Smart MXene-based Bioelectronic Devices as Wearable Self-powered Health Monitor for Sensing Human Physiological Signals

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Abstract

Biosafe wearable healthcare monitor has attracted significant attention owing to their applicability to wearable electronics. However, the narrow sensing range and poor response limit the application of flexible devices for comprehensive monitoring of human health-related physiological signals (i.e. pulse diagnosis). Critical challenges remain in the development of biocompatible materials and the design of flexible bio-integrated platforms for these purposes, targeting performance approaching those of conventional wafer-based technologies and long-term operational stability. In this context, this work presents a robust and flexible MXene/polydopamine (PDA)-composite-film-based pressure sensor in a portable/wearable fashion, which establishes a unique intercalated spherical-like PDA molecules structure, thereby resulting in excellent sensing performance. The MXene/PDA-based pressure sensor has sensitivity of up to 138.8 kPa-1 in the pressure range of 0.18-6.20 kPa with fast response and recovery speed (t1<100 ms; t2<50 ms). Associated embodiment involves real-time precise measurements of a variety of health-related physiological signals, ranging from wrist pulse, to finger motions, to vocalization and to facial expressions, with high sensitivity and accuracy. Studies on human subjects establish the clinical significance of these devices for future opportunities of health monitoring and intelligent control to predict and diagnose diseases.

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Biosafe wearable healthcare monitor has attracted significant attention owing to their applicability to wearable electronics. However, the narrow sensing range and poor response limit the application of flexible devices for comprehensive monitoring of human health-related physiological signals (i.e. pulse diagnosis). Critical challenges remain in the development of biocompatible materials and the design of flexible bio-integrated platforms for these purposes, targeting performance approaching those of conventional wafer-based technologies and long-term operational stability. In this context, this work presents a robust and flexible MXene/polydopamine (PDA)-composite-film-based pressure sensor in a portable/wearable fashion, which establishes a unique intercalated spherical-like PDA molecules structure, thereby resulting in excellent sensing performance. The MXene/PDA-based pressure sensor has sensitivity of up to 138.8 kPa^{-1} in the pressure range of 0.18-6.20 kPa with fast response and recovery speed (t_1<100 ms; t_2<50 ms). Associated embodiment involves real-time precise measurements of a variety of health-related physiological signals, ranging from wrist pulse, to finger motions, to vocalization and to facial expressions, with high sensitivity and accuracy. Studies on human subjects establish the clinical significance of these devices for future opportunities of health monitoring and intelligent control to predict and diagnose diseases.

Keywords: MXene, Polydopamine, Flexible pressure sensors, Health monitor

1. Introduction

Recent interest exists in materials and fabrication techniques that enable wearable electronics to be constructed with applications in daily health monitoring, movement tracking, disease diagnosis, and intelligent medicine. [1-5] Such technologies utilize the capability of sensing materials to convert health-related physiological signals into electrical forms of energy. Pressure sensors, to some extent, play a pivotal role in intelligent wearable devices. A significant challenge of conventional pressure sensors exposes the limitations in narrow detection limit, low sensing performance, and unsuitability of large-scale production. Suitable choices of sensing materials, reasonable layout design and integration fashion have access to establish flexible platforms, in some ways that overcome such limitations. The exertion of sensors formed in compatibility and flexibility could offer opportunities for directly monitoring and long-term stable performance on the soft interfaces.

Effective efforts to the achievement of pressure sensors in high performance involve electronic materials and mechanics principles. For example, carbon materials are highly positive candidates for the construction of pressure sensors, ranging from rigid activated carbon, graphene, carbon black, to carbon nanotubes.[6-8]However, the binding forces of these materials must be further improved for practical applications. Additionally, recent works demonstrated that pressure sensors generated by the conversion of textiles or conductive hydrogels usually exhibit excellent performance. In some cases, the sensitivity of micro-pressure monitoring far exceeds the range attainable to the detection,[9] thereby photo-lithographically manufacturing microstructures (such as pyramids) is more easily achieved than non-structured counterparts. Although the construction of microstructures is intuitively accessible to the enhancement of sensitivity, there are limitations to the production of large areas and the cost of per unit area.[10] The above limitations lead to a better understanding of the selection of pressure-sensitive materials. Previously reported extensive exploration of 2D materials in terms of transition metal carbides, carbonitrides, and nitrides (MXenes) tends to accomplish sophisticated electronic functions. The chemical composition of MXene is $M_{n+1}X_nT_x$, where M, X, and T represent the transition metal, C/N ratio, and surface functional group, respectively, and n represents the number of surface functional groups.[11] The practical possibilities of MXene for the construction of multifunctional devices to advantages in good electrical conductivity, photothermal conversion ability, rich surface chemical properties, and large specific surface area are promising. By comparison with conventional two-dimensional materials (such as GO), MXene also shows better antibacterial properties, es-

pecially in terms of gram-positive and gram-negative bacteria. However, because of the low length-to-width ratio, directly assembling MXene nanosheets into the desired macrostructure with the ability of monitoring in real time is difficulty, thus limiting their applications.[12]Therefore, MXene displays excellent electrical and mechanical properties, and can be formed in the formats of large-areas, the suitable utility of flexible bio-integrated platforms.

Previous studies have confirmed that composites of polymers and MXene have the potential for flexible sensing owing to good processability and low cost. In such systems, polymers such as polyvinyl alcohol, polyvinyl butyral, polydimethylsiloxane, polyvinylidene fluoride, and polyaniline serve as the monomer with MXene for diverse functional composites.[13-18] Dopamine (DA) is a natural, non-toxic biomolecule that mimics the structure of the adhesion proteins in mussels and seaweed. It has a large number of amine and catechol functional groups, which are environmentally friendly and can reduce the health risks when applied to human skin, making it very attractive for flexible electronic products.[19] DA adhesively polymerized on the target substrate in situ is well configured for the protection of the circuit. In addition, DA polymerized into polydopamine (PDA) leads to the spontaneous formation of hydrogen bonds with abundant active functional groups.[20] Therefore, the construction of a three-dimensional (3D) MXene/PDA composite film is expected to provide flexible pressure sensors with excellent sensitivity and biosafety.

Here, we report a portable and wearable MXene/polydopamine (PDA)-composite-film-based pressure sensor, in which the key functional constituent consists of the molecular structure of intercalated spherical PDA, thereby contributing to the large-area fabrication technique and the high-performance operation. Systematic studies of formulations of composite films with or without polymerization necessitate the layout design and integration fashion. The sensitivity of the MXene/PDA-based pressure sensor is up to 138.8 kPa^{-1} when the pressure range is 0.18-6.20 kPa with fast response and recovery speed ($t1<100$ ms; $t2<50$ ms). The sensor enables sensitive and accurate modes of precise measurements of various health-related physiological signals in real-time, involving wrist pulse, finger motions, vocalization and facial expressions. The results demonstrate that MXene/polydopamine (PDA)-composite-film-based pressure sensor serve as the basis of portable and wearable platforms relevant to health monitoring and prediction of disease diagnosis.

2. Results and discussion

Figure 1a illustrates the fabrication process for the MXene/PDA composite film. First, MXene ($Ti_3C_2T_x$) nanosheets are prepared by selectively etching an Al layer in the $Ti_3Al_2C_2$ MAX phase. A large number of surface functional groups, such as -O, -OH, and –F, can be generated on the MXene nanosheets, as demonstrated through Raman characterizations (Figure S1).[21, 22]Subsequently, DA molecules are adsorbed and further polymerized in situ into the PDA macromolecules on the MXene nanosheets. PDA can be used as a bridge between the MXene sheets to form a unique 3D cross-linked structure. In addition, the hydrogen bond between MXene nanosheets and PDA molecules facilitates the formation of the organic-inorganic hybrid membrane after vacuum filtration. The MXene nanosheets are stacked in a lamellar stacking structure after vacuum filtration (Figure 1b). The fabrication of the pressure sensor is illustrated in Figure 1c. The MXene/PDA composite membrane is designed as a sandwich structure by covering both sides with flexible Cu electrodes. The flexible film is further encapsulated in PVC films for continuous detection. The pressure sensor can be integrated with intelligent devices for commercial medical applications such as monitoring and analyzing the heartbeats and pulse, movements, and subtle signals, including those of sound and swallowing (Figure 1d), to achieve early warning of physical abnormalities.

In Figure 2a, the cross-section image of MXene/PDA composite film shows an ordered layered structure with many interlayer gaps, and the PDA molecules are embedded between the interlaced MXene nanosheets. Top-view SEM image in Figure 2b indicates that the MXene/PDA film surface was uniform and dense with the PDA molecules. Pristine MXene films are also shown for comparison (Figure S2a and 2b). The atomic force microscopy images further confirmed the rough surface morphology and change in thickness after DA polymerization (Figure S2c). Transmission electron microscopy (TEM) is used to clarify the morphology and

composition of the MXene/PDA composites. As shown in Figure 2c, both the lamellar structure of MXene and amorphous structure of PDA can be observed in the TEM image, showing that PDA was uniformly composited on the surface of MXene. The XRD patterns of MXene/PDA films of different ratios are shown in Figure 2d. The diffraction peak at 2ϑ=6.4° is attributed to the (002) plane of the MXene nanosheets. Compared with the XRD pattern of the $Ti_3Al_2C_2$ MAX phase (Figure S3), there is no characteristic peak at $2\vartheta = 39°$, indicating that the product obtained by etching has no impurity-phase Al. To prove the successful compounding of PDA and MXene, the (002) peaks of the original $Ti_3C_2T_x$ MXene and composite films are further compared. After polymerization, the blue-shifted (002) diffraction peaks indicates an increase in the nanosheet spacing. The increased layer spacing clearly evidences that the PDA is inserted between the $Ti_3C_2T_x$ MXene nanosheets.[23] With increase in the DA content, the (002) diffraction peak increases, however, the (002) peak shows red-shifted with further improvement in the composite ratio with the ratio of 1:3. Furthermore, the composite film becomes fragile and it is no longer suitable for flexible monitoring of physical activities. The structures of the MXene, PDA, and MXene/PDA composites are studied by Raman spectroscopy in Figure S1, the Raman peaks at 204, 390 and 627 cm^{-1} corresponded to the active groups such as -O (A1g of $Ti_3C_2O_2$), -F (E_g of $Ti_3C_2F_2$), and-OH (E_g of $Ti_3C_2(OH)_2$),[24] respectively. The Raman peak at 1585 cm^{-1} is assigned to C=C stretching benzenoid ring. The Raman peaks of the PDA molecular at 1338, 1415 and 1565 cm^{-1} are ascribed to C=O catechol stretching vibrations.[19] The characteristic Raman peaks for both MXene and PDA prove that the MXene/PDA composite have been successfully prepared. The FTIR spectra of MXene and MXene/PDA composite film are shown in Figure 2e. For the spectrum of MXene, a prominent absorption band at 3452 cm^{-1} is observed, which could be attributed to the stretching vibration of -OH.[13] The strong absorption band at 3431 cm^{-1} assigns to the strong hydrogen bonding interaction between MXene and PDA molecules in composite film.[20, 23] The chemical states of MXene and MXene/PDA composite are characterized by XPS. As shown in Figure S5, the XPS survey spectrum indicates that the MXene nanosheets is mainly composed of Ti, C, O and F elements.[25, 26] No related peaks of Al element are detected, indicating that the Al layer in the raw material Ti_3AlC_2 has been completely removed during the etching process.[27]. The peak intensity of the MXene/PDA composite corresponding to F element is weaker than that of MXene, which could be attributed to the change of the surface after compounding dopamine (Figure 2f).[28] On the other hand, the introduction of dopamine increases the content of N, O and C elements. The fine chemical state of MXene and MXene/PDA is further studied. As shown in Figure 2f, it shows the high-resolution Ti 2p spectra of MXene/PDA composite film, which spectrum could be deconvoluted into four peaks at 461.3, 457.1, 455.9, and 455.0 eV, which corresponded to Ti-C $2p_{1/2}$, Ti^{3+}, Ti^{2+}, and Ti-C $2p_{3/2}$, respectively.[20, 29] The C 1s spectrum for the MXene/PDA flexible film can be divided into three peaks centered at 286.2 eV, 284.8 eV and 281.9 eV, and correspond to the presence of C-O/C-N, C-C and C-Ti bonds, respectively.[30] The Ti-C peak could be attributed to the Ti element in the $Ti_3C_2T_x$ nanosheets, which is a special feature of the MXene. The C-N bond comes from the PDA. The high-resolution O 1s spectrum of MXene/PDA containes three specific peaks at 532.3 eV, 531.0 eV and 529.7 eV, which are attributed to C-O, C-Ti-$(OH)_x$ and Ti-O bonds, respectively.[29] Compared with the O 1s spectrum of MXene in Figure S4c, the ratio of TiO_2 is determined by 41.9% is approaching 22.9%. The strong reduction of dopamine molecules would inhibit the oxidizable nature of MXene. The specific peak at 685.2 eV is attributed to the F-Ti bonds. The absence of F-Al peak indicates the successful removal of Al element in $Ti_3C_2T_x$.[29, 31]

The influence of dopamine on the sensing behavior is studied at a pressure of 0.93 kPa by adjusting the mass ratios of the MXene nanosheets and dopamine to 1:0, 3:1, 1:1 and 1:3,as shown in Figure S5. The pressure response increases with the increase in the ratio of MXene to DA (1:0, 3:1, 1:1). With an increase in the DA content, the interlayer spacing gradually increases, which produces high deformation and high response under loading pressure. As the ratio of DA is increased further (1:3), the corresponding response decreases under the loading pressure. The above results can be attributed to the following reasons: (i) regulation of the conductive path from PDA to MXene nanosheets. Polymers are typically weak conductors of electricity. After compositing with PDA, the increased number of conductive paths of the MXene nanosheets resultes in lower initial current. The interlamellar spacing of the MXene nanosheets decrease with increasing in applied pressure. As shown in Figure3a and 3b, dopamine molecules are embedded in the MXene nanosheets,which

increase the distance between them; this is consistent with the XRD pattern analysis. The increased contact area and conductive paths between the MXene flakes help improve the current conduction of the composite membrane after the application of pressure. (ii) The unique spherical-like structure of PDA provides good deformation performance. The DA molecules are polymerized on the surface of the MXene nanosheets, and a porous structure is fabricated. These porous networks extending into the soft substrate can adjust the compressive deformation distribution in the MXene/PDA hybrid film. Thus, such a layered porous network structure can induce good pressure-sensitive properties in flexible sensors. The I-V curves of the flexible pressure sensor with the ratio of MXene to dopamine of 1:1 are obtained at different voltages under different applied pressures. As shown in Figure 3c, the current signal increases with the increasing of the voltage, and the slope of the I-V curves is positively correlated with the exerted pressure, indicating a stable sensitive performance. The dynamic sensitivity curve of the MXene/PDA-based flexible sensor is obtained under different pressure gradients. The sensitivity S is defined as $S= \delta(\Delta I/I_0)/\delta_P$, where ΔI is the difference between the current after the load pressure and the initial current, I_0 is the initial current, and P is the pressure applied on the sensor.[23] As shown in Figure 3d, the sensitivity of the device gradually increases in the range of 0.18-6.20 kPa, and each dynamic response curve is uninterrupted and stable. Figure 3e shows that the sensor could clearly distinguish the signals generated by the water drops with different weight. As shown in Figure 3f, the MXene/PDA-based flexible sensor exhibits fast response/recovery speeds of 100 and 50 ms, respectively, and the fast response and recovery capabilities ensure timely and flexible sensing under the action of external forces. The relationship between sensitivity and applied pressure in Figure 3g can be divided into two areas: (i) in the low-pressure (0.18-2.90 kPa) range, the sensitivity of S_1 was 24.7 kPa^{-1}; (ii) in high-pressure (2.90-6.20 kPa) range, the sensitivity of S_2 was 138.8 kPa^{-1}. Overall, the sensitivity of the MXene/PDA-based pressure sensor is higher than that of the MXene film (Figure S6). The repeatability of the MXene/PDA thin-film flexible sensor shown in Figure 3h indicates that the sensitivity of the device is maintained after 350 cycles of compression loading/unloading tests. The inset of Figure 3h shows that the sensitivity of the pressure sensor is maintained at almost the same amplitude over multiple compression cycles. As shown in Figure S7, the morphology of the MXene/PDA composite film remained uniform and dense after more than 350 loading/unloading cycles. Figure 3i compares the sensitivity obtained in this study with that reported in the literature.[14, 32, 33, 37-42] Compared with other MXene-based flexible sensors, MXene/PDA-based pressure sensors generally exhibit higher sensitivity, such as MXene/PVB-based porous composite sensor (S_1=11.9 kPa^{-1}, S_2=1.15 kPa^{-1}, S_3=0.20 kPa^{-1}), Ti_3C_2Tx/PVDF-TrFE-based composite nanofiber sensor (0.51 kPa^{-1}), elastic microstructure-based MXene/PDMS thin film sensor (S_1=2 kPa^{-1}, S_2=0.003 kPa^{-1}) MXene@CS@PU-based pressure sensor (3 kPa^{-1}), pressure sensor based on MXene/Cotton (12.095 kPa^{-1}), pressure sensor based on MXene/PI (22.32 kPa^{-1}), thin-film sensor based on LS-MXene/PVA (S_1=5.5 kPa^{-1}, S_2=1.5 kPa^{-1}), elastic aerogels of MXene/rGO (22.56 kPa^{-1}) and pure MXene-based thin film sensor. In addition, the response and reply times of our proposed sensor are shorter than those mentioned above, enabling higher-accuracy detection.

As dispalyed in Figure 4a, the MXene/PDA flexible pressure sensor with the flexibility and biocompatibility can be integrated with intelligent system for health monitor, owing to its high sensitivity over a wide sensing range, which can be used for health warning, physiological monitor, dignose deasease and telemedicine ranging from small physiological signals to human posture. Figure 4b shows the MXene/PDA-based composite thin-film flexible pressure sensor attached to the curved skin of the index finger joint to stably record the real-time signal changes caused by different finger motions of varying amplitudes, thereby distinguishing between the different bending angles of the fingers (from 30° to 90°). The pulse of the human wrist has clinical significance for indicating the changes in the heart rate and arterial conditions. Therefore, the use of pressure sensors to detect the pulse on a person's wrist has immense value.. As shown in Figure 4c, the MXene/PDA flexible pressure sensor can be attached to the human wrist to accurately detect the pulse and diagnose potential diseases. The enlarged waveform of the heartbeat with two distinguishable characteristic peaks is displayed, which can be ascribed to the systolic (P_1) and diastolic peaks (P_2).[33] These results further confirm that the MXene/PDA flexible sensor has excellent sensitivity even when detecting small strains. When different words are spoken, the strain sensor shows different characteristic peaks, that is, different syllables are reflected as changes in the sensor current. The words "flexible" and "carbon" may have

complex throat movements, which generate different peak-shaped patterns and differences in insensitivity in Figure 4d. Excellent voice recognition capabilities make the sensor very promising for voice recovery exercises and human–computer interactions. It holds great promise in helping speech-impaired people convey information by recognizing vocal signals. In addition, this flexible sensor can recognize subtle changes in the facial expression. For example, when the eyebrows are raised, there was an obvious response with a $^{I}/I_0 value of approximately 12 (Figure 4e). Furthmore, it can sensitively identify the bending of devices at different angles; the b$
$based pressure sensor is shown in Figure 4f. The LED is dark in the absence of external pressure but lighted up faintly after placing$
$computer interaction and intelligent control.$

3. Conclusion

In conclusion, MXene/PDA film-based flexible pressure sensors with low health risk and high sensitivity are successfully prepared via dopamine intercalation polymerization. The dopamine molecule embedded in MXene nanosheets not only increases the layer spacing of MXene but also acts as a linker to form a 3D cross-linked structure, effectively improving the mechanical and pressure-sensing performance of the device. The sensor has excellent sensitivity (S1=24.7kPa^{-1}; S2=138.8 kPa^{-1}) and a relatively short response/recovery time (<100ms/<50 ms). Flexible pressure sensors can be safely applied to human skin to collect various human vital signs, including biological signs that show tiny deformations, for example, finger bending, pulse, facial expressions and vocalization. This demonstrates the immense potential of MXene/PDA-based sensing materials for artificial skin and wearable devices.

4. Materials and Methods

Materials. 3-Hydroxytyramine hydrochloride (dopamine) was purchased from Adamas. Lithium fluoride (LiF, 99.9%) was purchased from Adamas. Ti_3AlC_2 MAX phase (400 mesh) was purchased from 11 Technology Co., Ltd. Hydrochloric acid (HCl, 37%) was purchased from Adamas.

Synthesis of MXene nanosheets . To synthesize $Ti_3C_2T_x$ nanosheets from Ti_3AlC_2 MAX phase, the etching solution is LiF/HCl.[55] First, a certain amount of LiF and HCl (9 M) was stirred for 30 min to ensure that LiF is dissolved. Then the solution was heated to 35deg C and the precursor MAX phase was slowly etched at a certain speed for 24 h to etch the Al layer. The resulting solution was repeatedly washed with deionized water until the pH value was higher than 6 to obtain a few layers of $Ti_3C_2T_x$ nanosheets.

MXene/PDA biocomposite film preparation. First of all, the concentration of MXene solution was calculated by vacuum filtration. Subsequently, dopamine of different masses was used to control the mass ratios to 1:0; 1:1/3; 1:1 and 1:3. We stirred the solution magnetically for 30 minutes at a certain speed, then adjusted the pH of the solution to about 8.5, and continued to stir continuously for 12 hours under certain conditions. Subsequently, the solvent was removed and a composite membrane was formed by vacuum filtration using a cellulose acetate membrane as a substrate. At last, it was named as MXene/PDA film and dried overnight at room temperature.

Fabrication of flexible pressure sensor. Copper films were first drawn from both ends of the MXene/PDA flexible film as electrodes, and then the two flexible PVC films were cleaned with deionized water, ethanol and acetone, respectively, and covered at both ends of the pressure-sensitive material to prevent material contamination and ensured the stability of the equipment during the test.

Characterization . The crystalline structures of MXene film and MXene/PDA were characterized by X-ray diffraction (XRD, D8-Advance, Germany) with 2ϑ in the ranges of 5˜60° at room temperature. The morphology of the $Ti_3C_2T_x$ MXene film and MXene/PDA biocomposite film were observed with field emission scanning electron microscopy (FE-SEM, JEOL JSM-7001F, Japan) and transmission electron microscopy

(TEM, Tecnai G220S-Twin, USA). The thickness and surface topography of sensitive materials were characterized by atomic force microscopy (AFM, Cypher, USA). The surface compositions and chemical states were examined by X-ray photoelectron spectroscopy (XPS, VG ESCALAB 210, USA). The sensing performances are measured by the flexible device analysis system (AES-4SD, SINO AGGTECH, China).

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Competing interests

The authors declare no competing interests.

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