A. Aljarb, C. Chen, J. H. Fu, X. Wei, K. W. Huang, Y. Han, *Sci. Adv.* **2020**, *6*, abb5367.
- [10] S. Umrao, R. Tabassian, J. Kim, V. H. Nguyen, Q. Zhou, S. Nam, I. K. Oh, *Sci. Robot.* **2019**, *4*, aaw7797.
- [11] C. Ye, M. Wang, J. Min, R. Y. Tay, H. Lukas, J. R. Sempionatto, J. Li, C. Xu, W. Gao, Nat. Nanotechnol. 2024, 19, 330.
- [12] P. Srimuk, X. Su, J. Yoon, D. Aurbach, V. Presser, Nat. Rev. Mater. 2020, 5, 517.
- [13] A. Agresti, A. Pazniak, S. Pescetelli, A. Di Vito, D. Rossi, A. Pecchia, M. Auf der Maur, A. Liedl, R. Larciprete, D. V. Kuznetsov, D. Saranin, *Nat. Mater.* 2019, *18*, 1228.
- [14] K. Huang, Z. Li, J. Lin, G. Han, P. Huang, *Chem. Soc. Rev.* **2018**, *47*, 5109.
- [15] J. Zhang, Y. Zhao, X. Guo, C. Chen, C. L. Dong, R. S. Liu, C. P. Han, Y. Li, Y. Gogotsi, G. Wang, *Nat. Catal.* **2018**, *1*, 985.
- [16] A. Morales-Garcia, F. Calle-Vallejo, F. Illas, ACS Catal. 2020, 10, 13487.
- [17] Y. Song, R. Y. Tay, J. Li, C. Xu, J. Min, E. Shirzaei Sani, G. Kim, W. Heng, I. Kim, W. Gao, *Sci. Adv.* **2023**, *9*, adi6492.
- [18] M. Shekhirev, J. Busa, C. E. Shuck, A. Torres, S. Bagheri, A. Sinitskii, Y. Gogotsi, ACS Nano 2022, 16, 13695.
- [19] D. Li, W. Zheng, S. M. Gali, K. Sobczak, M. Horák, J. Polčák, N. Lopatik, Z. Li, J. Zhang, D. Sabaghi, S. Zhou, *Nat. Mater.* 2024, 23, 1085.
- [20] F. Cao, Y. Zhang, H. Wang, K. Khan, A. K. Tareen, W. Qian, H. Zhang, H. Ågren, *Adv. Mater.* **2022**, *34*, 2107554.
- [21] C. J. Zhang, S. Pinilla, N. McEvoy, C. P. Cullen, B. Anasori, E. Long, S. H. Park, A. Seral-Ascaso, A. Shmeliov, D. Krishnan, C. Morant, *Chem. Mater.* 2017, *29*, 4848.
- [22] W. Li, T. Zhou, Z. Zhang, L. Li, W. Lian, Y. Wang, J. Lu, J. Yan, H. Wang, L. Wei, Q. Cheng, *Science* **2024**, *385*, 62.
- [23] V. Kamysbayev, A. S. Filatov, H. Hu, X. Rui, F. Lagunas, D. Wang, R. F. Klie, D. V. Talapin, *Science* **2020**, *369*, 979.
- [24] H. Yun, Y. Chae, E. Kim, H. K. Kim, S. Jang, M. H. Baik, C. W. Ahn, Y. Lee, Adv. Funct. Mater. 2022, 32, 2203296.

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- [25] C. Zhou, D. Wang, F. Lagunas, B. Atterberry, M. Lei, H. Hu, Z. Zhou, A. S. Filatov, D. E. Jiang, A. J. Rossini, R. F. Klie, *Nat. Chem.* **2023**, *15*, 1722.
- [26] Y. Cheng, X. Li, Y. Qin, Y. Fang, G. Liu, Z. Wang, J. Matz, P. Dong, J. Shen, M. Ye, *Sci. Adv.* **2021**, *7*, abj1663.
- [27] J. Yang, M. Li, S. Fang, Y. Wang, H. He, C. Wang, Z. Zhang, B. Yuan, L. Jiang, R. H. Baughman, Q. Cheng, *Science* **2024**, *383*, 771.
- [28] Y. Liu, W. Zou, N. Zhao, J. Xu, Nat. Commun. 2023, 14, 5342.
- [29] X. Zhu, Y. Zhang, Z. Man, W. Lu, W. Chen, J. Xu, N. Bao, W. Chen, G. Wu, Adv. Mater. 2023, 35, 2307186.
- [30] H. Yang, G. X. Zhang, H. J. Zhou, Y. Y. Sun, H. Pang, Energy Mater. Adv. 2023, 4, 0033.
- [31] C. Liu, Y. Bai, W. Li, F. Yang, G. Zhang, H. Pang, Angew. Chem., Int. Ed. 2022, 61, 202116282.
- [32] T. Ramachandran, F. Hamed, Y. A. Kumar, R. K. Raji, H. H. Hegazy, J. Energy Storage 2023, 73, 109299.
- [33] R. Ramachandran, K. Rajavel, W. Xuan, D. Lin, F. Wang, Ceram. Int. 2018, 44, 14425.
- [34] T. D. Bennett, A. K. Cheetham, Acc. Chem. Res. 2014, 47, 1555.
- [35] J. Yan, C. E. Ren, K. Maleski, C. B. Hatter, B. Anasori, P. Urbankowski, A. Sarycheva, Y. Gogotsi, *Adv. Funct. Mater.* **2023**, *27*, 1701264.

- [36] Y. T. Liu, P. Zhang, N. Sun, B. Anasori, Q. Z. Zhu, H. Liu, Y. Gogotsi, B. Xu, Adv. Mater. 2018, 30, 1707334.
- [37] M. Garai, M. Mahato, S. Nam, E. Kim, D. Seo, Y. Lee, V. H. Nguyen, S. Oh, P. Sambyal, H. Yoo, A. K. Taseer, *Adv. Funct. Mater.* **2023**, *33*, 2212252.
- [38] A. Li, S. Singh, D. Sievenpiper, Nanophotonics 2018, 7, 989.
- [39] X. Huang, X. Zhang, Z. Hu, M. Aqeeli, A. Alburaikan, Microw. Antennas Propag. 2015, 9, 307.
- [40] X. Fan, Z. Pan, S. Chen, Y. Li, Z. Zhao, Y. Xin, T. Pan, Soft Sci. 2021, 1, 13.
- [41] Y. Saifullah, Y. He, A. Boag, G. Yang, F. Xu, Adv. Sci. 2022, 9, 2203747.
- [42] W. M. Zhu, A. Q. Liu, X. M. Zhang, D. P. Tsai, T. Bourouina, J. H. Teng, X. H. Zhang, H. C. Guo, H. Tanoto, T. Mei, *Adv. Mater.* **2011**, *23*, 1792.
- [43] J. Choi, J. Ahn, J. Kim, Y. Kim, J. Lee, I. Oh, *Small* **2016**, *12*, 1840.
- [44] M. Li, L. Shen, L. Jing, S. Xu, B. Zheng, X. Lin, Y. Yang, Z. Wang, H.
- Chen, Adv. Sci. 2019, 6, 1901434.
- [45] Y. Zheng, K. Chen, W. Yang, L. Wu, K. Qu, J. Zhao, T. Jiang, Y. Feng, Adv. Funct. Mater. 2022, 32, 2107699.
- [46] S. Nam, M. Mahato, K. Matthews, R. W. Lord, Y. Lee, P. Thangasamy, C. W. Ahn, Y. Gogotsi, I. K. Oh, *Adv. Funct. Mater.* **2023**, *33*, 2210702.
- [47] M. Mahato, M. Garai, V. H. Nguyen, S. Oh, S. Nam, X. Zeng, H. Yoo, R. Tabassian, I. K. Oh, *Sci. Adv.* **2023**, *9*, adk9752.