

# The Need for Smart Materials in an Expanding Smart World: MXene-Based Wearable Electronics and Their Advantageous Applications

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including mobile healthcare sensing and monitoring, humanmachine interfaces, portable energy storage and harvesting, and more. Because of their enriched surface functionalities, large surface area, and high electrical conductivity, transition metal nitrides and carbides (also known as MXenes) have recently come to be extensively considered as a group of functioning twodimensional nanomaterials as well as exceptional fundamental elements for forming flexible electronics devices. This Review discusses the most recent advancements that have been made in the field of MXene-enabled flexible electronics for wearable electronics. The emphasis is placed on extensively established nonstructural features in order to highlight some MXene-enabled electrical devices that were constructed on a nanometric scale.



These attributes include devices configured in three dimensions: printed materials, bioinspired structures, and textile and planar substrates. In addition, sample applications in electromagnetic interference (EMI) shielding, energy, healthcare, and humanoid control of machinery illustrate the exceptional development of these nanodevices. The increasing potential of MXene nanoparticles as a new area in next-generation wearable electronic technologies is projected in this Review. The design challenges associated with these electronic devices are also discussed, and possible solutions are presented.

## **1. INTRODUCTION**

As a result of recent developments in intelligent electronics and wearable technologies, flexible and miniaturized equipment has gained an exponential amount of interest.<sup>1–3</sup> These devices introduce an innovative paradigm to the realm of consumer electronics, which contributes to this trend. These advancements in wearable technology have enabled significant utilization of flexible electronics, which may now be used in applications such as self-powered gadgets,<sup>4,5</sup> intelligent health and household devices,<sup>6</sup> soft robotics,<sup>7</sup> smart sensors,<sup>8</sup> human–machine interfaces,<sup>9</sup> and many other areas. It is rather remarkable that the present development of flexible electronics may be achieved through their inherent properties, such as the fact that they are lightweight, affordable, and straightforward to mount on skin.

Wearable electronics will one day become commonplace in the field of healthcare as a result of the natural interactions that occur between the human body and electronics. These interactions allow for the measurement and collection of information from the human body. Thus, the substances should be conformable to human skin, robust, and sustainable for noninvasive surveillance.<sup>10,11</sup> This is especially desired in electronic sensing devices in which human presence is anticipated and thus the materials should meet all three of these requirements. In recent years, significant progress has been achieved in the field of wearable electronics by combining a wide variety of nanomaterials, both inorganic and organic, for use in operational applications.<sup>12,13</sup> Concerns regarding biocompatibility and gadget effectiveness are continually being taken into consideration, despite the fact that great advancement has been achieved in the field of nanomaterial electronics to work smoothly and reliably over the human body. It is interesting to note that more advanced devices have

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Figure 1. Upcoming and developing applications of MXene electronics, including flexible sensing, wearable energy storage, EMI shielding, and HMI.

investigated the utilization of a variety of nanomaterials in great depth.

The fabrication of flexible electronics for a wide variety of applications has seen significant adoption of two-dimensional (2D) nanomaterials. When it comes to the design and fabrication of flexible electronics, one category of 2D substances that has garnered a significant amount of attention is recently discovered MXenes. 2D MXenes constitute the layered transition metallic nitrides or carbides that are formed from  $M_{n+1}AX_n$  (MAX) phase precursors. M represents an early metallic element, A denotes an element coming from group IVA or IIIA of the periodic table, X indicates nitrogen or carbon, and the numerical value of n can range from 1 to 3.<sup>14,15</sup> Initially, Naguib et al. produced  $Ti_3C_2T_x$  using Al layer selective etching assisted by hydrofluoric acid (HF) derived from the MAX phase precursor of Ti<sub>3</sub>AlC<sub>2</sub>. In the formula for MXene, the symbol " $T_x$ " refers to a variety of surfaceterminating functional groups (including -Cl, -F, -OH, and -O) that can be created by a variety of synthetic procedures. It is possible to adjust the characteristics of MXenes by altering either the M or X components as well as the surface functions.<sup>16-18</sup> MXenes offer outstanding optoelectronic and electrochemical capabilities as well as strong conductivity for electricity, and their surface functionality can potentially be tuned. MXenes have found use in a variety of electronic systems due, in particular, to their increased electrical conductivity and their ability to feature surface functional groups that can be tuned. These applications include healthcare systems,<sup>19,20</sup> improved flexible electronics,<sup>21,22</sup> and sectors connected to wearable energy.<sup>23,24</sup> The enriched structural characteristics of MXene make it possible to fabricate electronics from bioinspired sources<sup>25</sup> and integrate

those devices into planar or textile substrates for wearable electronics.<sup>26,27</sup> This allows for the realization of different geometries, including printed electronic devices, intelligent textile-based electronics, and 3D-configured systems (for example, hydrogels and aerogels) for practical uses.<sup>28–30</sup> MXene is one of the most interesting substances for wearable applications because of its better hybridization and coupling to other substances at the nanoscale level.<sup>31</sup> It is anticipated that the next few years will see enormous progress in the efficient production of MXene-based nanoelectronic devices and the processing of these materials. For the engineering of MXene-based functional electronic devices, however, challenges regarding oxidation, aggregation, scalable synthesis in certain solvent systems, and poor mechanical characteristics must be addressed.

This Review provides a summary of the most recent developments in nanoengineered flexible electronics based on MXene that are applicable to wearable applications. In the first part of this Review, the recent developments in functional nanostructures formed from MXene are covered, along with a discussion of the various production methods for these structures. Elucidation is also provided for a number of important morphological characteristics that are shown by these nanostructures. In the next section, several system-level applications based on MXene nanodevices will be illustrated. Some examples of these applications include healthcare, electromagnetic interference (EMI) shielding, energy harvesting and storage, and human-machine interaction (HMI). The important uses of MXene-based flexible wearable devices are illustrated in Figure 1. The assessment comes to a close by discussing a number of obstacles and picturing new prospects

for electronics that are wearable and flexible and that are based on MXene.

## 2. ELECTRICAL AND MECHANICAL PROPERTIES OF MXENE

According to the findings of the experiments, the Young's modulus of each single-layered  $Ti_3C_2T_x$  was 330 ± 30 GPa. This value is significantly higher than the values for 2D reduced graphene oxide (rGO) and GO.32 However, the tensile strength of a single sheet of  $Ti_3C_2T_x$  was measured to be 17.3  $\pm$  1.6 GPa at the point of fracture. Density functional theory (DFT) has been used to make some theoretical predictions, and these results support those predictions.<sup>3</sup> Additionally, the transition components that are found throughout the MXene structure have a significant impact on the mechanical characteristics. Additionally, the intrinsic tensile characteristics of MXene may be adjusted further by modifying the surface of the material. For instance, the Young's modulus of a single-layered Nb-based Nb<sub>4</sub>C<sub>3</sub>T MXene was measured at  $386 \pm 14$  GPa, which is more than the Young's modulus of a single-layered Ti-based  $Ti_3C_2T_x$  MXene.<sup>34</sup>

Simulations using molecular dynamics (MD) were used to investigate the influence of the thickness of the layer on the mechanical characteristics of 2D  $\text{Ti9}_{n+1}\text{C}_n$ . The results of these simulations showed that reducing the thickness of the layer led to a spike in the Young's modulus regarding MXenes. It was discovered that the Young's moduli of  $\text{Ti}_4\text{C}_3$ ,  $\text{Ti}_3\text{C}_2$ , and  $\text{Ti}_2\text{C}$ , respectively, were 534, 502, and 597 GPa. This indicated that the thinnest  $\text{Ti}_2\text{C}$  carbide that was formed of three atomic layers had the greatest Young's modulus.<sup>35</sup> In a separate set of experiments, the researchers found that increasing the thickness of a single-layered MXene resulted in a lower value for the material's Young's modulus.<sup>36</sup>

MXene possesses superior electrical conductivities that are higher in comparison to those of other solution-processed 2D substances.<sup>37</sup> Surprisingly, the electrical conductivity levels of MXene might reach as high as 20 000 S cm<sup>-1</sup> with the incorporation of a variety of X and M elements and the utilization of a controlled manufacturing procedure.<sup>38</sup> The significant electrical conductivity of individual Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> nanosheets with a single layer is demonstrated by a value of around 4600 S cm<sup>-1.39</sup> The M layer is one of the most important components that plays a role in determining the electrical characteristics of MXene. Therefore, by preserving a careful equilibrium between the temperature and the reaction time of etchants, one is able to change the exterior transition layer that contains 2D carbide and thereby control the electrical characteristics of the material, which range from metallic to semiconductor. During the production of MXene, the addition of a variety of surface termination groups, etchants, and flaws can also have a significant influence on its capacity for electrical conductivity. Investigations, for instance, have shown that the conductive property of HF-etched powdered  $Ti_3C_2T_x$  is approximately 1000 S cm<sup>-1</sup>, which is approximately fivetimes weaker compared to that of Ti<sub>3</sub>C<sub>2</sub> nanosheets containing few flaws, which have a conductivity of 6500 S cm<sup>-1, 31,40</sup>

#### 3. **BIOCOMPATIBILITY**

When integrating wearable devices based on MXene into the human body, considerable consideration must be given to allergic reactions, toxic behaviors, and biocompatibility tests.<sup>41</sup> When designing an experiment to be conducted on humans,

certain criteria need to be taken into consideration. The interface must be biocompatible for ongoing tracking of biosignals (EEG, EMG, ECG, etc.) with a good signal-to-noise ratio in order to allow for the long-term integration of MXenebased wearable devices. In the event that this does not occur, it may gradually raise the impedance of the body caused by the immunological response.<sup>42</sup> Recent research has shown that there is potential for MXene to be used in diagnostic nanomedicine, image processing, biosensing, as an antibacterial agent, and in the treatment of cancer.<sup>41</sup> MXenes have been described as multipurpose theragnostic agents for effective magnetic resonance (MR) and photoacoustic (PA) dualmodality imaging-guided photothermal treatment (PTT) against cancer because of their great biocompatibility, which is a crucial route.<sup>43,44</sup> This is possible because of the MXenes' ability to inhibit the formation of reactive oxygen species. MXenes make it easier to manufacture mediator-free biosensors by allowing for direct electron transfer between the bioreceptors and the electrode and by functioning as biomolecule immobilization frameworks to preserve active proteins. This makes it possible for MXenes to be used in the fabrication of mediator-free biosensors. Studies using spectroscopy and electrochemistry have shown that the TiO<sub>2</sub>-Ti<sub>3</sub>C<sub>2</sub> nanocomposite is an effective immobilization matrix. This matrix shows compatibility with redox proteins, and it also provides greater durability and protein bioactivity.<sup>45</sup> These fascinating biological features, along with the multifunctionalities of 2D MXenes, open the way for a new avenue of diverse sensing modalities that may be used in wearable biosensing applications.

MXene materials have gained recognition for their excellent biocompatibility properties, making them suitable for integration into wearable devices for various applications.<sup>46,47</sup> MXene materials in the context of wearable devices have key biocompatibility properties, including low cytotoxicity, inertness, biostability, electrical conductivity, mechanical strength, compatibility with biological fluids, skin-friendly, biocompatible coatings, and reduced inflammatory response. When in contact with living cells, MXene materials do not harm or interfere with normal cellular functions. This is crucial for wearable devices that may have a prolonged skin contact. MXenes are chemically inert, which means that they do not easily react with surrounding biological tissues or fluids. This chemical stability is essential to prevent adverse reactions when MXene-based devices are used in or within the body. MXene materials are biostable, meaning they do not degrade over time upon exposure to biological environments. This characteristic ensures the long-term functionality of wearable devices. MXenes are good conductors of electricity. In the context of wearables, this property allows for the development of biocompatible sensors and electrodes, which can monitor physiological parameters without causing harm to the skin or underlying tissues. MXene materials have good mechanical properties, providing durability and resistance to wear and tear. This is important for wearable devices that may be subjected to daily use and physical stress. MXenes are compatible with various biological fluids such as blood, sweat, and interstitial fluids. This compatibility ensures that MXene-based sensors and devices function effectively when measuring biomarkers and health parameters. MXene materials are skin-friendly, meaning they do not cause skin irritation, redness, or allergic reactions when in contact with the skin. This property is essential for the comfortable and safe use of wearable devices.

MXenes can be used as coatings on other materials to enhance their biocompatibility. For instance, MXene coatings on metallic components of devices can make them more suitable for medical and biological applications. MXenes are known for their ability to reduce the inflammatory response in comparison to some other materials, which is advantageous for long-term use in the body.<sup>48–54</sup>

It is worth noting that while MXene materials offer these promising biocompatibility properties, the specific application and design of the wearable device, as well as the surface modifications and functionalizations, will play a crucial role in determining the overall biocompatibility and performance of the device. Continuous research and development are ongoing to optimize MXene-based materials for specific wearable device applications in the fields of healthcare, sports, and beyond.

### 4. MXENE-BASED WEARABLE ELECTRONICS

4.1. Textile and Planar Structures. Because of their exceptional adaptability for skin-wearable and mountable electronics, innovative fabrics have witnessed an exponential growth in the variety of applications they may be used for. Textiles were originally designed to be aesthetic, daily consumables; nevertheless, they have since evolved into more complex and demanding structures for today's wearable electronics. The development of nanomaterials such as carbon nanotubes (CNTs),<sup>55</sup> MXene,<sup>56</sup> graphene,<sup>57</sup> metallic nanoparticles and nanowires,<sup>58</sup> conducting polymeric materials,<sup>5</sup> and so on has made it feasible for textiles to undergo these groundbreaking developments and alterations. Electronic devices that are constructed on textiles have found widespread usage in a variety of contexts, including but not limited to thermal management,<sup>54,60,61</sup> flexible sensing<sup>62</sup> and displays,<sup>6</sup> real-time healthcare surveillance,<sup>64,65</sup> biomedical treatment,<sup>66</sup> and others. In spite of the fact that a number of nanostructures have made it possible to create smart fabrics for wearable electronics, innovative 2D MXene has lately garnered increasing interest. Insulating textiles can receive functionality thanks to the functional groups of the MXene surface, which may interact directly with the fiber. In addition, MXene is a great choice for the fabrication of innovative textiles due to the simplicity with which it can be processed as an alternative and its mechanical flexibility.<sup>19</sup> It offers a variety of material structures and morphologies that are suitable for use in wearable applications.

MXene may be processed in a variety of ways; for instance, it can be combined with a variety of spinning dopes in order to generate simply wet-spun or electrospun MXene fibers, or it can be combined as a second dopant in a spinning dope in order to generate intelligent composite fibers. MXene possesses varied processability.<sup>67</sup> Because MXene is able to disperse in an extensive range of solvents, such as dimethyl sulfoxide (DMSO), water, N-methyl-2-pyrrolidone (NMP), and dimethylformamide (DMF), it possesses high treatment possibilities. This potential may be because MXene can dissolve in many solvents. Particularly, numerous coating processes, including dip-coating, spray-coating, and others like them, have been intensively researched because of their simplicity of scalability and processing.<sup>37</sup> This is one of the various methods of processing used for MXene smart textiles. Additionally, the many functional groups on the surface that are present on MXene, in addition to the rheological characteristics of MXene suspensions that can be adjusted,

make it easier to prepare MXene for use in a variety of printing processes, most notably inkjet printing, screen printing, and 3D printing. Because an unsuitable flake size can hamper the manufacturing of smart textiles and lead to a decline in material performance, ensuring that the MXene flake diameter is maintained at an appropriate level is an incredibly important element.<sup>67</sup> For example, tiny flakes of MXene nanosheets can cover the center and interstitches of yarns and fibers in a conformal manner; nevertheless, this reduces the electrical conductivity of the material, which is essential in numerous electronic applications. However, covering fibers with big flakes of MXene results in increased electrical conductivity, but the coating has poor adhesion to fiber surfaces and reduces the fiber's mechanical flexibility.<sup>68</sup> As a result, the size of the MXene flakes must be tuned in accordance with the many applications that are sought, particularly for supercapacitor electrodes (SCs) that are based on textiles. In general, in order to create an effective MXene-mediated textile interface, it is necessary to functionalize the surfaces of the textiles, and one should also synthesize, modify, and refine MXene relative to the applications for which it will be used for.

Wet-spinning, electrospinning, and coating are three widely examined textile processing processes that are currently being researched to generate MXene-mediated yarns, fibers, and textiles.<sup>69</sup> In order for MXene to perform its role as a codopant in numerous spinning solutions that contain polymers, it has been utilized as an efficient nanofiller and introduced into these solutions. The outcome is nanofibers that have several functions in contrast to pure polymeric spun fibers. For instance, Levitt and his colleagues have developed a novel method that utilizes both wet-spinning and electrospinning to manufacture MXene-infiltrated nylon nanoyarns.<sup>70</sup> This method combines the advantages of both processes. This combination spinning method made it possible to disperse MXene nanoflakes across the full cross sections of nanoyarns, and it attained the all-time greatest MXene loading (90%) because of the spinning procedure it underwent. The size of the MXene flake has an effect on the process of fiber spinning, and it exhibits considerable morphological variations in the microstructure. At a relatively low MXene level ( $10 \text{ mg mL}^{-1}$ ), the nanoyarns spun using a tiny flake diameter (220 nm) displayed a circular microstructure. However, greater MXene flakes (850 nm) revealed randomly oriented fibers featuring entangled MXene flakes. In addition, the electrospun nanothreads that were created from tiny flakes (220 nm at 10 mg mL<sup>-1</sup>) displayed mechanical integrity, and there were no discernible morphological alterations seen after the yarn was knotted. In conclusion, the nanoyarns show considerable potential for use as a SC. In a separate piece of research, Levitt and colleagues inserted delaminated MXene flakes into an electrospinning dope of polyacrylonitrile (PAN). The fibers produced by the electrospinning were then carbonized in an argon environment at several temperatures. Because of the tiny diameter of the fiber and high electrical conductivity, the incorporation of carbon into nanofiber substances led to a considerable improvement in the electrochemical characteristics of the substance. The production of many MXenereinforced electrospun nanofibers for SCs has shown that there are great possibilities for exploration in the fiber-based platform for energy storage uses.

Wet spinning is a method that may be used to generate plain MXene fibers or MXene-based hybrid fibers. This method is made possible by the exceptional dispersibility of MXene in a variety of polar solvents as well as its pseudoelasticity, liquid crystalline (LC) behavior, and adjustable rheological characteristics. For instance, Zhang et al. created a pure wet-spun MXene fiber for use in flexible heating elements and a fiber-shaped SC for thermal applications by imbuing  $Ti_3C_2$  MXene with LC behavior.<sup>71</sup> These fibers are intended to be used in thermal electronics. Multiple factors, including MXene flake concentration and size, in addition to needle gauges and different types of coagulation baths, were investigated throughout the fiber creation process. For example, fibers generated from an acetic acid bath comprising smaller  $Ti_3C_2$  flakes displayed a porous microstructural morphology, while fibers generated from a chitosan coagulant solution comprising larger  $Ti_3C_2$  flakes demonstrated a tightly packed microstructure.

Shin et al. devised an efficient two-way technique to manufacture a robust mechanical Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene fiber using a flexible MXene gel as part of their research.<sup>72</sup> Initially, a highly reinforced MXene gel was generated from an MXene dispersion by strengthening the gel framework through electrostatic interactions between the sheets of MXene in a basic solution. After that, the MXene solution was subsequently wet-spun throughout a coagulation bath to form gel fibers, which resulted in highly oriented fibers that were assigned from the tight gel web. In addition to this, the gel-spun fibers demonstrated a capacity to endure significant shear stress while the mechanical drawing was being carried out. The small-angle X-ray scattering (SAXS) and morphological investigations revealed a strongly compacted lamellar morphology and an anisotropic streak pattern throughout the material as the fiber draw ratio increased from 1 to 3. These findings demonstrated the presence of microvoids that were the dimensions of the needles within the MXene fibers. The production of MXene fiber with a denser structure was confirmed by further investigation of the porosity and density.<sup>73</sup> The resulting highly aligned fibers showed greatly improved characteristics, including electrical conductivity (12 504 S cm<sup>-1</sup>), Young's modulus (122 GPa), and tensile strength (344 MPa).

MXene-derived smart-textile electronic devices are still an active area of study, and scientists are always looking for novel methods to utilize the one-of-a-kind features of MXenes for applications in the field of wearable technology. These technological breakthroughs have the potential to bring about a revolution in the field of smart textiles, making it possible to create wearable electronic devices that are extremely useful, pleasant, and multifunctional.

4.2. MXene Hydrogel. Because of their unique qualities, such as their extensibility, transparency, biocompatibility, and self-adhesion, hydrogels have garnered a significant amount of interest in recent years.<sup>74</sup> They are able to transfer electrical impulses, which opens up the possibility of applications in areas such as flexible electronics. MXene is a good candidate for wearable electronics because of its biocompatibility. Nevertheless, durability has been shown to be a limiting concern for applications based on MXene, and it has been demonstrated that synthesizing MXene into hydrogels considerably improves the stability of these applications. Because of their one-of-a-kind and intricate gel structure as well as their distinct method of gelation, MXene hydrogels call for extensive investigation and innovation at the nanoscale level.<sup>75</sup> Although there has been a significant amount of research conducted on the use of MXene-based composites in

electronics, the synthesis techniques and utilization of MXenebased hydrogels for wearable electronics are still rather uncommon. Therefore, in order to allow the effective fabrication of MXene hydrogel electronics, the design techniques, production techniques, and potential uses of MXene hydrogels for wearable and flexible electronics are described in this section.

4.2.1. Fabrication of MXene Hydrogels. As a result of the powerful van der Waals attraction between interlayers, MXene nanosheets invariably have a tendency to restack and polymerize; thus, it is challenging to produce hydrogels from MXene on its own.<sup>76</sup> Nevertheless, due to its vast flexibility, good hydrophilicity, and large number of surface terminations, MXene is sensitive to combination with a wide range of different substances, which can result in the production of a wide variety of hybrids. As a consequence of this, it is frequently essential in the process of manufacturing MXenebased hydrogels to include an additional component (i.e., a cross-linker) throughout the hydrogel framework. This is accomplished in order to counteract the hydrophilicity of MXenes and keep the 3D assembly of the 2D nanosheets intact. In addition, MXenes always interact with oxygen that is present in the atmosphere, which restricts the applications for which they may be used. It has been observed that certain chemical and physical surface engineering procedures can shield MXenes against the effects of oxidation.<sup>77</sup> The strong electronegativity and reduced functioning of the atoms on the surface of MXene enable organic ligands to release protons and attach to the MXene surface during the production of the MXene hydrogels. As a result, the oxidation of MXene does not need to take place.<sup>7</sup>

4.2.1.1. Inorganic MXene Nanocomposite Hydrogels Assisted by Materials. The inadequate number of available cross-linking spots on the MXene surface limited its functionality. On the other hand, GO, which is a 2D carbonbased substance, shows a specific surface area, excellent conductivity, and mechanical characteristics, which allowed for the creation of interactions that utilized the MXene nanosheet surface as an agent for gelling.<sup>79</sup> As a result, the phenomenon of self-stacking that occurs between MXene nanosheets was drastically minimized and the role that MXene plays in the hydrogel was improved.

The first report of a 3D macroscopic hydrogel made of rGO/MXene was presented by Xu et al., and it was done by using an organic-free self-convergence approach. In this process, GO was converted to rGO through the use of  $Ti_3C_2T_x$  as a reducing agent under mild circumstances.<sup>79</sup> This resulted in the elimination of a number of water-friendly oxygen-containing groups, as well as a strengthening of the  $\pi$ conjugated structure and hydrophobic properties of rGO, which ultimately made it possible to assemble the rGO/ MXene 3D skeleton. The team led by Shang suggested developing 3D hydrogels based on MXene and assembling them utilizing ethylenediamine (EDA) and GO, employing an approach of self-assembly.<sup>80</sup> While  $Ti_3C_2T_x$  is responsible for inducing the reduction of GO, EDA is responsible for releasing the epoxy rings that surround GO flakes, which allows for the promotion of the creation of oxygen suspension bonds. After that,  $Ti_3C_2T_x$  was linked to these dangling bonds in order to generate the MXene-rGO heterostructures. These structures were then changed into hydrogels via van der Waals forces that were present between layers of heterogeneous nanosheets. The submicrometer level all the way up to several micrometers was



Figure 2. (a) MXene hydrogel and (b) its uses in human healthcare surveillance. Figure taken with permission from 97. Copyright 2022 Wiley-VCH.

the size range that the interconnected, 3D porous, and welldefined structure of the MXene-based hydrogel emerged in.

4.2.1.2. Polymer-Assisted MXene Nanocomposite Hydrogels. Because this network can be widely swelled by water, which provides great adaptability in electrical devices, the insertion of MXene into polymeric hydrogel frameworks has lately gained a significant amount of attention. By employing MXene surface groups to interact with different types of polymers (such as poly(3,4-ethylenedioxythiophene):poly-(styrenesulfonate) (PEDOT:PSS), polyvinylpyrrolidone, poly-(vinyl alcohol) (PVA), chitosan, cellulose, etc.),<sup>81-84</sup> one may create hydrogels through the process of polymerization reactions. Contacts between MXene nanosheets and other polymer compounds within the hydrogel network are often caused by the interweaving of polymer chains in addition to hydrogen bonds, covalent bonds, and ionic interactions.

Nicolosi et al. proposed an easy method for 4D printing MXene-based hydrogels that consisted of PEDOT:PSS, MXene, and additives (H<sub>2</sub>SO<sub>4</sub>, sodium L-ascorbate, and DMSO). This method has now been expanded to the MXene family, which also includes Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub>, Mo<sub>2</sub>Ti<sub>2</sub>C<sub>3</sub>T<sub>x</sub>, and Nb<sub>2</sub>CT<sub>x</sub>.<sup>85</sup> Using 3D printing, the combination of inks was initially shaped into a variety of designs. Following that, using self-assembly, the MXene sols were converted to MXene hydrogels. The 4D MXene hydrogels that were developed exhibited better specific capacitance (232.9 F  $g^{-1}$  at 10 V  $s^{-1}$ ) and permitted operation at low temperatures  $(-20 \, ^{\circ}\text{C})$ . Wang et al. created a triboelectric nanogenerator (TENG) that was flexible and stretchy by employing MXene nanosheets and PVA hydrogel capsules as electrodes. They then employed this TENG in portable, self-powered sensors for tracking body motion.86

4.2.1.3. Metal–MXene Nanohybrid Hydrogels. During the process of gelation that was detailed earlier, there was unavoidable oxidation that led to a loss of some of the characteristics of the MXene-based hydrogels that were created.<sup>87</sup> Therefore, in order to mitigate the effects of oxidation and speed up the phase separation of MXenes from water, a more rapid gelation mechanism is required to greatly inhibit the restacking of the MXene nanosheets. This is necessary in order to achieve the goals of both of these processes.<sup>88</sup> In their study, Ye et al. developed a metal-assisted electrogelation approach that could be used to directly create MXene hydrogels with porous architectures and features that could be tuned.<sup>89</sup> This programmable technique allows more comprehensive patterning with more complexity and effective-ness than laser patterning or 3D printing, both of which entail

many-step procedures or have substantial device dependency. It also provides these benefits in a more straightforward manner. In the electrogelation procedure, the metallic cations are liberated by electrolysis, which is followed by a series of electrostatic interactions during which the cations engage with the MXene nanosheets. Yang et al. were the first researchers to report the fast gelation of MXene within aqueous dispersions when it was triggered by divalent metallic ions ( $Fe^{2+}$ ). The significant interactions that took place between the -OH groups on the MXene surface and metallic ions were extremely important in this process. The gelation process makes use of metal ions  $(Fe^{2+})$  as connecting points to facilitate the assembly of MXene nanosheets into a 3D network. This is possible as a result of the metal ions' strong binding power to the –OH groups that are located on the MXene surface.<sup>90</sup> The effective synthesis of the metal-MXene hybrid hydrogel structure by the metal-ion-mediated gelation approach was further validated by the presence of other metal ions, specifically Co<sup>2+</sup>, Al<sup>3+</sup>, Mg<sup>2+</sup>, and Ni<sup>2+</sup>.

4.2.2. MXene Hydrogels for Wearable Sensors. Since hydrogels are a material that is soft and flexible, they may react to an extensive variety of physical and chemical stimuli by producing detectable alterations to their optical, geometric, and electrical characteristics. Because of this, they have found widespread use in the field of wearable electronics. In spite of this, the majority of traditional hydrogel sensors have a very poor sensitivity when subjected to mechanical stimuli (pressure or strain), and they frequently experience signal hysteresis and variations as a result of the viscoelastic qualities they possess.<sup>91</sup> MXene materials play an essential role in the production of conductive hydrogels because they have good electrical conductivity, are mechanically deformable, and have a wide range of functional groups on the surface.<sup>92,93</sup> For example, MXene may react with a hydrogel matrix to improve its electrochemical and mechanical characteristics. As a result, the sensing capabilities and biocompatibility of the hydrogel may be enhanced in a proportional manner. Because of this, it is anticipated that MXene-based hydrogels will be employed in wearable electronic devices, such as smart sensors, electronic skin, and individualized healthcare monitoring equipment.

4.2.2.1. MXene Hydrogels for Pressure Sensors. In general, pressure sensors may be divided into the capacitive, piezoresistive, triboelectric, and piezoelectric pressure sensor categories according to the various signal transformations that they use.<sup>94,95</sup> Because of their high sensitivity, great temperature stability, and quick reaction time, piezoresistive sensors are the basic components of a variety of wearable electronic



Figure 3. Printing method of MXenes bonded with a polyurethane/poly(vinyl alcohol) hydrogel using direct ink writing (DIW) (left) and a schematic representation of temperature sensing (right). Figure taken with permission from 92. Copyright 2022 Springer Nature.

devices. In general, the relative sliding layers and metallic conductivity provide MXenes with a broad tunable resistance spectrum. These properties are highly respected in the field of pressure-sensitive substances and have significant application possibilities.

Conductive hydrogel sensors that are flexible have seen extensive use in the fields of personalized health care and eskin; however, it is still a significant challenge to concurrently acquire extremely accurate sensing (in particular electrophysiological signals) for wearable human-computer associations while also increasing wound healing for aftercare. Therefore, Wan et al. created a degradable, healable, and antimicrobial epidermal sensor for the purpose of sensing weakened physiological signals and mimicking the medical management of wound infections.<sup>96</sup> As can be seen in Figure 2a, the MXene-based hydrogel that was ultimately formed was manufactured by incorporating Ag nanoparticles and MXene into the polymer structure that was constituted of guar gum (GG) and sodium alginate grafted with phenylboronic acid (Alg-PBA). This MXene-based hydrogel is capable of creating a multipurpose epidermal sensor that is adept at sensitively tracking an extensive variety of energetic activities, as well as tiny electrophysiological signals. This can provide essential medical data for rehabilitative training as well as cardiovascular-related disorders (Figure 2b).

4.2.2.2. MXene Hydrogels for Strain Sensors. There is a definite chance that incorporating MXene into hydrogels would increase the effectiveness of the hydrogel strain sensors. This will be accomplished by combining the benefits of MXene and hydrogel, each of which possesses promising electrical characteristics and adaptable mechanical flexibility.<sup>98</sup>

A MXene-based hydrogel (containing antidehydration additives, PVA, MXene ( $Ti_3C_2T_x$ ), and water) was created by Alshareef et al., and it displayed exceptional tensile strain sensitivity with a gauge factor (GF) of 25, which was 10-fold greater than that of the basic hydrogel.<sup>93</sup> Furthermore, the hydrogel possesses extraordinarily high tensile qualities, the ability to self-heal, and excellent compatibility and adhesion to a range of surfaces. These characteristics make the hydrogel a prospective candidate for use in applications such as biosignal surveillance and touch sensing. The research team led by Yu was able to create a strain-sensitive MXene hydrogel (MNH) by substituting ethylene glycol (EG) for water as the medium for dispersion.<sup>99</sup> In comparison to other types of hydrogels, the MNH demonstrates superior resilience to freezing temper-

atures (-40 °C) and an extended period of fluid retention stability (8 days). In addition, this MNH might be designated as a flexible sensor that can measure the biological motions of the human body at extremely low temperatures. It possesses a fairly large strain range (up to 350% strain) as well as a high measurement factor (44.85). Alshareef et al. used  $Ti_3C_2T_x$ MXene as a versatile cross-binding agent that triggers different hydrogels and quickens the gelation process, beginning with multiple monomer or polymeric precursors (for example, PDMA-, PAM-, PAA-, PHEMA-, PANI-, PEGDA-, and PNIPAM–MXene hydrogels). They acted as stretchable electronics.<sup>100</sup>

4.2.2.3. MXene Hydrogels for Chemical Sensors. The demand for novel diagnostic approaches for people's wellbeing is continually expanding, and straightforward-to-use biosensors have attracted a significant amount of scientific attention. Personal diagnostic sensors are able to monitor both sick people and individuals in good health, which enables doctors to make earlier diagnoses and facilitates disease prevention at an earlier stage.<sup>101,102</sup> To this point, a wide variety of hydrogel biosensors have been developed effectively for the concurrent detection of a range of physiological signals or biomarkers through optical and electrical transduction techniques. These biomarkers include glucose, which is related to diabetes;<sup>103</sup> triglycerides, which are related to cardiovascular disease; and heartbeat (pulse), which is related to pulse.<sup>104,105</sup> Due to their high electrical conductivity, one-of-a-kind surface chemistry, and biological compatibility, MXene materials are ideal candidates for the development of novel electrochemical biosensing devices. This demonstrates their enormous promise to be used in mobile and wearable health surveillance and diagnostic tool sets.<sup>106</sup>

The adaptability of flexible sensing devices such as hydrogels continues to provide challenges in terms of cost, complexity of integration, and device manufacture, which impede particular application scenarios. In order to effectively create MXenebonded hydrogel sensors that have exceptional strain and temperature-sensing qualities concurrently, the group led by Huang suggested a 3D-printed direct ink writing process that both has a low cost and is adaptable to a variety of scenarios.<sup>92</sup> Figure 3 illustrates the schematic procedures for the direct ink writing printing of MXene Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> in combination with polyurethane/poly(vinyl alcohol) (PU/PVA) hydrogel as well as the MXene-based hydrogel for temperature sensing. A GF of 5.7 (0–191% strain) and strong response to temperature

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electrochemical

excellent electrochemical efficiency (sensitivity of 600 nA  $\mu$ M<sup>-1</sup> cm<sup>-2</sup>)

resistance to mechanical stress of up to 80 kPa

conductivity up to  $0.4 \text{ S m}^{-1}$ 

host materials

hydrogel hydrogel hydrogel

metal-ion-initiated interaction of

MXene/Fe<sup>2+</sup>

MXene

nanofiller

conductive

hydrogel

electrogelation in a single step was used to facilitate in situ coassembly

Fi<sub>3</sub>C<sub>2</sub>/ sodium alginate (SA) conductive nanofiller conductivity,

a method based on the induction of self-assembly by chitosan

chemical cross-linking method

/IXene/PVA

chitosan

MXene/

dynamic cross-linking

contains an electrode as a supercapacitor (  $\approx 226~F~g^{-1}$  at 1 V  $s^{-1})$ 

sensing

ref 92

application

temperature and strain sensing 108

wearable strain

sensors

exceptional levels of both mechanical strength (reaching up to 1900%) and suppleness, an ideal tensile strength of 190 kPa, exceptional electroconductivity (4 104 S cm<sup>-1</sup>), and responsiveness (a gauge factor of 11)

45 days of deterioration in quality

stretchability of around 200%

cross-linking

conductive nanofiller

96 96

electronic devices

self-powered

spidermic sensor

8

equipment for the storage of energy

(TCR of -5.27% °C<sup>1-</sup> at 0 to 30 °C and -0.84% °C<sup>1-</sup> at 40 to 80 °C) are characteristics of this MXene hybrid hydrogel. Table 1 provides a summary of the synthesis methodologies for MXene-based hydrogels as well as the electrical characteristics of different MXene structural devices.

**4.3. Printed Structures.** Because of the hydrophilicity and extremely negative charge of MXene flakes that have a zeta potential of around -30 mV, MXene is able to undergo homogeneous water dispersion, which has led to a fast rise in MXene-based printed electronics. These qualities of MXene contribute, in addition, to the process of generating a dispersion of colloidal particles in a wide variety of organic and water-based solvents.<sup>109</sup> In addition, recent advancements in MXene ink have shown promising results in the creation of additive-free printed devices. This method is seen as an innovative strategy in terms of its impact on the environment, especially when compared with previous printing techniques that are additive-mediated.<sup>19</sup>

A number of different materials, including metallic nanoparticles, conductive polymeric inks, and carbon-based materials (2D graphene and 1D CNT), have been developed as active substances for printing.<sup>110,111</sup> The formulation of graphene ink is often carried out using solvents based on Nmethyl-2-pyrrolidone (NMP). Large-scale manufacturing of NMP colloidal solutions is greatly hampered by the difficulties related to active components having high boiling temperatures and being unstable in the colloidal solutions.<sup>112</sup> In light of this, investigators examined several methods of producing inks that mitigate the problems described above by using binders, additives, and cosolvents in the formulation process.<sup>11</sup> Researchers have been dissuaded from employing such chemicals due to the severe environmental issues, postprocessing needs, and poor device performance. In recent years, additive-free MXene dispersions in aqueous media have been utilized in the processes of screen printing,<sup>114</sup> inkjet printing,<sup>115,116</sup> and 3D printing<sup>117,118</sup> in order to design electrical devices.

Mei et al. came up with a novel design for the thermoplastic polyurethane  $(TPU)/Ti_3C_2T_x$  (MXene)/MnFe<sub>2</sub>O<sub>4</sub>/multiwalled carbon nanotube (MWCNT) (i.e., TMMM) composites by utilizing a parametric Voronoi structure with a variety of pore widths and porosities during the fused deposition molding (FDM) printing process.<sup>119</sup> The 2D MXene/zerodimensional (0D) MnFe<sub>2</sub>O<sub>4</sub> hybrid fillers were built in the beginning using an electrostatic bonding strategy. After that, the 1D MWCNTs were added as a conductive bridging agent, and this process endowed the nanofillers with different dimensions that were uniformly dispersed to form an excellent conductive network structure in the TPU matrix. The TMMM composites, when used as pressure sensors, displayed an extensive sensing compression range (~89% strain with 12.03 MPa stress), an adjustable gauge factor (GF = 1.33 - 3.73), and excellent durability (6000 s cyclic compression). These properties were particularly suited for tracking human motion such as wrist bending, finger bending, speech recognition, and other similar activities. The sensing processes were also investigated using finite element modeling, and the simulation findings and the pattern of variation of resistance change were found to be in excellent agreement with one another. The printed composite with a thickness of 2.1 mm had an EMI effectiveness rating that averaged 31.2 dB, which indicates that about 99.9242% of electromagnetic waves could be blocked. The technique that was offered not only provided a solution to

	main characteristics	a gauge factor (GF) of $5.7$ (0–191% strain), a response time of 240 s, and stability over 5000 cycles	
	MXene role	conductive nanofiller, cross- linker	
hue and	derivative	hydrogel	
	synthesis method	direct ink writing by 3D printing	
hwdrogel	structure	MXene/PU/ PVA	

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MXene/GG/ Alg-PBA

AgNPs/

Table

lower the weight of electromagnetic shielding substances but also provided an effective method for manufacturing porous 3D printed components with tunable sensing qualities at the macroscopic scale in the direction of wearable electronic devices.

In another novel study, Zhang et al. constructed a wearable self-powered toroidal triboelectric sensor (STTS) featuring a pyramidal structure for self-powered interactions between humans and machines based on a very basic design strategy.<sup>120</sup> Using 3D printing technology, pyramidal arrays are fabricated on MXene/Ecoflex nanocomposites. This creates a comfortable distance between the finger skin and the negatively charged layer, which helps conventional triboelectric-based sensors overcome their space limitations. In addition, the wearability of the sensing system was not compromised by the established pyramidal structure of the nanocomposite, which consisted of flexible conductive fabric electrodes. These electrodes were combined with 3D-printed gloves that were based on a flexible TPU material. The STTS utilizes a flexible and streamlined single-electrode design strategy that is readily worn on the human hand for the purpose of providing a pleasant and natural connection with various machines and gadgets. The high sensitivity (0.088 VkPa1), high peak-to-peak voltage (19.91 V), and broad pressure detection range (0-120)kPa) allow the manufacture of high-quality output signals for the precise detection of different finger movements, displaying significant promise for applications in human-machine interaction uses in next-generation artificial intelligence and interactive devices.

Processing MXene inks for screen printing is made easier by the inks' distinctive rheological behavior and their ability to have a viscosity that can be adjusted. For example, Abdolhosseinzadeh and colleagues developed a sustainable method called "turning trash into treasure" in order to produce MXene ink that did not contain any additives.<sup>121</sup> During the sonication and centrifugation processes, the unetched MAX phase as well as the unexfoliated sediments are obtained by separating the supernatants, which consist of few-layered Ti<sub>3</sub>C<sub>2</sub>Tx nanosheets. A three-roll mill method was used to generate sediment ink from the tiny fraction of delaminated MXene  $(d-Ti_3C_2T_r)$  nanosheets that remained in the decanting supernatant mixture. This was possible since  $d-Ti_3C_2T_r$ provided both electrical conductivity and mechanical integrity. In addition to having a viscosity of 35 Pa s, the ink displayed a shear-thinning tendency and non-Newtonian features. Last but not least, the ink was able to be screenprinted on a wide variety of surfaces because of its excellent resolution and homogeneous printing spatiality.

The combination of MXenes with these printing techniques enables the development of wearable electronic devices such as sensors, energy storage devices, and electronic circuits.<sup>122</sup> MXene-based wearable electronics offer advantages such as light weight, flexibility, and compatibility with various substrates. These devices can be integrated into clothing or accessories or even directly onto the skin, providing a wide range of applications in fields such as healthcare, sports, and smart textiles.

**4.4. Biomimetic or Bioinspired Structures.** Investigators have been successful in fabricating a variety of functional nanostructures by drawing inspiration from nature. These nanostructures have been used to achieve effective skin-inspired microelectronics,<sup>123</sup> soft electronics,<sup>124</sup> actuators,<sup>125</sup> and sensors. Among them, leaf-like structures, polymeric

woods, and bioinspired nacre have all received a significant amount of attention and investigation.<sup>126,127</sup> In particular, structures that resemble nacre are responsible for inducing remarkable mechanical reinforcing effects, which result in the production of substances that have great stiffness and strength. These strengthening effects can be achieved by consciously designing a "brick and mortar" assembly to take advantage of the inherent structural qualities of the material. The scientists used a variety of techniques and different nanomaterials, such as MXene,<sup>128</sup> graphene,<sup>129</sup> and nanoclay,<sup>130</sup> to create an exact replica of this structure.

Recently, bioinspired materials based on 2D MXene have shown promise in their ability to simulate nacre-like nanocomposite frameworks. These materials have also found wide electrical applications, particularly in sensors<sup>131</sup> and EMI shielding.<sup>131</sup> In a recent work, Cao and colleagues incorporated d-Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene into nanofibers of cellulose (CNFs).<sup>132</sup> They then made a multilayer composite paper that resembled nacre by using a technique called vacuum-assisted filtrationassembly. The layered structure that resembles nacre was validated by the cross-sectional morphology of the hybrid paper. This revealed that the function of 2D MXene is "brick", while the role of 1D CNF is "mortar". Because of its biomimetic nacre structure, the  $d-Ti_3C_2T_r/CNF$  paper displayed synergistic mechanical characteristics enhancement. This improvement came about as a result of the paper's structure. The framework of that composite material was formed using 2D MXene nanosheets as "bricks" throughout the composite construction. 1D CNFs, on the other hand, closely bonded MXene nanosheets and enabled stress transport throughout the structure, which ultimately resulted in the dissipation of energy. Overall, the paper's durability and strength were significantly increased as a result of these synergistic reinforcing and toughening effects, and the overall improvement was even greater than the individual improvements brought about by the paper's parent ingredients, MXene and CNFs. In spite of the fact that nacre-like structures are said to be used primarily for mechanical strengthening, the EMI shielding effectiveness is improved by of their presence.<sup>133</sup> The nacre-like laminated framework of d-Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub>/CNF, which is endowed with many internal reflections, enabled the aforementioned composite paper to induce outstanding EMI shielding capabilities. These factors led to the electromagnetic (EM) waves being absorbed and their energy being dissipated, which eventually resulted in a high EMI efficiency.

Structures built on MXene have shown the potential to mimic the appearance of leaves for high-performance applications, such as EMI shielding and programmable actuators. For instance, Cai et al. designed and constructed a natural leaf-inspired intelligent actuator.<sup>134</sup> Initially, am MXene-cellulose (MXCC) ink was created by mixing the two substances together homogeneously. Next, a polycarbonate (PC) (MXCC/PC) ink and bilayered MXCC were developed by filtering the MXCC ink through a PC membrane. The MXCC/PC membrane that was developed displayed a remarkable capacity for successfully imitating many significant leaf microstructures. A typical leaf structure includes a palisade mesophyll, epidermis, cuticle, stomata, and vein skeleton; the combination of all of these features makes the leaf capable of functioning as an efficient photosynthetic medium.<sup>135</sup> Stomata are openings in the cuticle that enable water and carbon dioxide to pass through. The MXCC/PC membrane may achieve a number of different tasks thanks to its leaf-like form,

which was inspired by nature. Nanosheets made of 2D MXene  $(Ti_3C_2T_x)$ , for instance, can collect electric energy and transform it to thermal energy. This is accomplished by imitating the structure of palisade mesophyll. CNFs, on the other hand, are designed to imitate the structure of the vein skeleton. This helps to improve the resilience of the actuator while also facilitating quick shape changes. In addition to this, PC acts as a mimic for the epidermis and stomata, playing the role of allowing water entry as well as extraction through the MXCC. The proposed soft actuator exhibited great responsiveness when exposed to a variety of external stimuli, such as electricity, near-infrared (NIR) light, and humidity. A programmable soft device may make excellent use of a nature-inspired intelligent actuator like this one. The potential is enormous.

In order to construct MXene-based fibers that include a mechanically and functionally important support layer, investigators emulated the hierarchical layered structure of wood.<sup>136</sup> Coaxial wet spinning was used as the method of construction for the biomimetic core-shell MXene/GO fiber that was developed and manufactured. The morphology of the hybrid fiber indicates that, in comparison to a standard MXene/GO fiber, the core-shell MXene/GO fiber features a tightly layered structure of the MXene core; there are no voids or gaps that can be seen, as GO covers the core efficiently. These structural assemblages allow the fiber to have improved mechanical characteristics, which is a benefit to the fiber. In addition, the highly conductive biomimetic fiber has shown some potential as an EMI shield. Cheng et al. announced the development of a piezoresistive sensor based on MXene that emulated a bioinspired microspinous microstructure for the measurement of very modest pressure.<sup>137</sup> In the course of this research, a thermal spray-coating technique was utilized in order to deposit a single layer of  $Ti_3C_2T_x$  in a uniform manner on polydimethylsiloxane (PDMS). Imaging using scanning electron microscopy (SEM) showed that the MXene-based PDMS microstructure had a rough surface and microspines that were randomly dispersed throughout the material. The sensor has a lot of potential in a number of pressure-sensitive applications, including human-machine interactions and the monitoring of human physiological signals.

4.5. MXene Wearable Electronics for Energy Storage and Harvesting. Supercapacitors are essential electrical devices that are desperately needed for the development of future technologies because of their capacity to store energy. Supercapacitors have emerged as the principal power source for intelligent electronics, particularly nanoscale devices, due to the fact that they combine the benefits of features including high power density, extended cycle life, and rapid charge and discharge.<sup>138</sup> Utilizing materials with a high capacitance, also known as those with a pseudoactive species or large surface area, is one of the most important factors in ensuring that electronic devices have a high energy density. The current breakthroughs with graphene are boosting studies on 2D nanomaterials in the realm of supercapacitors. Despite this, the continued development of graphene is being hampered by its poor energy density and capacitance.<sup>139</sup> On the other hand, 2D-layered MXene nanostructures can make it easier for water and electrolyte ions to intercalate between adjacent layers. MXenes are able to effectively retain charges through a technique known as cation adsorption, and they may also promote the swelling of MXene galleries, which is made possible by fast layer expansion and contraction.<sup>140</sup> For

example, it was reported that  $Ti_3C_2T_x$  MXenes have a durable specific volumetric capacitance that is close to 1500 F cm<sup>-3</sup>,<sup>140</sup> which is higher than those of all supercapacitors that are based on carbon.<sup>141</sup> In addition, the majority of MXenes are metallic conductors that can have an electrical conductivity of up to 15 000 S cm<sup>-1</sup>, which enables rapid charge transfer as well as a large current charge.<sup>142</sup> As a result of all of these advantages, MXene-based supercapacitor electrodes have a significant amount of untapped potential, which has attracted a lot of interest from investigators in the field of MXene-based supercapacitors.

There is a future opportunity for flexible devices that store energy to play a significant role in the development of contemporary electronics. These components are employed in the energy storage and harvesting systems of portable smart phones as well as outdoor wearable electronics.<sup>143</sup> The widespread use of technology that can be worn to store energy has necessitated its miniaturization. Nevertheless, the manufacture of typical sandwiched electrode supercapacitors has shown enormous volumes and poor power density. Because of this, future prospective applications, like micropower units for consuming energy targeting microelectronic equipment incorporated on the same circuit board, are restricted. As a result, the new micro-supercapacitors, which cut down significantly on the overall size of the device, have resulted in an easy fabrication procedure that makes it possible for the device to be further integrated with other coplanar microelectronic functional electronics.<sup>144</sup> Because of these cutting-edge characteristics, micro-supercapacitors have the potential to be utilized in a wide variety of fields, including wearable and flexible electronics, nanorobotics, maintenancefree wireless sensor networks, and micro-electromechanical systems (MEMS).<sup>145</sup>

MXenes are capable of helping form a viscous, stable, and water-based colloidal suspension or ink without the need for further surfactant or polymer additives. In addition to their exceptional electrochemical properties, MXenes benefit from superior characteristics like a microscale lateral size, an atomically layered thickness, and superior hydrophilicity. Therefore, MXenes are intriguing candidates for use as emerging materials in MSC electrodes. It is extremely desirable to build flexible electrodes that have both outstanding mechanical qualities and high electrochemical effectiveness in order to manufacture high-performance, robust MXene-based micro-supercapacitors.<sup>146</sup>

The term triboelectric nanogenerators (TENGs) refers to a type of energy harvesting device that is recognized as being successful despite its tiny volume, low cost, friendliness toward humans, and other desirable characteristics.<sup>147,148</sup> TENGs are applied to convert kinetic energy to electricity. They are developed by linking electrostatic induction and triboelectrification across diverse materials. This concept was inspired by many dynamic human biomechanical motions. Depending on the triboelectric series and electrostatic induction,<sup>149</sup> the process of charge transfer and electrification in TENGs takes place as a result of sliding, contact separation, or friction between different substances like metals and polymers.<sup>150</sup> The increased dielectric constants that result from the presence of functional oxygen and fluorine groups in MXenes make these molecules attractive options for use in TENG applications. In addition to this, the 2D structure of MXene stores charge between the polymer matrix and the interlayer sheets. Through an increase in both the electrical conductivity and the



Figure 4. (A) The preparative procedure of MXene 3D aerogels utilizing a cryo-assembly method that is straightforward in nature. (B) MXene aerogels that exhibit superelasticity capabilities that remain stable throughout a range of temperatures. (C) The designed MXene-based supercapacitor. (D) Mechanical property curve shown against MXene loading. (E) The sandwich structure of an MXene solid supercapacitor exhibits remarkable energy storage capabilities. Figure reproduced with permission from ref 155. Copyright 2020 American Chemical Society.

electronegativity of the substances, MXenes are able to bring about an improvement in performance for TENGs. The performance of the TENG was highly impacted by the water molecules that bonded with the device surface, which prevented its effective energy collection capabilities. A fabricbased TENG created by Xiong et al. demonstrated promise in harvesting energy from the flow of water.<sup>151</sup> As a result, researchers developed a water-repellent fabric-based MXene/ Ecoflex composite TENG nanosystem. This system was inspired by a variety of human biomechanical movements as well as environmental circumstances.<sup>149</sup> The newly designed TENG has a peak output power of 3.69 mW at its highest and a power density of 9.24 W m<sup>-2</sup> throughout its whole surface area. Cao et al. developed a shape-adaptable TENG for use as a



**Figure 5.** (A) The preparative procedure employed for the fabrication of electrochemical micro-supercapacitors based on MXene  $(Ti_3C_2T_x)$ . (B) The development and implementation of wearable power systems that possess the capability to recharge autonomously. (C) The mechanism by which electricity is generated for the purpose of charging micro-supercapacitors. (D) The utilization of the micro-supercapacitor for powering a commercial temperature-humidity meter and a digital watch. Figure reproduced with permission from 158. Copyright 2018 Elsevier.

self-powered biomechanical sensor by incorporating CNFs, liquid MXene, and soft silicon rubber into their design.<sup>152</sup> In this TENG system, CNFs served the purpose of both a dispersion and an interlocking agent in order to ensure that an efficient interconnected web of MXene was created, while silicone rubber served the purpose of both a packaging material and a triboelectrification layer. This TENG has the ability to capture energy and has shown promise to power many electronic devices, including those for the real-time monitoring of a variety of human movements.

Controllable 3D structures were fabricated by employing 2D MXene nanosheets through a freeze-induced coassembly methodology. This method enables the incorporation of MXenes into certain heterogeneities, hence enabling the customization of 3D aerogels with both structural integrity and multifunctionality.<sup>153</sup> The utilization of functionalized cellulose nanocrystals as modifiers for the structural and framework aspects was employed in the production of multilayer MXene 3D aerogels at various length scales. This approach provides mechanical strength and different functionalities, as depicted in Figure 4. The aerogels demonstrated exceptional electrochemical properties, including temperature-

invariant superelasticity ranging from 0 to 150 °C and a high rate capacity. These features can be attributed to the presence of a high-ion route resulting from the low-tortuosity topology. The MXene quasi-solid-state supercapacitors exhibited exceptional electrochemical performance, which was characterized by an energy density of approximately  $\sim$  38.5  $\mu$ W h cm<sup>-2</sup> and a remarkable cycle stability of approximately 86.7% after undergoing 4000 cycles. These structures, which possess a significant capacity for photoresponses, can also be utilized in conjunction with photodetectors. The utilization of MXene 3D aerogels has been implemented in the development of intelligent and self-sustaining lightweight wearable electronics for the purpose of monitoring a range of human movements.<sup>153</sup> This application capitalizes on their inherent mechanical adaptability and robustness. In another investigation, cobalt-metal organic framework frameworks were produced on an MXene-carbon nanofiber mat featuring great electroconductivity and flexibility.<sup>154</sup> The composites obtained from the experiment were utilized in the development of capacitive- and battery-type functioning multicomponent electrodes. These electrodes were then employed in the creation of wearable hybrid quasi-solid-state supercapacitors

that exhibited exceptional mechanical flexibility and effectiveness. Consequently, the electrodes exhibit a narrow operating voltage range of 1.5 V, resulting in an energy density of 72.5 W h kg<sup>-1</sup> and a power density of 832.4 W kg<sup>-1</sup>. Additionally, these electrodes demonstrate long-term stability, with around 90.36% retention of capacitance. These characteristics highlight the important potential of these electrodes in the development of various smart wearable devices.

The utilization of micro-supercapacitors for the purpose of powering integrated wearable systems for monitoring has experienced notable advancements.<sup>156,157</sup> In an investigation, a self-powered wearable tracking device was designed by integrating MXene electrochemical micro-supercapacitors with a triboelectric nanogenerator (Figure 5).<sup>158</sup> The microsupercapacitor exhibited a capacitance of 23 mF cm<sup>-2</sup> and demonstrated a capacitance retention of approximately 95% following 10 000 charge-discharge cycles. Additionally, the triboelectric nanogenerator achieved a maximum output power of 7.8  $\mu$ W cm<sup>-2</sup>. The power system has the potential to be continuously charged through typical human motion, occurring at a frequency of approximately 5 Hz, without any significant observable current leakage. Micro-supercapacitors possess the potential for use in the development of diverse sensors and electronic devices.

4.6. Human-Machine Interaction (HMI). The term "wearable human-machine interaction" (or "HMI") refers to an emerging field of technology that enables humans and machines to carry out tasks, collaborate, and communicate with one another in a fashion that is colocated or coordinated.<sup>159</sup> It is desired for the flexible sensor, which serves as the main component of wearable HMI electronics, to be accurate, intelligent, collaborative, and multipurpose for feedback control. Through the use of radical polymerization, Zhao et al. were able to produce a multifunctional MXene/ poly(acrylic acid) (PAA) hydrogel. It has a high sensitivity (gauge factor of  $\approx$ 4.94 or lower), a broad detection range (0– 1081%), and a steady signal output for 500 cycles, making it suitable for use as a flexible strain sensor.<sup>160</sup> In addition to this, they were able to control a manipulator in real time and could detect human movement, showing that it has significant promise in the fields of wearable artificial intelligence and HMI. In addition to this, the MXene/PAA hydrogel exhibits photothermal conversion capability along with the capability to regulate the surface temperature in a regulated manner (RT  $\approx$ 67 °C) when exposed to near-infrared radiation. Through the integration of a flexible sensor with a personal thermal management system, their research gives fresh insight into how to construct collaborative and multipurpose wearable HMI electronics.

Self-actuated sensor systems that do not require an external power supply, like flexible TENG-based strain sensors, have garnered a lot of attention in recent years thanks to the rapid growth of Internet of Things (IoT) technology.<sup>161,162</sup> This is because these systems have a straightforward construction and are capable of self-powered active sensing. Nevertheless, in order to fulfill the actual applications of human-wearable biointegration, flexible TENGs demand stricter criteria for striking a balance between the flexibility of the material and the good electrical characteristics that it possesses. As a result of research conducted by Xu et al., the strength of the MXene– substrate interface was significantly increased using leather with a distinctive surface structure as the substrate material.<sup>160</sup> As a result of this study, an MXene film was produced that was

both mechanically robust and electrically conductive. The surface of the MXene film was given a rough texture thanks to the natural fiber structure that was present on the leather's outside. This resulted in an improvement in the electrical output performance of the TENG. The greatest output power density of the MXene film on leather based on the singleelectrode TENG can reach 0.469 mW cm<sup>-2</sup>, and the electrode output voltage can reach 199.56 V. The efficient array preparation of MXene and graphene was achieved, and it was used in a wide variety of HMI applications. This was made possible by the utilization of a technology supported by lasers. In a separate piece of research, Zhang and colleagues constructed a stretchy and flexible sensor with a sandwich construction consisting of two layers of PDMS and one layer of sensing material.<sup>163</sup> The highly conductive sensing layer consisted of silver nanoflowers and MWCNTs. The films' flexibility and conductivity were effectively increased as a result of the synergistic impact of these two nanomaterials. The sensor has a high gauge factor (1187.07), a high scalability (100%), a rapid reaction time (60 ms), and a high level of stability, enabling the detection of physiological and motionrelated signals like voice recognition, pulse, and the locations of joints. In order to implement the multipoint-distributed detection of skin surface pressure, an electronic skin array was manufactured using sensor array technology. This array was used to collect the necessary data. In addition, an intelligent glove was developed with the help of this electronic skin, and hand motion tracking was accomplished with the help of a virtual display interface, wireless transmission, and technology based on 3D modeling. The sensor has significant opportunities for further development in a variety of areas, including human-computer interfaces, mechanical control, and wearable electronic systems.

Conductive hydrogels are flexible electronic devices, which means that in addition to their one-of-a-kind appeal they also satisfy the fundamental demand for mechanical flexibility and intelligent sensing. This is a distinct facet of conductive hydrogels. It is still difficult to figure out how to give standard homogeneous conductive hydrogels and flexible sensors anisotropy while also providing a wide application temperature range. In this regard, the organized structure of a muscle served as inspiration for the development of anisotropic MXene conductive hydrogels, which were prepared via a directed freezing process.<sup>164</sup> Because MXene conductive hydrogels have anisotropic features, their mechanical qualities and electrical conductivity are improved in certain directions. Through the process of solvent substitution, the hydrogels have a temperature resistance range that extends from -36 to 25 °C. Therefore, wearable flexible sensors based on muscle-inspired MXene conductive hydrogels featuring anisotropy and tolerance to low temperatures may be created and utilized. The sensing signals are then presented on the mobile phone as images by means of wireless technology. In order to accomplish motion detection, the displayed images will vary in accordance with the signals that have been acquired. Multiple flexible sensors were also built into a 3D sensor array in order to detect the amount and geographical distribution of any stresses or forces that may be present. The MXene conductive hydrogels, exhibiting an ordered orientation as well as anisotropy, provide opportunities for flexible sensors. Flexible sensors have vast application potential, particularly in HMI compatibility and healthcare surveillance.

Wearable HMI electronic technologies are rapidly advancing, and their applications span various domains such as healthcare, gaming, productivity, and industrial settings. They aim to enhance the user experience, improve productivity, and enable more natural and efficient ways of interacting with machines and computer systems.

# 5. CONCLUSION, CHALLENGES, AND FUTURE DIRECTION

MXene is a thriving material that belongs to the 2D family. It has enhanced surface chemistry, which offers adjustable MXene effectiveness for many applications, especially in flexible electronics. According to the information presented in this Review, MXene affords enormous prospects for the construction of flexible and wearable electronic devices for mobile medical surveillance, energy storage devices, humanmachine interfaces, and EMI shielding. It is possible to notice the rapid growth of technologies based on MXene; nonetheless, prototyping and downsizing of electronic devices are still mostly confined to research facilities. In the long run, it is anticipated that freshly created technologies will eventually generate technologies that are more diversified and functionally rich than those already available in the marketplace and will complement existing electronic technological devices.

5.1. MXene Manufacturing That Is Both Sustainable and Scalable. Following the discovery of MXene, a number of different approaches to the synthesis of MXenes have been studied. Nevertheless, due to the presence of a potentially dangerous HF solution throughout the reaction system, MXene synthesis presents a number of technical challenges in addition to significant safety issues. For instance, in order to etch MAX phases, investigators used a combination of lowcontent (5% weight) HCl and HF. The quality of the MXene end product generated using this approach was comparable to that of MXene generated from a highly concentrated HF solution.<sup>165</sup> Several methods have been devised by researchers to reduce the concentration level of HF. In spite of the fact that numerous salt-based etching methods have been established for the production of MXenes, an efficient etchant for Ti<sub>3</sub>AlC<sub>2</sub> was found to be a combination of hydrochloric acid and lithium fluoride.<sup>166</sup> Recently, quantities of delaminated  $Ti_3C_2T_x$  MXene nanosheets ranging in weight from 1 to 50 g have been created. Furthermore, wet chemical etching of MXene may enhance its manufacturing scalability to levels greater than kilograms or even volumes of tons, which can be done by scaling up the capacity of the etchants and the reactor. This might be important if you want to produce MXene in larger numbers. In addition, the manufacture of the device requires that the integrity of the MXene used and the functional parameters of the material, including specific surface layers, flake sizes, interlayer spacing, etc., be assured. Exploring different forms of MXene involves more rigorous efforts due to the fact that it is a vast family of 2D substances. This is accomplished in order to promote a variety of properties that may be used in device applications. As a result, the development of accurate synthesis processes and the utilization of sophisticated characterization methods to uncover the features of MXene are both extremely important.

Oxidation is frequently found to be one of the most significant obstacles that must be overcome during the manufacturing process of MXene-based electronics. MXene nanosheets have a propensity to oxidize when exposed to air, which is problematic for applications that take place in ambient

conditions and run for extended periods of time.<sup>167</sup> In addition to the parameters used for the synthesis and the overall quality of the initial MXene, there are a few elements that directly impact the oxidative degradation of MXene. These factors include the temperature of the solution, the environment in which it is stored, and the level of concentration of the dispersion. As a result, a number of studies have been carried out in an effort to enhance the oxidation stability. These studies have included the following: the synthesis of minimally defective layered MXenes, the improvement of storage requirements, the process of passivation of the defects of nanosheets of MXene within aqueous dispersions, the replacement of the water medium in organic solvents, and many others.<sup>168</sup> In addition, the rate of oxidation of MXene can be greatly reduced by encapsulating it within a polymeric matrix (for example, polydopamine (PDA), polyvinyl acetate (PVA), etc.).<sup>168</sup> To date, a great number of solutions have been devised to combat the oxidative reactions that occur in MXenes, demonstrating the materials' potential for use in forthcoming advanced applications.

5.2. Challenges with the Engineering of Wearable MXene-Based Electronic Devices. 5.2.1. Reliability in Collecting Data for Portable Devices. Wearable electronic devices, wireless networking, healthcare, and human-machine interfaces are some of the fields that have made use of products based on MXene. For instance, electronics based on MXene are now being employed in a variety of wireless devices for the capture of bioelectrical signals (electrocardiogram (ECG) and electromyogram (EMG)).<sup>169</sup> Nevertheless, these biophysical statistics must be repeatable, reliable, and exact, and any change in the process of acquiring accurate information may appear to go against common sense Because of this, electrodes made out of MXene should have a high level of effectiveness, and the signals that they create should not be impacted by any additional disturbances or noise. Nevertheless, in order to ensure the dependability as well as the utilization of the data, an integrated strategy is necessary for the processing of biophysical information as well as the evaluation of biophysical data.

5.2.2. Developing Electronics That Can Adapt to Human Skin. Electrical devices that may be worn and attached to the skin must have an adequate degree of adaptability and elasticity. In recent times, electrodes based on MXene have been used for multifunctional skin biosensing (electroencephalogram (EEG), EMG, and ECG surveillance), and the devices have demonstrated skin-adhered effectiveness on a variety of bending phenomena.<sup>169,170</sup> MXene has been utilized in a number of skin-conforming structures due to its great mechanical rigidity. On the other hand, it is expected that using these devices for an extended period of time would not have any adverse effects on human skin. Even though earlier research demonstrated that MXene has a low degree of toxicity, further systematic research is necessary to verify the compound's biocompatibility for use in bioelectronics.

5.2.3. Creating Functioning Structures with Newly Acquired Characteristics. MXene is very compatible with different substrates or nanomaterials, and as a result it develops synergy when combined with others, which is how its efficiency in device development may be described. For the purpose of developing multipurpose applications, MXene has been investigated in a wide variety of nanostructural forms, including 2D + 1D, 2D + 2D, 3D, and 2D + 0Dheterostructures. It is possible that high levels of efficiency and functionality can be achieved through the development of multiple application-wise customized structures. As a result, foamlike porous MXene and a segregating structural aerogel may be useful for applications involving EMI shielding. In a similar vein, a number of unique bioinspired structures could constitute new additions for improved practical uses. When compared with alternative stiff and cumbersome constructions, textile-based technologies have the potential to open up a greater number of avenues for potential skin applications. Across the board, the primary focus should be on the creation of innovative nanostructures that exhibit extraordinary features. This is something that is attainable with the use of logic and deliberation in the name of MXene composite nanostructures.

5.2.4. Developing Comprehensive, All-in-One Wearable Alternatives. Even if many of the currently available wearable electronic devices serve a particular purpose, it is possible that their overall effectiveness might be improved by integration into a single electronic system. It would be worthwhile to design sensors that include more functionalities in a single portable system, eliminating the necessity for several electronic modules or sensors to be housed within a body. In comparison to a single electrode, for instance, a smart-clothing platform that includes built-in sensors providing Joule heating, strain sensing, energy harvesting and storage, etc., might be attractive for diverse functions with all-in-one wearable operation. In recent times, a number of incorporated MXene electrodes have been produced, but their functionality is restricted to solely strain sensing and Joule heating. As a result, it is of the utmost importance to build further integrated systems in order to create an all-in-one wearable approach and a self-powered platform.

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#### Notes

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