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Recent progress in flexible sensors based on 2D materials

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Abstract: With the rapid development of the internet of things (IoT) and wearable electronics, the role of flexible sensors is becoming increasingly irreplaceable, due to their ability to process and convert information acquisition. Two-dimensional (2D) materials have been widely welcomed by researchers as sensitive layers, which broadens the range and application of flexible sensors due to the advantages of their large specific surface area, tunable energy bands, controllable thickness at the atomic level, stable mechanical properties, and excellent optoelectronic properties. This review focuses on five different types of 2D materials for monitoring pressure, humidity, sound, gas, and so on, to realize the recognition and conversion of human body and environmental signals. Meanwhile, the main problems and possible solutions of flexible sensors based on 2D materials as sensitive layers are summarized.

Key words: 2D materials; flexible sensors; layered structure; solution method

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1. Introduction

Sensors can convert physical or chemical signals (e.g., physiological signals from human body, mechanical deformation from the environment) into electrical signals[1–5], thermal signals[6–8] or acoustic signals[9–11], serving as a bridge for connecting human beings and nature. Flexible sensors play an irreplaceable role in the development of flexible and wearable electronics. Sensors generally consist of electrodes and sensitive layers. Researchers commonly select the appropriate materials as the sensitive layer according to the application requirements of the sensors.

2D materials have been on the upward trend since the discovery of graphene in 2004 by the group of Geim from the University of Manchester[12] and have attracted great interest from researchers. Graphene has excellent mechanical stability, flexibility and excellent thermal conductivity[13–15]. Graphene holds sensing performance for temperature, pressure, sound, and humidity. Graphene-based flexible sensors have excellent mechanical properties and high durability, thus can be applied to pulse, respiration, sound, etc.[16–19]. Following the discovery of graphene, a series of 2D materials have been discovered and extensively studied, for example, MXene, boron nitride (BN), and 2D transition metal chalcogenide (TMDs, e.g., molybdenum disulfide (MoS_2), molybdenum diselenide ($MoSe_2$), tungsten disulfide (WS_2), tungsten diselenide (WSe_2)).

The rise of MXene has received global recognition from researchers due to its excellent physical and chemical properties. For example, the large specific surface area of MXene leads to surface rich in functional groups, which increases interfacial bonding. In addition, their excellent metallic conductivity, adjustable interlayer spacing, and electromagnetic properties make them promise for exploration in electrodes, pressure sensing, piezoelectric sensing, and electromagnetic shielding[20–23].

BN has also exerted a vigorous impetus in the development of 2D materials on account of its exceptionally high thermal conductivity and temperature stability. In addition to chemical vapor deposition (CVD), BN can also be obtained by cost-effective liquid-phase exfoliation[24, 25]. BN was initially applied as the dielectric material due to its excellent photoelectric performance[26, 27], and later researchers found that BN-based heterojunctions could be adopted to detect gases[28–30], which drove the development of flexible gas sensors.

TMDs are ideal candidates for dielectric materials in field effect transistors, because of their uniform and dangling-bond-free surface affording them extremely high carrier mobility, I_{on}/I_{off} ratio, and tunable band gaps[31–35]. In particular, the large number of exposed active sites at the edges of TMDs makes them ideal candidates for gas molecule detection[30]. TMDs are generally obtained by physical or chemical vapor deposition, while researchers have discovered liquid-phase exfoliation in recent years, which greatly improves the processability of TMDs and saves energy. TMDs have excellent photoelectric properties and temperature sensitivity[36–39]. Their layered and assembled structure makes them sensitive to pressure to some extent[40–42], allowing them to be deployed as pressure, temperature, and strain sensors.

In summary, the gradual discovery of the 2D materials in recent years and their continuous optimization of prepara-

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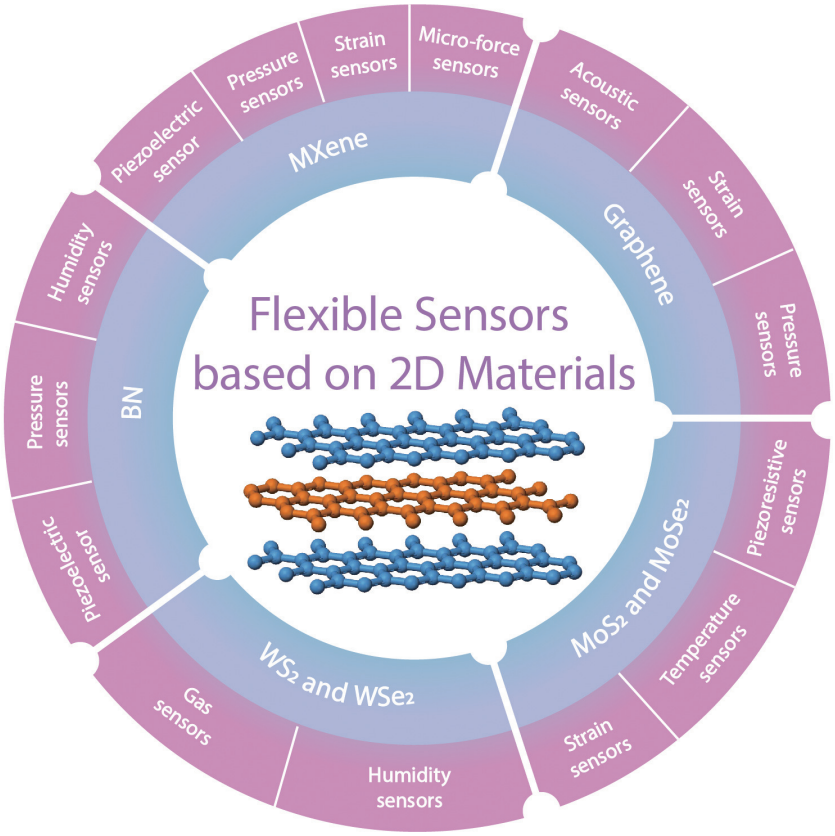

Fig. 1. (Color online) Summary of the review. Different kinds of materials with different sensing mechanism, thus for different sensors.

tion methods, especially the solution method, have been crucial advancement for the preparation of sensors based on 2D materials. The application of different types of 2D materials (i.e., graphene, MXene, BN, MoS_2 and $MoSe_2$, as well as WS_2 and WSe_2) for flexible sensors is described in detail sections, as shown in Fig. 1. Their different sensing mechanisms are introduced according to the physical or chemical properties, thus being applied to different sensing fields. We believe this review will provide systematic and comprehensive knowledge of the 2D materials in sensing, advance the development of gas sensing, strain sensing and other fields, which will contribute to human health, integration of the internet of things and so on.

2. Flexible sensors based on 2D materials

The most popular 2D materials employed in sensing can be categorized as graphene, MXene, BN, MoS_2, and $MoSe_2$, as well as WS_2 and WSe_2. The following section describes the applications of the different materials in sensing in the following order.

2.1. Graphene

Graphene is a two-dimensional material consisting of a single atomic layer of honeycomb carbon atoms, with the carbon atoms being sp^2 hybridized and the one remaining electron in the p-orbital of the carbon atom collectively forming a large π-bond. The σ-bond constituted by the sp^2 bond of carbon atoms enables graphene to be structurally stable and flexible. Graphene is a zero-bandgap semi-metallic material[43]. The edge of graphene determines its electrical and magnetic properties[44]. Graphene exhibits high electron mobility but low carrier concentration, so the carrier concentration of graphene can be increased by doping to increase the electrical conductivity[45]. Accordingly, graphene has extremely excellent mechanical property[46, 47], electrical property[48, 49], and thermal conductivity[50, 51], due to its unique atomic arrangement as an extremely hot member of 2D materials. When subjected to deformation, the carbon atoms bend, avoiding atomic rearrangement and maintaining stability under stress[52]. Unexpectedly, the nearly perfect structure of graphene brings significant drawbacks in its application due to the inert surface, leading to agglomeration during dispersion[53]. To expand its utility, researchers modified it to introduce abundant active functional groups on its surface, such as carboxyl and hydroxyl groups[54, 55], resulting in oxidized graphene (GO) and reduced oxidized graphene (rGO).

To address the sensitivity and detection range compatibility of the flexible sensors, Tao *et al.* attached graphene onto the paper and then reduced the paper, resulting in the pressure sensor named rGO-paper, as shown in Fig. 2(a). rGO-paper could achieve different sensitivity and linearity by adjusting the number of layers. When it was 5 layers, the response time and recovery time were 120 and 60 ms, respectively. The sensitivity and fatigue resistance of the sensor were excellent at pressure below 800 Pa, so it can detect small pressure, such as respiration and pulse, realizing the feedback of human physiological signals[56]. Sun *et al.* obtained strain sensor with wide operation range, high sensitivity, and low detection limit by depositing carbon nanotube (CNT), rGO, dopamine, and Hf-SiO_2 sequentially on dissolved elastic rubber band. The strain sensor exhibited operation range of 482% with gauge factor of 685.3 at maximum strain. The synergistic effect of rGO and CNT enhanced the operating range of the sensor[57].

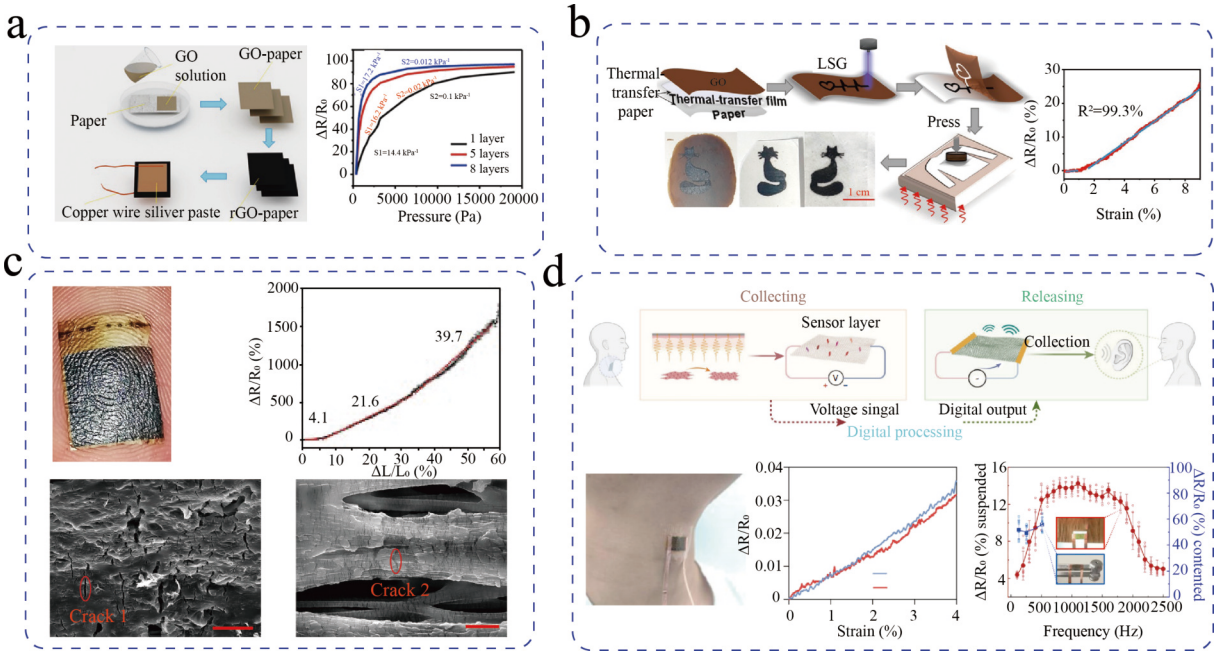

Fig. 2. (Color online) Graphene-based sensors. (a) Graphene-paper pressure sensors for detecting physiological signals. Reproduced with permission from Ref. [56], Copyright 2017, American Chemical Society. (b) Graphene-based multifunctional sensors. Reproduced with permission from Ref. [58], Copyright 2021, American Chemical Society. (c) Conformal electronic skin based on graphene. Reproduced with permission from Ref. [59], Copyright 2022, Wiley. (d) Mixed-modality speech recognition and interaction using a wearable AT. Reproduced with permission from Ref. [60], Copyright 2023, Nature Publishing Group.

Textiles-based wearable electronics are of great advantage because they can be integrated directly into clothing. By combining the advantages of graphene in electrical, mechanical and thermoacoustic, Wei *et al.* prepared multifunctional flexible sensor on fabric by laser scribing and thermal transfer, named graphene-based textile (GT), as shown in Fig. 2(b) The pressure linearity of GT was 98.2%, the maximum pressure reached 1000 kPa, and the tension linearity was 99.3%. In addition, the unique structure of graphene realized the transformation from heat to sound, which is rare in other materials. Under the action of alternating voltage, GT generated periodic heat, causing the surrounding air to alternately expand and contract, producing sound, which was then collected into the microphone to achieve sound[58]. Although both paper and fabric can be attached to clothing to meet wearable requirements, yet they are too thick, resulting in poor conformality between sensors and skin. Qiao *et al.* produced conformal electronic skin by dribbling GO solution onto electrospinning films and then laser sintering the dry films to reduce the resistance and improve the sensitivity, as is shown in Fig. 2(c). With the gradual increase of strain, the crack network changed from crack of relatively large size to double crack mechanism containing large crack and small crack. As a result, the gauge factor (GF) reached 40, and the signal of human epidermis was monitored[59]. Sharma *et al.* integrated a tactile glove with temperature, humidity and pressure sensors by utilizing CNT-coated laser-engraved graphene as the sensitive layer and designing different circuit structures. The same material eliminated interface problems at the connection of different sensors. The clever design allowed the glove to detect change in temperature, humidity, and pressure respectively when gripping a ball in hot water[61].

In addition to motion detection sensors that most commonly used, sensors that monitor physiological signals also have extremely important research value. The stable monitoring of physiological signals concerns human life and health. Lorestani *et al.* obtained sensors for epidermal sweat and temperature monitoring by dropping glucose-sensitive solution on the surface of sintered porous graphene (The solution consists of silver/gold alloy nanoparticle graphene oxide-carbon nanotube nanocomposite). Glucose undergone oxidation/reduction reactions leading to change in the concentration of H^+ and electrical signals changed[62]. Moreover, sound is one of the important ways in which people communicate with the world. However, the current flexible acoustic sensors can only emit sound and receive sound respectively, and cannot achieve dual-functional at the same time[63]. Yang *et al.* prepared the flexible sensors with dual-functional by combining the pressure-sensitive properties and thermoacoustic effects of graphene. The polyimide (PI) film was transformed into porous graphene by laser ablation, named artificial throat (AT), as shown in Fig. 2(d). The ultra-thin feature of AT allows it to fit perfectly on the skin, so it could detect the tiny vibrations emitted by the throat of people with voice disorders, and convert them into electrical signals that can be read due to the piezoresistive effect. In addition, the thermoacoustic effect of graphene can also produce sound under the action of alternating currents[60]. The prepared acoustic devices promote the application of flexible electronics in the wearable field, and bring facility to some people with voice disorders.

2.2. MXene

As an emerging 2D material, MXene has attracted the interest of researchers at an alarming rate. MXene has extremely outstanding mechanical properties and electrical properties[7, 20–23]. MXene, as a polyatomic material, has various components that affect the electrical conductivity,

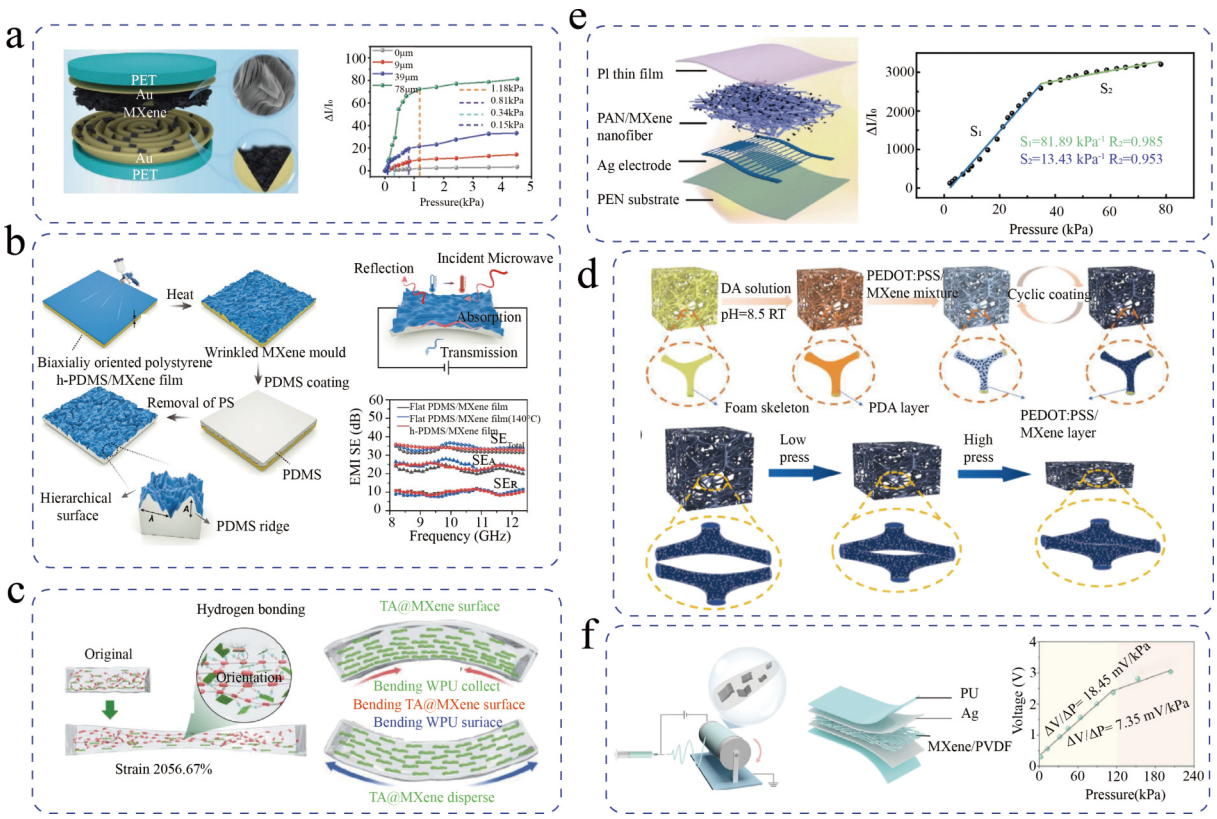

Fig. 3. (Color online) MXene-based sensors. (a) Micro-force sensor combined with MXene and microchannel. Reproduced with permission from Ref. [68], Copyright 2020, Wiley. (b) Pressure sensor with electromagnetic shielding based on MXene of kirigami-inspired structure. Reproduced with permission from Ref. [69], Copyright 2021, American Chemical Society. (c) Self-healing strain sensor based on PU-TA@MXene Janus architecture. Reproduced with permission from Ref. [70], Copyright 2023, Wiley. (d) Piezoresistive sensor based on MXene/PEDOT:PSS. Reproduced with permission from Ref. [71], Copyright 2021, American Institute of Physics. (e) Pressure sensor with PAN/MXene nanofiber as the sensitive layer. Reproduced with permission from Ref. [72], Copyright 2022, Wiley. (f) Piezoelectric sensor with confined MXene/PVDF nanofiber. Reproduced with permission from Ref. [73], Copyright 2023, Springer.

such as transition metals, X atoms, surface functional groups, and defects, etc. The electrical conductivity of MXene is higher when the X atoms are nitrogen or carbon and nitrogen than when X is C[64]. Yury's group obtained MXene by etching and intercalation in 2021, which achieved conductivity of 20 000 S/cm[65]. Different preparation and assembling methods affect the conductivity. In addition, there are a good deal of polar functional groups (e.g., hydroxyl(–OH), –Cl, –F, –I,) on the surface of MXene[66]. MXene can interact with a variety of materials, equipping with a particularly wide range of research and application prospects.

The force sensitivity of MXene is attributed to its excellent layered structure. Ma *et al.* obtained MXene with controllable sensitivity by controlling the temperature and degree of oxidation during preparation. As the oxidation on the edges of the MXene increased, the sheet spacing increased. As a consequence, greater oxidized MXene sheets produced greater deformation and more contact area up to a point that the electrical signal change were more significant[67]. Gao *et al.* designed piezoresistive mechanical sensor to detect micro-force and expanded the detection range by adding microchannel structure similar to the optical disc, as shown in Fig. 3(a). In addition, the depth of the channel also affected the sensitivity of the sensor. The sensitivity of the sensor was up to 99.5 kPa^{-1}, the minimum detection stress was 9 Pa, and response time was only 4 ms[68].

Apart from the channel structure, the wrinkle structure also improves the performance of the sensors greatly. Chen *et al.* prepared the resistive strain sensor with wrinkle structure of the heat shrinkable film, as shown in Fig. 3(b)[69]. With the assistance of the wrinkle structure, the strain sensor could withstand maximum of 100% strain, which overcomed the non-stretchablity of MXene. In addition, the sensor also equipped with excellent electromagnetic shielding properties, which is attributed to the excellent electromagnetic shielding properties of MXene[74, 75], promoting multi-functional applications based on MXene devices.

With the increase of components required by people, it is urgent to exploit devices with special properties such as anisotropy. Bai *et al.* designed the flexible sensor that could detect directional bending inspired by the non-uniform and symmetrical skin, as shown in Fig. 3(c). By mixing the MXene modified by tannic acid (TA) with the elastic polymer of water-soluble polyurethane (PU), TA@MXene with gradient distribution in gravity direction were obtained, named PU–TA@MXene. In addition, TA improved the binding force between MXene and PU. The fracture strain of PU–TA@MXene was up to 2057% and the tensile strength was 50.78 MPa, which can withstand the weight of 2.287 kg. Importantly, the backward bending could be detected by PU–TA@MXene because of the gradient distribution of TA@MXene, which provided a broader idea in stress detection[70]. The foam type porous structure has the natural advantage for pressure detection. Zhang *et al.* prepared foam-struc-

tured piezoresistive sensor by filling dopamine-modified commercial melamine foam (MF) with the mixture of Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PDEOT:PSS) and MXene, as shown in Fig. 3(d). The rich internal pore structure made it provide vital deformation space, and the contact area changed under pressure. The final compressive strain reached 60 kPa, and the linearity was 0.99. In addition, Melamine foam were also preeminent elastomer, so the sensor could withstand strain of 80%, and GF of 171.688 at large strain[71].

Spinning films are also a class of excellent structures in the research of sensors. Furthermore, the mixing of micro-sized or nanometer-sized fillers with polymer in spinning film provides confinement effect on conductive fillers, further enhancing its sensing performance. Liu *et al.* obtained an ultra-thin pressure sensor by airflow spinning the mixture of MXene and polyacrylonitrile (PAN) as the sensitive layer, as shown in Fig. 3(e). The sensitivity and linearity of the sensor were 81.95 kPa^{-1} and 0.985, respectively, when the pressure was less than 40 kPa. The sensitivity and linearity were 13.43 kPa^{-1} and 0.953, respectively, when the pressure increased to 80 kPa. In addition, MXene exhibited the best sensitivity among many conductive fillers, such as Au, carbon black (CB), CNT and AgNWs. Mentioned that airflow spinning is energy-saving and is a method that can be popularized massively[72]. Similarly, Zheng *et al.* reported multilayer fiber structure based sensor with pressure and humidity response. AgNWs/MXene was sprayed on the fiber as the electrode/sensing layer. The porous structure enhanced the working range and sensitivity owing to the increased contact area under pressure. The final sensitivity and working range were 770.86–1434.89 and 0–100 kPa^{-1}, respectively[76]. Zhang *et al.* obtained the piezoelectric type of sensor with wider operating range by electrospinning the mixed solution of PVDF and MXene, as shown in Fig. 3(f). F in PVDF interacted with MXene. The sensitivity of the sensor increased and then decreased with the increase of MXene content. The prepared sensors can be adhered to the skin, attributing to the flexible and ultrathin properties of the electrospinning film[73].

Similar to graphene, MXene-based sensitive layers can also be employed to detect acoustic signals attributed to the piezoresistive properties. Intercalating bacterial cellulose in MXene improved flexibility and satisfied the strain range of the throat[77]. Shao *et al.* obtained full MXene functional circuits by directly printing aqueous solutions of MXene. The line spacing could be as small as 3 μm due to the lamellar structure and relatively homogeneous size of MXene. The sensitivity of the MXene-based sensitive layer to temperature was 0.066% $°C^{-1}$ and also responded quickly to humidity. The high resolution printing of the inks under room temperature conditions advanced the mass manufacturing of flexible electronics[78].

2.3. BN

BN consists of atomically planar layers of alternating hexagonal B and N atoms that are holds together by van der Waals interactions (vdW). Crystalline BN exhibits a graphene-like layered structure consisting of sp^2 hybridized, strongly covalent and highly polarized B–N bonds along the planes. However, unlike graphene, the B–N is strongly polarized with electronegative nitrogen atoms, giving rise to possible anisotropic properties[79]. Li *et al.* reported that BN presents wide band gap, high thermal conductivity, excellent strength, good flexibility, and excellent thermal and chemical stability[79–83]. The large surface area of BN provides an even greater advantage, and its surface is also easily modulated by covalent/non-covalent, which results in gas molecules being able to adsorb a greater number of active sites[84–86]. Similar to graphene, the structure of BN deforms when subjected to pressure[79]. In addition, BN can be prepared by solution methods[84], which is energy-efficient and environmentally friendly. Therefore, lots of researchers have utilized BN as the sensitive layer for gas sensors, pressure sensors, and humidity sensors.

Researchers usually prepare BN nanosheets by liquid-phase exfoliation, but the high hydrophobicity of BN nanosheets leads to their low solubility in water[87, 88]. Chen *et al.* obtained stabilized dispersion solution of BN by adding 1-pyrenesulfonic acid sodium salt (PS1) as surfactant during the stripping process to improve the interaction between BN and water, as shown in Fig. 4(a). The humidity sensor was obtained by dropping the BN solution directly onto the interdigital electrode of the PI film. The linear response of the humidity sensor in the relative humidity range of 5%–95% was extremely high, as its resistance varied by more than 6 orders of magnitude. The response time of the sensor was 0.1 ms. In addition, the sensing sensitivity would not cross over at 25–60 °C[89]. These aid in the need to detect extremely small change in humidity in our daily lives (e.g., faint breathing of the patients). Harley-Trochimczyk *et al.* obtained aerogels with 3D-like stacking structure consisting of BN and Pt nanoparticle. The large surface area of the aerogel allowed loading of more gas. In addition, BN exhibited higher thermal conductivity compared to the alumina widely adopted and thus can maintain stability at temperature below 900 °C[90].

Di *et al.* designed thermally comfortable pressure sensors by combining the thermal conductivity of BN as well as the pressure sensitivity of MXene, as shown in Fig. 4(b). MXene was attached to the surface of micro-dome structure composed of the mixture of thermoplastic polyurethane (TPU) and BN, with the other side in contact with interdigital electrode. Under pressure, the contact area between the micro-dome structure and the interdigital electrode changed, thus producing the change in current. The maximum sensitivity of the device was up to 288.95 kPa^{-1} and the sensing range was up to 300 kPa. The device was temperature comfortable to wear because the BN conducted the heat away in time and there was very little decrease in thermal conductivity in the range of 20–100 °C[91]. The addition of thermal conductivity in pressure sensors improves the lifetime of the device and promotes more comfortable applications of wearable electronics in the human body.

With the gradual deterioration of the natural environment, acute outbreaks of various diseases cause the human body to face numerous unprepared moments, thus continuous blood pressure detection is crucial for human life and health, especially for comfortably wearable monitoring devices. Tian *et al.* designed multilayered piezoelectric-type sensor by electrospinning the mixture of BN and MXene with poly(vinylidene fluoride-rantrifluoroethylene) (P(VDF-TrFE)) layer by layer and then hot-pressed (Fig. 4(c)), which was a sim-

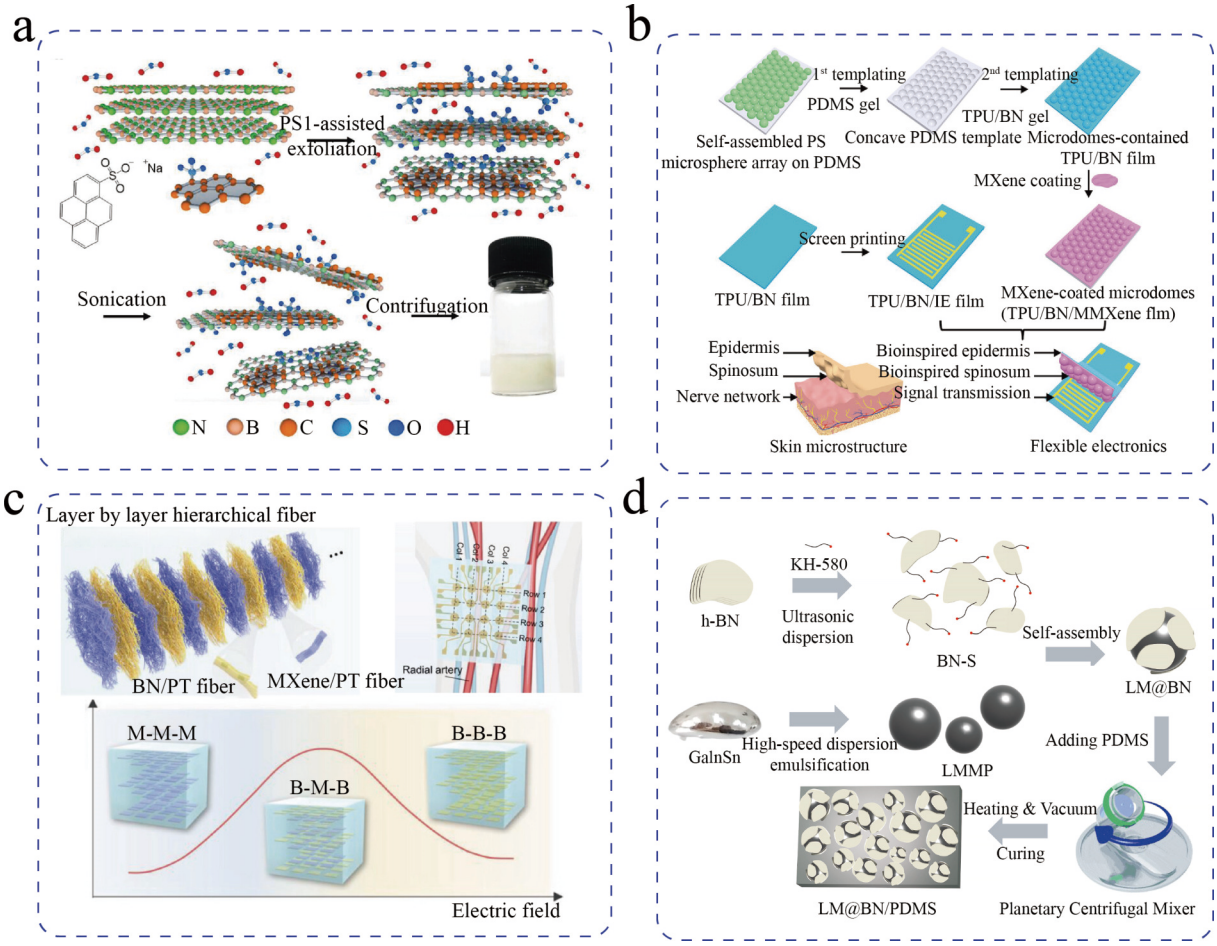

Fig. 4. (Color online) BN-based sensors. (a) Humidity sensor with BN as sensitive layer. Reproduced with permission from Ref. [89], Copyright 2024, Wiley. (b) Pressure sensor based on BN combining with MXene. Reproduced with permission from Ref. [91], Copyright 2024, Wiley. (c) Piezoelectric sensor with hierarchical structure for cardiovascular monitoring. Reproduced with permission from Ref. [92], Copyright 2024, Wiley. (d) Composites of liquid metal@BN with highly thermal conductive. Reproduced with permission from Ref. [93], Copyright 2023, Elsevier.

ple method that can be scaled up on large scale. In this multilayer structure, the excellent electrical conductivity of MXene improved the polarization of the interface, while the insulating property of BN enhanced the strength of the polarized electric field, largely reducing the risk of breakdown. Ultimately, the synergistic effect allowed the composite to withstand relatively large voltages. The 4 × 4 sensing arrays could continuously monitor blood pressure at the human wrist with accuracy comparable to that of commercial sphygmomanometers. In addition, machine learning simplified the tediousness of data processing, greatly saving workload[92]. The combination of continuous monitoring and machine learning facilitates the subsequent clinical application of flexible sensors. Antonova *et al.* obtained biosensors to detect glucose concentration, which consisting of graphene, boron nitride as well as PEDOT:PSS. When sweat accumulated, the charge accumulation raised and produced strong current response. In addition, the sensors also responded to humidity due to sweat accumulation. Importantly, the current response of the sensor was stronger than that of the sensors composed of graphene and PEDOT:PSS[94].

Although boron nitride has good thermal conductivity, it is not stretchable. Liquid metal (LM) as a class of new material with flexibility and processability has received the favor of researchers, but its thermal conductivity is lower than that of BN[93]. Therefore, Wang *et al.* prepared thermally conductive composites by utilizing the high thermal conductivity of BN combined with the deformable liquid metal, as shown in Fig. 4(d). The BN nanosheets were modified by sulfhydryl-containing surfactant and then composited with liquid metal particles to obtain the shell-core structure, named LM@BN, which was then mixed with the elastic polydimethylsiloxane (PDMS) to obtain flexible thermal management devices. The thermal conductivity of the composite was optimized when the volume content of LM@BN was 50%. In addition, the composite could also be employed as dielectric material because the LM was wrapped by the BN and its own oxide layer, and there was no continuous conductive pathway[93]. In other words, this multifunctional material can be excavated in fields such as robotics, which promotes the development of soft robots.

2.4. MoS_2 and $MoSe_2$

For flexible sensors, the sensitivity, accuracy and repeatability of semiconductors are superior compared to composites broadly employed consisting of conductive fillers and polymers[95–97]. Transition metal dichalcogenide (TMDs) compounds are favored by increasing number of researchers because they are equipped with bandgap and change from direct bandgap (usually single layer) to indirect bandgap (fewer layers and more layers) with the increase of the number of layers, which has bright prospect for the application in the field of sensing[98].

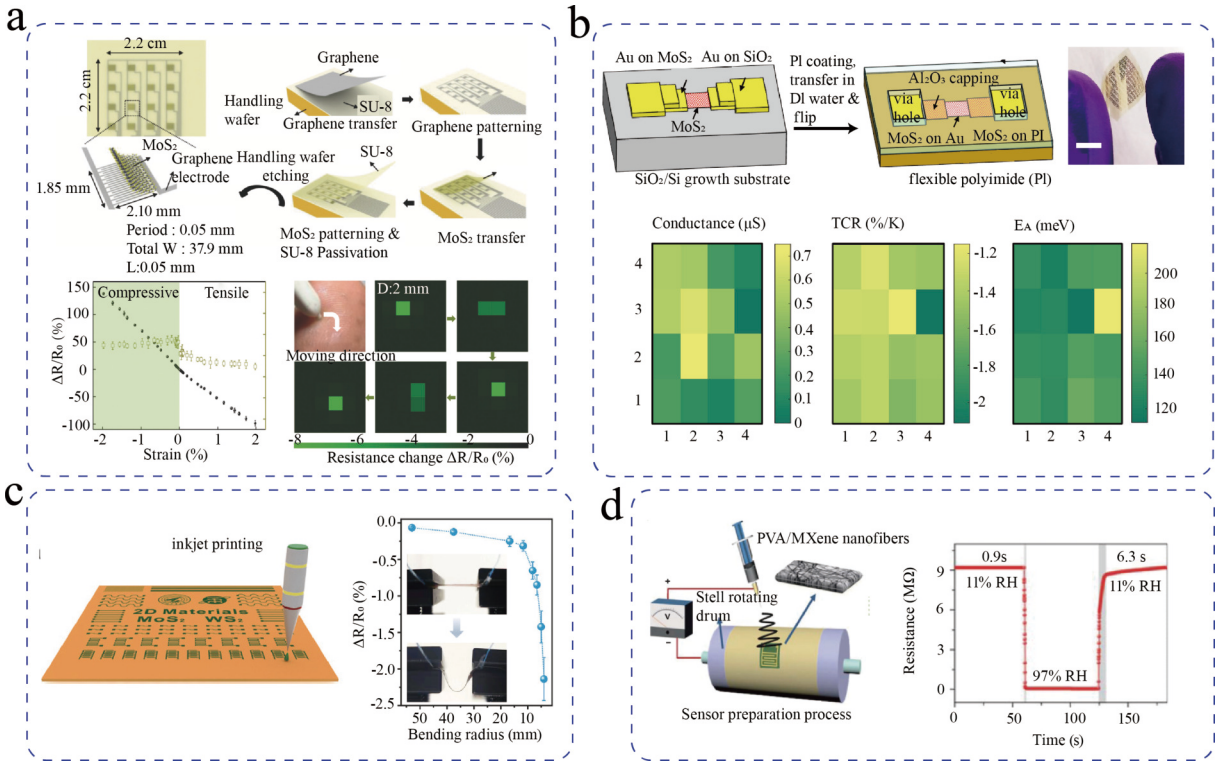

Fig. 5. (Color online) MoS_2-based and $MoSe_2$-based sensors. (a) Tactile sensor based on MoS_2. Reproduced with permission from Ref. [113], Copyright 2016, Wiley. (b) Temperature sensor based on monolayer MoS_2 channel. Reproduced with permission from Ref. [114], Copyright 2022, American Chemical Society. (c) Strain and tempture sensor based on MoS_2 by inkjet printing. Reproduced with permission from Ref. [115], Copyright 2023, Wiley. (d) Humidity sensor with PVA/MXene nanofiber. Reproduced with permission from Ref. [116], Copyright 2021, Springer.

MoS_2 exhibits favorable mechanical and electronic properties and is one of the most widely explored members of TMDs. The positions of the valence and conduction band edges of MoS_2 change with decreasing thickness and the indirect bandgap of the bulk material becomes the direct bandgap of the monolayer material[99]. The piezosensitivity coefficients of monolayer and bilayer MoS_2 are two orders of magnitude higher than those of graphene[100, 101]. The presence of piezoresistive and piezoelectric effects in MoS_2 makes it a promising candidate for ultra-low-power piezoelectric transconductance logic devices[102, 103].

In the initial studies, monolayer films were generally obtained by atomic layer deposition (ALD)[104–106], chemical vapor deposition[107, 108], physical vapor deposition (PVD)[109] growth, which were then transferred to flexible substrates to obtain flexible devices. Subsequently, Lin *et al.* proposed exfoliation of molybdenum disulfide crystals by electrochemical intercalation/exfoliation to obtain the monolayer film for the first time[110], which significantly improved the processability and assembly method of MoS_2, making it possible to assemble the film directly on the flexible substrate.

MoS_2 features high sensitivity and gauge factor as piezoresistive material, making it to be candidate for the sensitivity layer[111, 112]. Park *et al.* obtained patterned electrodes by transferring graphene grown by CVD as electrodes on a handling wafer covered with SU-8, followed by photolithographic processing, as shown in Fig. 5(a). The sensitive layer of MoS_2 was obtained through the same process. SU-8 was utilized as the encapsulation layer, and the overall thickness of the device was less than 75 nm, thus perfect fit was achieved on the human skin with excellent conformability. Under pressure, finger touch can be detected as the electrical signal changes[113]. MoS_2 exhibited high temperature coefficient of resistance (TCR) in addition to piezo-resistivity, which allowed it to be utilized for temperature sensors. Daus *et al.* prepared 4 × 4 temperature sensing arrays on PI film by transfer method, as shown in Fig. 5(b). The TCR of the sensor was about 1%–2% K^{-1} between 27–120 °C with a response time of 36 μs. It is worth mentioning that the response time was limited by integrated Ti/Pd microheaters and packaging of the device, so the response of the MoS_2 was much faster. This was more than 100 times faster than conventional temperature sensors consisting of metals and polymers. The thickness of the total devices encapsulated by Al_2O_3 was about 35 nm, showing excellent stability. This greatly improves the responsiveness of the temperature sensor and promotes its application in healthcare and wound healing[114].

Complicated transfer processes usually degrade the quality of the film, which affects the performance of the device. In addition, device preparation is inefficient and costly. Li *et al.* proposed simple and efficient approach for preparing large-area patterned MoS_2 films directly on flexible substrates by inkjet printing, as shown in Fig. 5(c). Large-area patterned films of MoS_2 were obtained by inkjet printing the precursor solution of MoS_2 and then annealing. The films were utilized to detect deformation and temperature changes because the resistance changed. In addition, due to its sensitive resistance change, the sensor could also be applied to monitor electrocardiography (ECG) and electromyography (EMG)[115].

Jung *et al.* realized NO_2 gas sensors by hybridizing MoS_2 and graphene. The sensitivity was four times higher than the graphene-based sensors because the high defect density of

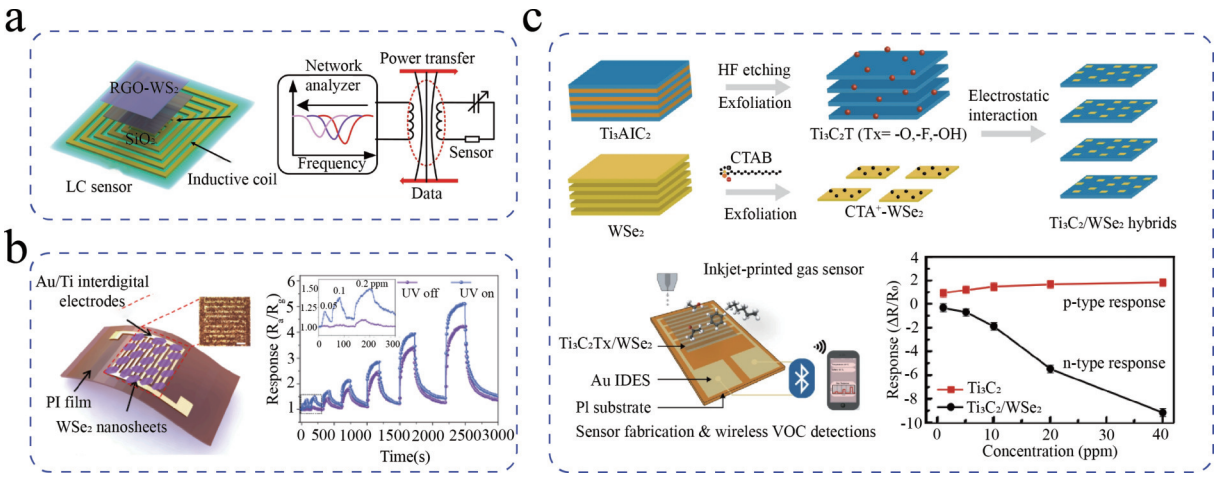

Fig. 6. (Color online) WS_2-based and WSe_2-based sensors. (a) Humidity sensor based on rGO-WS_2 heterojunctions. Reproduced with permission from Ref. [131], Copyright 2021, Elsevier. (b) Gas sensor for detecting NO_2. Reproduced with permission from Ref. [132], Copyright 2021, Elsevier. (c) Hybrid sensor for ethanol detection. Reproduced with permission from Ref. [133], Copyright 2020, Nature Publishing Group.

MoS_2 compensated for the low defect density of graphene. The sensitivity performance of the sensor remained unchanged at the bending radius of 14 mm when transferred to polyethylene glycol terephthalate (PET) substrates[117]. Liu *et al.* designed P/N junction for NH_3 gas sensors based on polyaniline/MoS_2. SnO_2 nanotubes were obtained by electrospinning followed by annealing, and then the surface was sequentially covered with MoS_2 and *in situ* polymerized aniline. The P/N junction ensured high sensitivity, and the resistance increased due to the compounding of NH_3 with the electrons on the surface of the polyaniline. The SnO_2 nanotubes provided high specific surface area for MoS_2, which enlarged the working range[118]. Analogously, Kim *et al.* possessed P/N junction for flexible chemical sensors (including NO_2 and NH_3) based on MoS_2/single-walled carbon nanotubes (SWCNTs). SWCNTs enhanced the adsorption of gases compared to single layer of MoS_2. The sensor can withstand 10^5 bending cycles on PET substrates[119].

$MoSe_2$ is also a generally exploited 2D material due to the principle that S and Se are from the same main-group elements. Wang *et al.* obtained a flexible piezoelectric nanogenerator (PENG)-based flexible humidity sensor (PEHS) by integrating $MoSe_2$ prepared by CVD on polyvinyl alcohol (PVA)/MXene hybrid electrospinning fibers, as shown in Fig. 5(d). The $MoSe_2$ converted mechanical energy into electrical energy to drive the humidity sensor. The response and recovery times of the whole device were 0.9 and 6.3 s, respectively. The prepared devices achieved energy harvesting and humidity detection in the human epidermis[116].

2.5. WS_2 and WSe_2

WS_2 and WSe_2, as an emerging type of TMDs materials, are also the most popular materials studied in the last two years[120–123]. These materials provide large surface area and favourable electrical properties, allowing more active sites, compared to one-dimensional (1D) materials such as CNTs[124, 125]. The crystal structure of WS_2 is layered, similar to graphene, and consists of alternating layers of tungsten and sulfur atoms. The bandgap of WSe_2 is located between the wide-gap semiconductor and the zero-bandgap graphene[126, 127]. This layered structure and wide bandgap significantly affect its electrical properties.

As the most important component in the human body, water affects human health directly. Therefore, there is an increasing demand for wearable humidity sensors. Guo *et al.* obtained flexible humidity sensor by transferring WS_2 to PDMS substrate. Pre-stretching and releasing yielded wrinkled structure between the sensitive layer and the electrode layer. The humidity sensor could detect 90% of the humidity range and the current changed when a finger was being close to it. And the mechanical bending was up to 5 mm, attributed to the wrinkled structure which strengthened the flexibility of the sensor[128]. rGO has a large number of functional groups, such as hydroxyl (–OH) and carboxyl groups (–COOH)[129, 130]. Although there are no hanging bonds on the surface of WS_2, it can form heterojunction with P-type materials as the N-type material, which can improve the electron mobility (Fig. 6(a)). Zhang *et al.* prepared humidity sensors based on heterojunctions of rGO and WS_2 by utilizing SiO_2 as the dielectric layer, which was achieved by combining multilayers of rGO with WS_2 composite. The electrons generated by the reaction between the functional groups on the surface of rGO and water were transported through the heterojunction, which improved the electron mobility and the sensitivity of the sensor. With the synergistic effect, the humidity sensor operated from 5% to 95% with response time of up to 0.56 s and sensitivity of up to ~140 kHz/% relative humidity (RH). The device showed excellent stability and was explored for the monitoring of water vapor in human breath. In addition, the sensing signals would be transmitted wirelessly with the LC circuit, which promoted the application of wireless sensors for human physiological signals[131].

Wang *et al.* constructed heterojunction consisting of WS_2 and polyaniline (PANi) for the detection of NH_3. polyaniline was present at the edges and in the middle of the layer of WS_2, providing more active sites for NH_3 adsorption. Therefore, the response time and speed of the heterojunction-based sensor were significantly higher than those of the WS_2-based and PANi-based sensors[134]. Qin *et al.* prepared NH_3 gas sensor based on S-defective WS_2. WS_2 containing S-defective adsorbed plenty of oxygen on the surface, and when NH_3 was adsorbed as electron donor, oxygen promoted the adsorption of more NH_3 as electron acceptor, and thus the sen-

sitivity was increased. In contrast, the NH_3 sensor prepared based on WS_2 crystals exhibited fewer surface defects and therefore less adsorption and lower sensitivity[135]. Nitrogen dioxide (NO_2) affects human life and health, such as respiratory diseases, as one of the most harmful pollutant gases today[136–138]. NO_2 exists hazardous when present in the air at levels above 200 ppd[139]. Therefore, trace detection of NO_2 has become one of the research hotspots. Yang *et al.* improved the sensing performance of WSe_2 by illuminating it with UV light, making full use of its excellent optoelectronic properties, as shown in Fig. 6(b). The sensor was more sensitive to NO_2, and down to 8 ppb when UV illuminated[132]. This promotes the development and application of WSe_2 flexible integrated sensors in the field of life and health.

In addition to NO_2 and NH_3, volatile organic compounds (VOCs) are also irreversibly harmful to human life and health[140]. Chen *et al.* quantified the volatile ethanol by constructing a hybrid structure of MXene/WSe_2 as shown in Fig. 6(c). Both MXene and WSe_2 were prepared by liquid-phase exfoliation, protecting the environment. In addition, both the sensing layer and electrodes were prepared by inkjet printing, which contributed to the advancement of large-scale fabrication of the devices. MXene compensated for the high surface resistance of WSe_2 that leads to electrical noise, and at the same time improved the gas responsiveness. The final sensor noise level with the composite material as the sensitive layer reached 0.15%, and the gas response and recovery time were 9.7 and 6.6 s. Compared with MXene, the sensitivity of the sensor based on the hybridized material was improved by a factor of 12. It is worth mentioning that the sensor on the PI substrate could work under bending conditions, which promotes the development of theinternet of things[133].

3. Summary and perspective

Sensing plays a determining role in human-machine interaction as a bridge between people and nature. In this review, we summarize different sensors with 2D materials as sensitive layers due to the different properties of each material. Sensors based on 2D materials can facilitate the development of sensors toward more flexible and ultra-thin due to their atomic-level controllable thickness and solution-processable preparation methods. However, there are still some problems that must be faced.

(1) Graphene has excellent mechanical and electrical properties as one of the most frequently utilized 2D materials. Due to the complexity of the preparation process, there is still necessary to study methods to reduce costs, so as to the graphene-based sensors can be promoted from the experimental stage to the stage of mass production.

(2) MXene has been widely broadly exploited in pressure sensors, due to its large specific surface area and abundant surface functional groups, but currently its poor environmental stability leads to low material utilization and poor sensor stability. The functional groups at the edges of MXene can be modulated to reduce the degree of oxidation at the edges, thus preventing its diffusion to the interior. In addition, although MXene is the accordion-like layered structure, current MXene-based pressure sensors are designed by combining MXene with microstructures. Therefore, there is significant need to expand the layer spacing of MXene. For example, intercalating the suitable molecules to improve the utilization of the laminar structure as well as the sensitivity of the sensor.

(3) BN is one of the ideal candidates for humidity sensors due to its excellent stability and large surface area that can be modified with a large number of functional groups. However, as the humidity sensor response time is usually on the order of seconds, heterojunctions can be involved to increase the electron mobility and therefore improve the response time. In addition, BN has a low conductivity and usually needs to be compounded with other materials to be available for resistive sensors, therefore, its surface can be modified to increase the conductivity.

(4) Further advancements are necessary in the research on TMDs as sensitive layers. Currently, the primary method of preparation involves CVD or PVD, which is not only costly but also environmentally polluting. Moreover, the fabrication of flexible sensors typically entails transferring the obtained materials to flexible substrates for assembly, resulting in numerous defects and significant contact resistance between the 2D material and the electrode. Additionally, crystal defects in 2D materials obtained through solution methods are relatively large, leading to reduced electron mobility and sensitivity. Therefore, there is urgent need to enhance the crystal quality of 2D materials prepared via solution methods to improve their electron mobility and sensitivity.

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