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Review

Formaldehyde Gas Sensors Fabricated with Polymer-Based Materials: A Review

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Abstract: Formaldehyde has been regarded as a common indoor pollutant and does great harm to human health, which has caused the relevant departments to pay attention to its accurate detection. At present, spectrophotometry, gas chromatography, liquid chromatography, and other methods have been proposed for formaldehyde detection. Among them, the gas sensor is especially suitable for common gaseous formaldehyde detection with the fastest response speed and the highest sensitivity. Compared with the formaldehyde sensors based on small molecules, the polymer-based sensor has higher selectivity but lower sensitivity because the polymer-based sensor can realize the specific detection of formaldehyde through a specific chemical reaction. Polymer-related formaldehyde sensors can be very versatile. They can be fabricated with a single polymer, molecularly imprinted polymers (MIP), polymer/metal-oxide composites, different polymers, polymer/biomass material composites, polymer/carbon material composites, and polymer composites with other materials. Almost all of these sensors can detect formaldehyde at ppb levels under laboratory conditions. Moreover, almost all polymer nanocomposite sensors have better sensitivity than single polymer sensors. However, the sensing performance of the sensor will be greatly reduced in a humid environment due to the sensitive coating on the gaseous formaldehyde sensor, which is mostly a hydrophilic polymer. At present, researchers are trying to improve the sensitive material or use humidity compensation methods to optimize the gaseous formaldehyde sensor. The improvement of the practical performance of formaldehyde sensors has great significance for improving indoor living environments.

Keywords: formaldehyde detection; gas sensor; polymer; composite

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1. Introduction

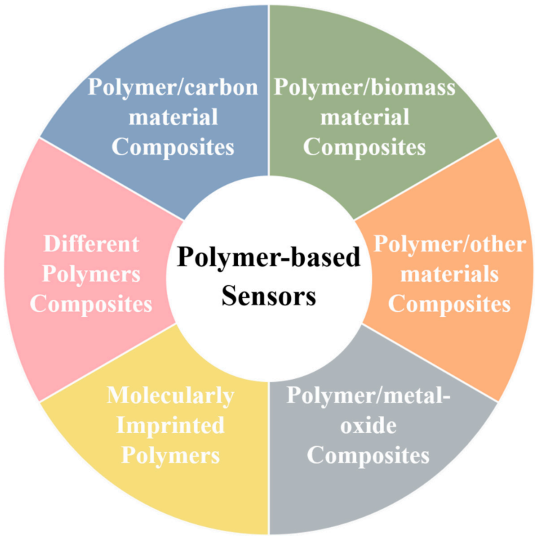
In society nowadays, the majority of human production and living activities are carried out indoors; that is, modern people usually spend 80–90% of their time indoors [1]. People's physical and mental health is directly affected by indoor air quality [1–3]. At present, more than 400 types of indoor pollutants have been identified, which is far more than human expectations [4–6]. Volatile organic compounds (VOC) are common indoor air pollutants and have a great impact on human health [7–9]. When the indoor concentration of VOC reaches a certain level, it will cause nausea, dizziness, vomiting, etc., in a short time, and even coma in a severe case [10–12].

Formaldehyde has penetrated all aspects of human production and life as a common VOC, which plays a significant role in the chemical industry, textile industry, anti-corrosion, and even cosmetics [13–17]. For ordinary families, indoor formaldehyde pollution mainly comes from indoor decoration [18]. For example, the main component of most adhesives used in artificial panels and furniture is formaldehyde [19]. In addition, the paint and coatings used in interior decoration also contain a lot of formaldehyde [20]. The residual

formaldehyde after decoration will gradually be released into the surrounding environment [21–23]. Generally, the formaldehyde content of newly decorated houses will exceed the standard by more than six times, and the formaldehyde release period will last for 3–15 years [24–26]. In 1995, the International Agency for Research on Cancer has already identified formaldehyde as a suspected carcinogen [27,28]; in 2004, formaldehyde was upgraded from a Class II carcinogen to a Class I carcinogen [12,29]; in 2010, formaldehyde was defined as one of nine indoor air pollutants in indoor air quality guidelines issued by the World Health Organization [10,16]. Besides, as a protoplasmic toxic substance, formaldehyde can be combined with proteins as well [30–32]. Inhalation of high concentrations of formaldehyde may lead to respiratory edema and induce bronchial asthma [33–36]. Direct skin contact with formaldehyde can cause dermatitis, necrosis, and even skin cancer [37–39]. And long-term exposure to formaldehyde will cause the decline of body function and poison the nervous system, cardiovascular system, and reproductive system to some extent [38,40–42]. Therefore, the real-time detection of gaseous formaldehyde in an indoor environment is very important for human beings.

Sensing technology is one of the commonly used methods to monitor indoor formaldehyde [10]. Compared with the colorimetric method, chromatographic analysis, spectrophotometry, and other methods for formaldehyde monitoring, the detection of formaldehyde by gas sensors has higher sensitivity and shorter reaction time [43,44]. Briefly, these sensors based on metal oxide semiconductor (MOS) material and polymer material are the most researched gaseous formaldehyde sensors [45–47]. Accurate identification of formaldehyde in complex atmospheric environments is a great challenge for MOS-based sensors [48–50]. Compared with the MOS sensor, the development of polymer sensors started late, but the development speed is also very fast [51,52]. The advantage of polymer sensors is that they can accurately identify formaldehyde based on specific chemical reactions.

At present, the superior sensing performance of polymer gaseous formaldehyde sensors has been verified in the laboratory. As far as we know, few people have reviewed the sensing mechanism and application of polymer sensors. Pan et al. [53] mainly described the application, advantages, and disadvantages of small molecule and polymer probes in monitoring formaldehyde in biological systems. In this paper, the research progress of gaseous formaldehyde sensors prepared by polymer-based materials for indoor formaldehyde monitoring is reviewed. First, we briefly introduce the common sensing techniques for formaldehyde detection. Second, the formaldehyde sensors fabricated with single polymer sensors, different polymers, molecularly imprinted polymers (MIP), polymer/metal-oxide composites, polymer/biomass materials composites, polymer/carbon materials composites, and other polymer composites were discussed in detail in regard to their materials preparation, sensors fabrication, and performance advantages (Scheme 1).



Scheme 1. The types of gaseous formaldehyde sensors based on polymers and polymer composites.

2. The Sensing Techniques for Formaldehyde Detection

Currently, resistance sensors and quartz crystal microbalance (QCM) sensors are the most common sensing types in the field of polymer and polymer nanocomposite sensors [54,55]. In addition, microelectromechanical system (MEMS) resonators, fluorescence probes, and other sensing techniques are also reported in some studies [56,57]. All of the above methods can be used to detect trace formaldehyde with high sensitivity [2]. The resistance sensor has the longest history, but its operation method is more complex [3]. The QCM sensor has the advantages of simple operation, high sensitivity, and low energy consumption [58]. At present, the QCM sensor is the most widely studied in the field of gaseous formaldehyde detection [59]. Compared with QCM sensors, MEMS sensors such as film bulk acoustic resonators have higher sensitivity due to the higher resonant frequency [60]. But now, the development of MEMS sensors is not mature enough, and the cost is high. Fluorescent sensors are used to visualize formaldehyde levels in living cells because of their good biocompatibility [61].

The resistance sensor is one of the most commonly used formaldehyde sensors with the longest history [62]. Further, the working principle of the resistance sensor is to detect the gas by recording the change in the resistance value when the sensitive material contacts the gas [63]. Good selectivity, high sensitivity, and low detection limit are the advantages of the resistance sensors [64].

In recent years, QCM has gradually replaced resistance testing and has become the most popular research direction in the field of organic polymer sensors for detecting gaseous formaldehyde [65,66]. QCM is a sensitive quality detection platform whose sensitivity is nanogram (ng) level [67]. In theory, the QCM can detect mass changes equivalent to a fraction of a monolayer or atomic layer, and the most basic principle of QCM is the piezoelectric effect of quartz crystals [68,69]. In 1959, Sauerbrey came to the conclusion that the resonant frequency change of QCM is proportional to the added mass on the quartz crystal [67,70]. On this basis, the Sauerbrey equation is summarized to represent the relationship between the mass adsorbed on the crystal sensor and the resonant frequency [29]. Based on the Sauerbrey equation, the QCM surface can be modified with different sensing materials to achieve highly sensitive detection of target gas [58]. The surface of QCM was modified with formaldehyde-sensitive materials [54]. Based on the Sauerbrey equation, the mass change on QCM is calculated according to the change frequency of QCM to realize the detection of formaldehyde gas [71]. As early as 2003, polyvinylpyrrolidone (PVP)-modified QCM has been used to determine ammonia [72]. Currently, the QCM sensor has been widely used in humidity, benzene vapor, formaldehyde vapor, and other detection fields [67,68]. Similar to QCM, MEMS are often used as mass-loading platforms when used as gas sensors. The sensitive layer is coated on the surface of the resonator to absorb the target gas molecules, and then the small mass change is monitored by the change of the resonant frequency [73]. Compared with traditional electro-acoustic resonators (such as QCM), MEMS resonators use 1–2 microns-thick piezoelectric films instead of crystal plates [74,75]. Therefore, MEMS resonators, such as thin-film volume acoustic resonators, have higher sensitivity, which has attracted wide attention in the field of gas detection [60,76].

Fluorescence is a cold luminescence phenomenon of photoluminescence [53]. In the ground state, there is no electron transition, that is, no fluorescence [35,77]. When the recognition site on the fluorescent molecule interacts with the analyte, the identified chemical signal is transmitted to the fluorophore through different signal transduction mechanisms [56]. The properties of fluorophore, such as the emission wavelength, intensity, or fluorescence lifetime of fluorescence, can be changed to realize the quantitative or qualitative detection of the measured object [78].

3. Formaldehyde Sensors Based on Polymers and Polymer Nanocomposites

This section may be divided into subheadings. It should provide a concise and precise description of the experimental results, their interpretation, as well as the experimental conclusions that can be drawn.

The aldehyde group can produce Schiff base by nucleophilic addition reaction with a primary or secondary amine under environmental conditions. Most polymer formaldehyde sensors rely on the reaction principle to select sensing materials. Conductive polymers rich in amines, such as polyaniline (PANI) and polypyrrole (PPY), are usually used as the selection layer of chemical resistance sensors to monitor formaldehyde vapor [79,80]. Some common ammonia-rich polymers, such as polyethyleneimine (PEI) and polydopamine (PDA), are often used as sensitive coatings for QCM and MEMS sensors to achieve the detection of formaldehyde vapor.

3.1. Formaldehyde Sensors Based on a Single Polymer

3.1.1. The Resistance Formaldehyde Sensor Based on a Single Polymer

PANI films prepared by plasma polymerization have a small specific surface area, low sensitivity, and poor selectivity to target gas [81]. To solve these above-mentioned problems, Sriniveset et al. [82] reported an amine-functionalized PANI nanofilm sensor. The reaction principle of this type of sensor is that the aldehyde group can react with the amine group on the sensitive layer of the sensor by a nucleophilic addition reaction, and the reaction products are H_2O and Schiff base without pollution. The formation of reaction products can change the resistance of PANI film. According to the principle, the functional relationship between the concentration of formaldehyde and the change in PANI resistance can be determined so as to calculate the concentration of formaldehyde to be measured. The lysine functionalized PANI nanofilm sensor shows good sensing performance, the limit of detection (LOD) for formaldehyde is as low as 400 ppb, and simultaneously, this formaldehyde sensor also has good selectivity for acetaldehyde, acetone, and formic acid.

3.1.2. QCM Formaldehyde Sensors Based on a Single Polymer

In 2016, Yan et al. [83] reported a QCM trace formaldehyde sensor based on thick-walled PDA nanotubes. The synthesis procedure of PDA nanotubes is shown in Figure 1. ZnO nanorods were used as templates, and the dopamine molecule containing the catechol and amino group was self-assembled onto the surface of the ZnO nanorod. Subsequently, the ZnO template was removed in NH_4Cl solution to form PDA nanotubes. The thickness of PDA nanotubes can be manipulated by adjusting the polymerization time. The formaldehyde sensor was constructed by coating PDA nanotubes on a QCM. Compared with amorphous granular PDA materials, PDA nanotubes have higher sensitivity, stronger selectivity, faster response speed, and lower LOD of less than 100 ppb.

3.1.3. MEMS Resonator Formaldehyde Sensor Based on a Single Polymer

Considering the high sensitivity of MEMS systems such as FBAR, Chen et al. [57] constructed a formaldehyde sensor based on a micro-electromechanical thin-film bulk acoustic resonator and nanostructured sensitive fiber. PEI nanofibers were deposited directly on the surface of a resonator by electrospinning as the sensitive layer of the sensor. The 3D nanofibers provide a large specific surface area for the adsorption and diffusion of formaldehyde vapor. The thin-film acoustic resonator, as a sensitive mass-loading platform, can detect the slight mass change after the amine group on the PEI react with formaldehyde. The sensor is capable of rapid response with LOD of formaldehyde as low as 37 ppb. Arabi et al. [73] also used a MEMS system to construct a gaseous formaldehyde sensor. This study mainly explored the desired sensitivity and selectivity of PANI and poly (2, 5-dimethylaniline) (P25DMA) to formaldehyde and benzene. The PANI and P25DMA sensors can achieve selective detection of formaldehyde in the presence of benzene with higher sensitivity and selectivity.

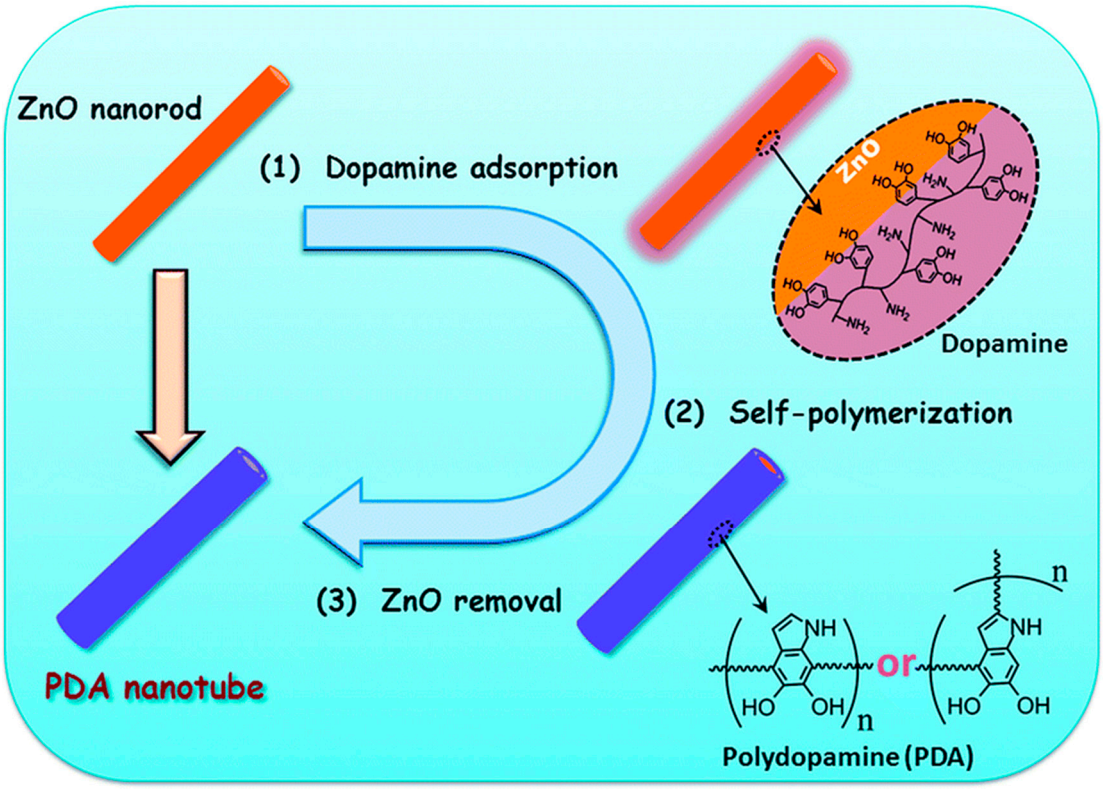


Figure 1. Roadmap of the synthesis procedure of PDA nanotubes [83]. (Copyright (2016) Royal Society of Chemistry).

3.2. Formaldehyde Sensors Based on MIP

MIP refer to polymers that have specific recognition and selective adsorption of specific target molecules without the interference of their similar structural analogs, which are synthesized by molecular imprinting technology [29]. Polyakov successfully used molecular imprinting technology for the first time in the study of the polymerization of sodium silicate and ammonium carbonate in 1931 [84]. It was not until 1972 that Wulff and Klotz introduced molecular imprinting into organic polymerization [85]. They found that the target molecules could be identified by covalently introducing functional groups into the imprinted cavity of the polymer [86]. In recent years, molecularly imprinted technology has been widely used in the recognition of small molecules, VOC, and proteins due to the excellent selectivity of MIP [87].

3.2.1. The Resistance Formaldehyde Sensor Based on MIP

After the study on a formaldehyde sensor composed of PANI and 4-amino-3-pentene-2-ketone (fluoro-P) by Carquigny et al. [27], Antwi-Boampong et al. [88] designed a novel formaldehyde sensor, which mainly used the specific Hantzsch reaction between formaldehyde and fluoro-P to identify formaldehyde vapor. Then molecular imprinting technology was used to combine PANI and ammonia to specifically encode the ammonia produced by the fluorine-P/formaldehyde reaction. The interdigitated electrode assembly was used to selectively detect formaldehyde vapor. In the study, molecular imprinting technology was applied to the double-layer platform for the first time, which improved the anti-interference performance of the sensor and made it more selective to formaldehyde. In addition, the sensor can realize real-time detection of formaldehyde vapor with good sensitivity and pleasant LOD as low as 30 ppb.

3.2.2. QCM Formaldehyde Sensors Based on MIP

In 2005, Feng et al. [28] first combined the MIP with a piezoelectric method to detect formaldehyde vapor at low concentrations. Based on the selective recognition of target molecules by molecular imprinting binding sites, the sensor can selectively recognize formaldehyde. The research has opened up a new development direction for the subsequent development of gaseous formaldehyde sensors. Based on previous studies on the MIP–QCM sensors, Hussain et al. [89] designed a copolymer film composed of styrene, methacrylic acid, and ethylene glycol dimethacrylate based on molecular imprinting technology. And then, the copolymer film was attached to the surface of QCM to detect formaldehyde vapor in the air. In dry air, the LOD of formaldehyde is 500 ppb in the presence of interfering gases such as methanol, acetic acid, dichloromethane, etc. When the relative humidity in the air was up to 50%, formaldehyde could not be recognized effectively due to the H_2O absorption saturation of the MIP surface. In order to improve the sensitivity of MIP coatings to formaldehyde vapor in a humid environment, the primary amino groups were introduced into MIP through allylamine, which transformed the morphology of the coatings into nanoparticles. And then, the modified sensor can detect formaldehyde in ppm levels in a humid environment. However, the sensor needs further functional modification to improve its sensing performance in a humid environment. Ultrathin MIP layers with ultra-high sensitivity can be obtained by deposition on porous materials. Titanium dioxide nanotube arrays (TiO_2-NTA) are widely used due to their abundant porous structure and low synthesis cost. On this basis, Tang et al. [64] designed a formaldehyde sensor based on polypyrrole molecularly imprinted polymers (PPY-MIP). The PPY-MIP layer deposited on the TiO_2-NTA has an orderly fishing net structure, and the thickness is only 20 nm. The special structure can minimize the gas diffusion resistance and further improve the binding ability of the PPY-MIP layer to formaldehyde. The sensing performance of the sensor is very stable, and it can even work stably in a humid environment. In order to further improve the specific surface area of MIP coating, Yang et al. [85] introduced a subsequent lithographic micro/nanoimprinting method to prepare formaldehyde imprinted hemispherical pore-patterned thin film composed of poly(2(trifluoromethyl)acrylic acid-co-ethylene glycol dimethacrylate-co-styrene) (poly (TFMAA-co-EGDMA-co-ST)), and subsequently, the thin film was combined with QCM to obtain a formaldehyde vapor sensor. The assembly process of the sensor and the characterization of the ultrathin concave hemispherical MIP film are shown in Figure 2. The results showed that the sensitivity of porous MIP film to formaldehyde (0.132 mg g^{-1} ppm^{-1}) was much higher than that of the porous non-imprinted polymer film (0.05 mg g^{-1} ppm^{-1}). In the presence of toxic gases such as hydrogen chloride and hydrogen fluoride, porous MIP film had higher selectivity than non-imprinted polymer film. The study also shows that the porous MIP surface composed of nanopattern features is very suitable for gas sensors.

Furthermore, most sensors have investigated the selectivity and sensitivity of target gas while ignoring the problem of response time. Considering the application of formaldehyde vapor sensors in real life, Lqbal et al. [86] designed a low-temperature imprinted poly(methacrylic acid) (imp-PMAA) using molecular imprinting technology for formaldehyde vapor. Besides, the prepared imp-PMAA and core-shell gold nanoparticles were assembled layer by layer to prepare imp-PMAA/Au-NPs sandwich or hybrid layers with ordered structure and adjustable thickness (100 $\pm$ 20 nm). The prepared imp-PMAA/Au-NPs mixed layer was coated on the surface of quartz microbalance (QMB) to detect formaldehyde vapor. As shown in Figure 3, compared with non-imp-PMAA, imp-PMAA, non-imp-PMAA/Au-NPs layers, and other tested materials, the response of the imp-PMAA/Au-NPs hybrid layer to formaldehyde increased by 2–9 times (when the concentration of formaldehyde vapor is 1 ppm). The hybrid sensor shows ultra-high selectivity to formaldehyde on account of the non-covalent dispersion interaction between the formaldehyde molecule and the molecular recognition site. More importantly, the LOD of the imp-PMAA/Au-NPs hybrid sensor is as low as 152 ppb, and both response (28 s) time and recovery (13 s) time are very short.

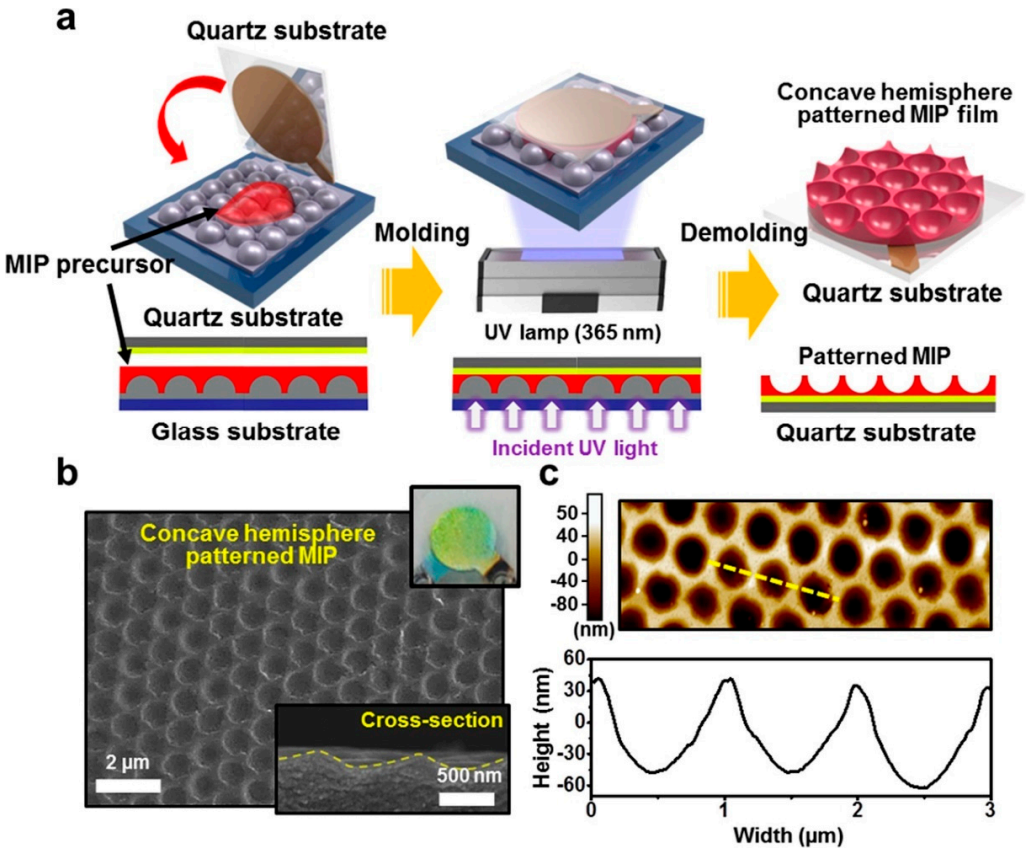


Figure 2. (**a**) Flowchart of the step and flash imprint lithography procedure employed to form pore-arrayed MIP films on quartz crystals. (**b**) Representative scanning electron microscopy image of the surface of the concave hemispherical patterned MIP film (upper inset: a digital image of quartz crystal microbalance—based MIP sensor; lower inset: a cross—sectional view of the ultrathin MIP film. (**c**) Atomic force microscopy image of the concave porous MIP film (top) and corresponding height profile [85]. (Copyright (2021) ELSEVIER).

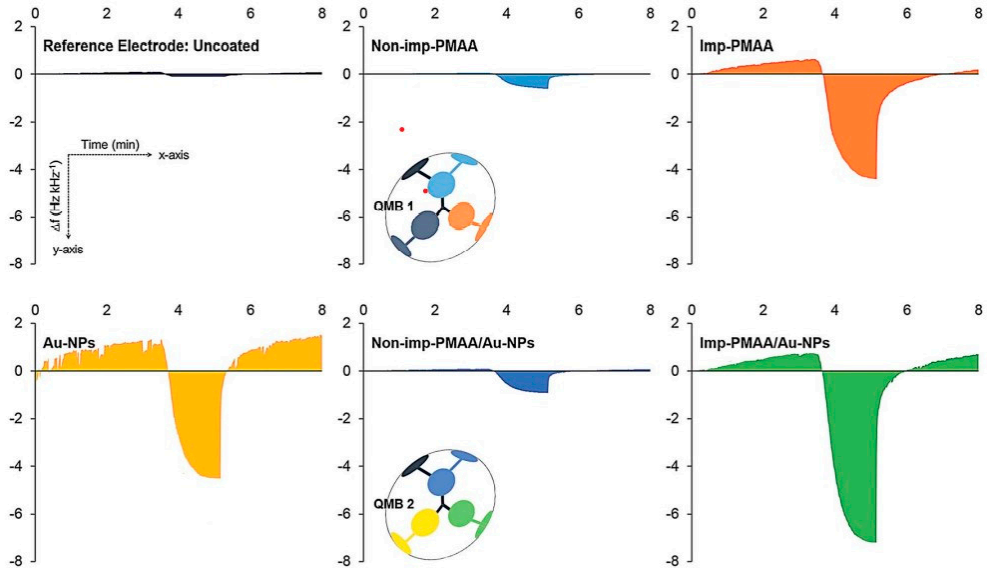


Figure 3. Control experiments: time-dependent frequency response of 2 tri—electrode QMB devices coated with different sensing materials toward 1 ppm formaldehyde gas at 25 °C and 50% RH. These sensor responses are normalized to 1 kHz or 40 nm of layer thickness [86]. (Copyright (2014) Royal Society of Chemistry).

3.3. Formaldehyde Sensors Based on Polymer/Metal-Oxide Composites

Up to now, a variety of metal oxides (such as titanium dioxide (TiO_2), zinc oxide (ZnO), indium oxide (In_2O_3), etc.) have been used as formaldehyde-sensing materials [90,91]. Compared with the single metal-oxide sensor, polymer nanocomposites (doped with metal oxide) can be customized according to the target analyte, such as formaldehyde. In addition, the polymer nanocomposites can operate at room temperature and improve the sensitivity and selectivity of the target analyte.

3.3.1. The Resistance Formaldehyde Sensors Based on Polymer/Metal-Oxide Composites

Molybdenum oxide (including MnO_2, MnO_3) is the earliest known metal oxide used in the formaldehyde sensor based on metal-oxide polymer nanocomposites [80]. Itoh et al. [80,92] had done a lot of research on layered organic/inorganic hybrid thin-film VOC sensors. Firstly, poly(N-methylaniline) (PNMA) was selected as the organic sensing layer, and MnO_3 thin films were prepared by chemical vapor deposition. Then, the sodium ions of PNMA were exchanged into the MnO_3 interlayer through the intercalation process to form $(PNMA)_xMoO_3$ hybrid film. The typical organic/MoO_3 hybrid film (polyaniline/MoO_3) is more sensitive to formaldehyde than acetaldehyde, while $(PNMA)_xMoO_3$ hybrid film has the same sensitivity to acetaldehyde as to formaldehyde. It has proved that the performance of organic/MoO_3 hybrid films could be controlled by modifying the interlayer organic components. Besides, Itoh et al. [80] continued to study polymer nanocomposites based on PANI or poly(o-anisidine) (PoANIS) by the same process. The $(PANI)_xMoO_3$ and $(PoANIS)_xMoO_3$ hybrid films prepared by this method can respond to dozens of ppb concentrations of formaldehyde and acetaldehyde, which indicates that the layered organic/inorganic hybrid films are extremely sensitive to target gases.

Moreover, Stewart et al. [63] prepared four different polymer nanocomposites by adding four different metal-oxide nanoparticles of copper oxide (CuO), alumina (Al_2O_3), nickel oxide (NiO), and TiO_2 to P25DMA. The measurement of sensing performance shows that the type of metal oxides has a great influence on the morphology of sensing materials and the adsorption capacity of the analyte. Moreover, TiO_2-based polymer nanocomposites are most sensitive to formaldehyde. Based on the high selectivity of the TiO_2 sensor to formaldehyde and ethanol under UV irradiation, Jo et al. [90] designed a monolithic flexible chemical sensor consisting of a TiO_2 sensing film, zeolite imidazole framework (ZIF-7) nanoparticles, and polymer mixed matrix membrane (MMM). The unique sensor designed in the study sandwiches the TiO_2 film between two flexible layers (the lower layer of the polyethylene terephthalate substrate and the upper layer of the MMM layer), enabling the formaldehyde sensor to achieve the specific detection of formaldehyde even in the presence of ethanol. Under ultraviolet light at 23 °C, the sensor showed a high response, and the concentration of formaldehyde was calculated as 25 ppb. As shown in Figure 4, this gas sensor could work at room temperature for the detection of formaldehyde at ppm level, showing ultra-high selectivity (response ratio > 50) and response (resistance ratio > 1100).

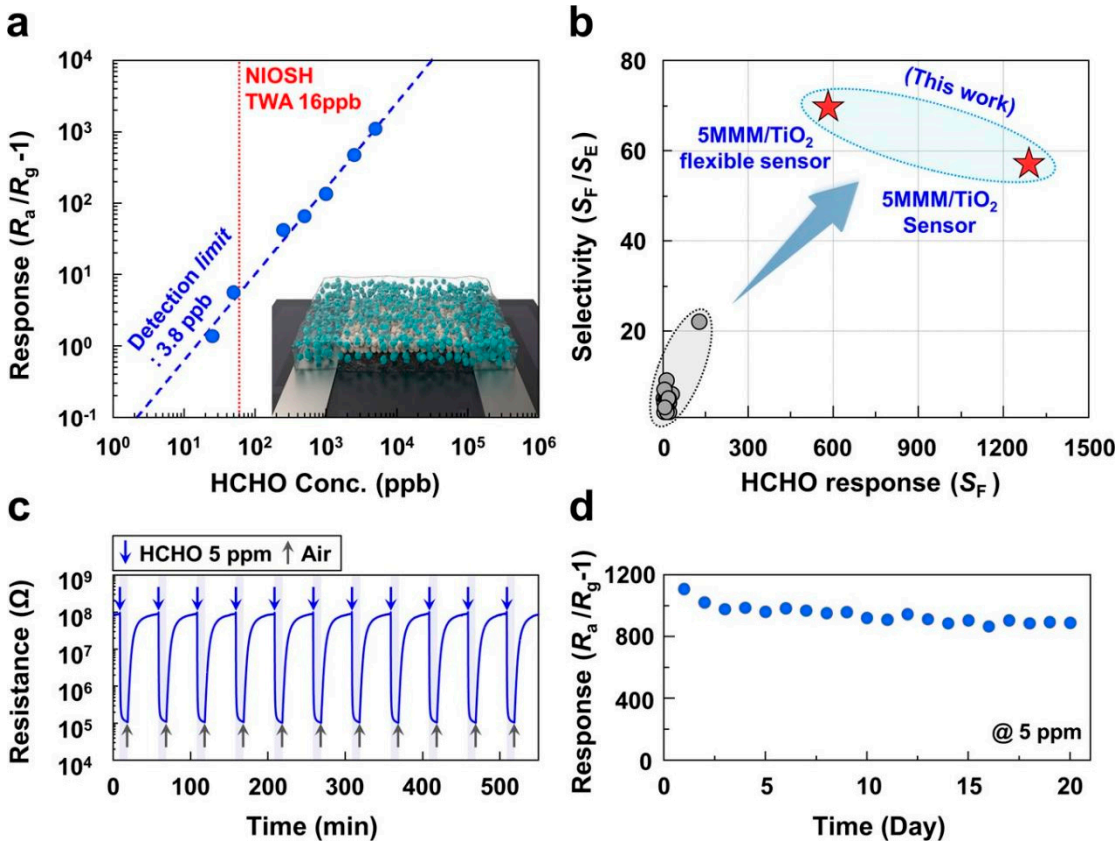


Figure 4. Low detection limit, formaldehyde selectivity/response, dynamic response, and stability. (**a**) Gas response as a function of formaldehyde concentration. (**b**) Formaldehyde selectivity (SF/SE) and response (SF) compared to the reported values in the literature. (**c**) Repeated sensing transients to 5 ppm formaldehyde at 23 °C under 365 nm UV radiation. (**d**) Long-term stability of 5 MMM/TiO_2 sensor (UV light illumination during sensor measurement) [90]. (Copyright (2021) Springer Nature).

3.3.2. QCM Formaldehyde Sensors Based on Polymer/Metal-Oxide Composites

Up to now, the research on QCM gas sensors based on polymer nanocomposites (doped with metal oxide) is relatively less. So far, Georgieva et al. [93] used the TiO_2 thin films deposited on QCM by liquid deposition method to detect NH_3. Due to the poor specific surface area of the film, the sensing performance is greatly limited. The detection sensitivity of the sensor is poor (the LOD is 100 ppm). After the above research, Wang et al. [71] developed a variety of novel PEI functionalized TiO_2 nanofiber complexes (PEI-TiO_2) as sensing coatings for QCM to detect formaldehyde. TiO_2 nanofibers with a high specific surface area were prepared by electrospinning method instead of flat film to enhance the sensing performance. And then, TiO_2 nanofibers are functionalized with PEI. The nanoporous TiO_2 fiber covered with the PEI layer has extremely high sensitivity and can provide the output signal of weight change when detecting formaldehyde. In general, QCM sensors have good selectivity and sensitivity, which can respond well to formaldehyde vapor at room temperature, with the LOD as low as 1 ppm.

3.3.3. Organic Thin-Film Transistor (OTFT) Formaldehyde Sensors Based on Polymer/Metal-Oxide Composites

Li et al. [78] prepared poly(3-hexythiophene) (P3HT)/ZnO organic-inorganic composite films and attached them to OTFT by spray-deposited method for the detection of formaldehyde at room temperature. Interestingly, the P3HT/ZnO hybrid thin-film OTFT sensor has a good sensing response to formaldehyde, and the sensor exhibited better-sensing performance when the weight ratios of P3HT/ZnO are 1:1 and 1:5. Tai et al. [56] further studied the approach to enhance the sensing performance and stability of the

P3HT/ZnO hybrid thin-film OTFT sensor. The effect of ZnO nanoparticles on sensing performance was systematically studied. Compared with the P3HT film sensor [94], the sensing response and reversibility of the P3HT/ZnO hybrid thin film are improved by more than two times. The transistor with organic-inorganic hybrid film has both the advantages of a thin-film transistor sensor and the excellent sensing performance of hybrid material, which can detect ppm levels of formaldehyde. The P3HT/ZnO hybrid thin-film transistor sensor provides a new idea for the development of formaldehyde sensing technology at room temperature.

3.3.4. Other Types of Formaldehyde Sensors Based on Polymer/Metal-Oxide Composites

In addition to the above-mentioned gas formaldehyde sensors, other sensing methods (such as ultraviolet-visible (UV-vis) spectrophotometry, electronic nose, and photonic crystals, etc.) have also been used for the detection of gas formaldehyde [23]. For example, Boersma et al. [95] reported a chemical sensor based on functionalized photonic crystals that can detect ppm levels of formaldehyde. The polymer device of the sensor is composed of TiO_2-UV resin nanocomposites with a nanoparticle fraction between 50–60%. The high refractive index polymer device prepared by the printing technology means that photonic crystals can be successfully transferred from the traditional dielectric or silicon material platform to plastic materials, which is a big step forward in the field of formaldehyde detection.

In recent years, UV-vis has also been used in the field of VOC detection, such as formaldehyde detection. Mazhar et al. [96] designed polytetrafluoroethylene (PTFE)/ZnO (PTFE-ZnO) composite nanofiber mats for VOC adsorption. The corresponding fabrication of the composite nanofibers is simple. Firstly, nanofiber mats are prepared by electrospinning in the precursor solution and then are treated at 280 °C for 5 min. The effects of different ZnO loads on the physicochemical properties of nanofibers are studied. Compared with pure PTFE, PTFE solution with ZnO added has higher conductivity [97]. The increase of the conductivity in the solution caused the formed nanofibers to have a thinner average diameter, stronger hydrophobicity, and higher surface roughness. The improvement of these properties has significantly boosted the adsorption efficiency of VOC. Meanwhile, the adsorptions of toluene, acetone, and formaldehyde on nanofiber mats were tested by UV-vis as well. The experimental results showed that the sample containing 20 wt.% ZnO had increased the adsorption of these test VOC by 10 times, seven times, and three times.

Additionally, Mavani et al. [98] evaluated the sensitivities of PANI and polyaniline derivative (poly(2,5-dimethyl aniline)) to formaldehyde by gas chromatography. Among them, PANI is more sensitive to formaldehyde. In order to further improve the sensitivity of the polymer skeleton to formaldehyde, different weight percentages of In_2O_3 were doped into PANI. And then sensing performance of the composite is further tested. The results show that PANI doped with 1.25 wt.% In_2O_3 is the most sensitive to formaldehyde. When the content of In_2O_3 increases to 5 wt.%, the selectivity of PANI to formaldehyde is still better than benzene. However, the trend of selectivity and sensitivity is the opposite. The best sensing material can be selected according to the specific application of the sensor.

Considering the practical application of formaldehyde sensor, Devadhasan et al. [99] designed a smartphone-coupled handheld array reader with integrated complementary metal oxide (CMOS) image sensor based on the colorimetric monitoring method. The sensor uses nano titanium (TiO_2-NPs) mixed with polyvinyl alcohol (PVA) hydrogel test strips to generate chemical reactions with toxic gases (such as hydrogen fluoride, chlorine, NH_3, and formaldehyde) to map dyes. Since the color of the dye can change according to the change of acid-base reaction, the color change can be monitored by a colorimeter and mapped in the form of data. The LOD of the sensor for the above toxic gas is only 1 ppm. In other words, the selectivity, sensitivity, and stability analysis can prove the reliability of the detection system.

3.4. Formaldehyde Sensors Based on Composites of Different Polymers

3.4.1. The Resistance Formaldehyde Sensor Based on Composites of Different Polymers

Sensors based on composites of different polymers have paid more attention to the detection of formaldehyde. For instance, Antwi-Boampong et al. [70] designed a PANI/PEI composite film as a conductive element and cast it on an interfinger electrode to detect gas formaldehyde. When exposed to formaldehyde vapor in the laboratory gas exposure room, the resistance of the film increases significantly. However, the reaction to other volatile organic vapors is weak. It can be shown that the combination of PANI and PEI improved the porosity and targeting of the composite films. The study provides a foundation for the development of organic elements in wearable sensors. In 2020, Wang et al. [100] developed a novel formaldehyde sensor by combining electrochemical analysis with functional nanochannels. Ethylenediamine (EDA) functionalized poly(ionic liquid)/polyacrylonitrile (PAN) nanofiber membrane was constructed as a formaldehyde-responsive ion microchannel. The formaldehyde detection process of the sensor is shown in Figure 5. The EDA fixed on the microion channel reacts with formaldehyde, the electron affinity of the microchannel increases, and the zeta potential decreases so that the output of the ion current can be switched from low to high. By establishing the linear relationship between the output of ion current and the concentration of formaldehyde, which concluded that the sensor has a high sensitivity to the formaldehyde in the concentration range of 360 ppm–0.036 ppt.

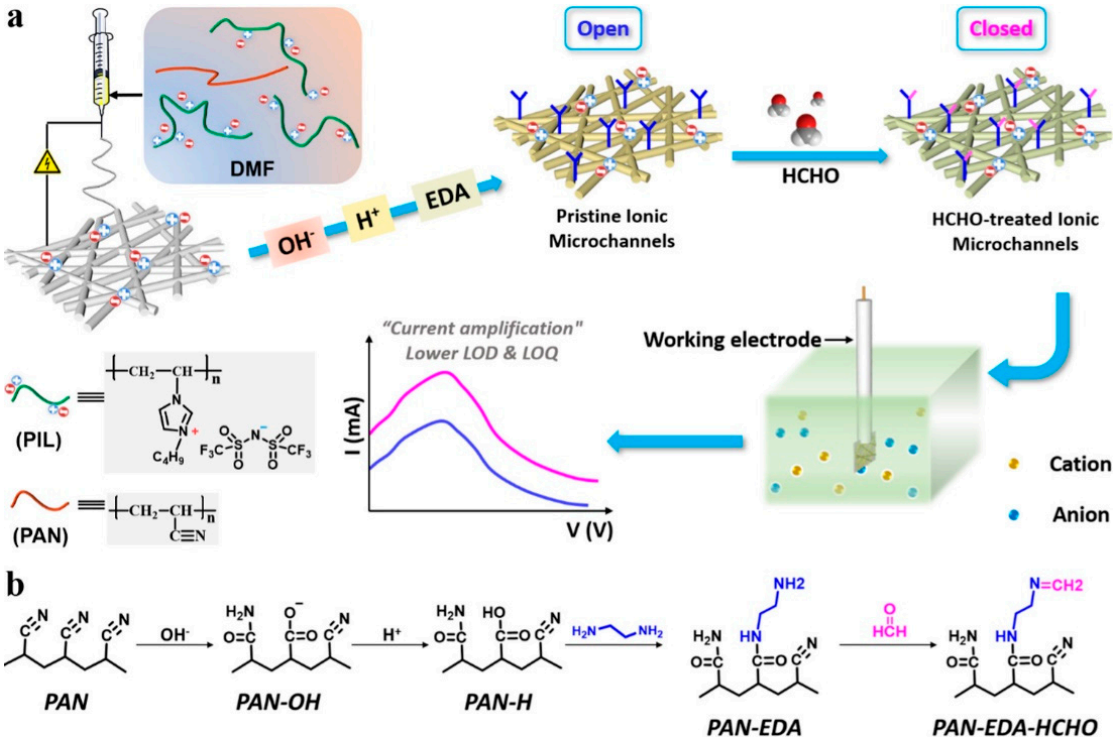


Figure 5. (**a**) Schematic illustration of ultrasensitive recognition of formaldehyde and stronger ionic current signals output of the designed ionic microchannels: the poly (ionic liquid) backbone inside of the ionic microchannels can promote a higher ionic current output. Moreover, when formaldehyde reacted with pristine ionic microchannels ("open state"), the surface could switch from "open" to "closed" (i.e., formaldehyde-treated ionic microchannels), and then the current signal outputs of formaldehyde-treated ionic microchannels were stronger. Therefore, it could be used to realize a lower LOD and lower limit of the quantity (LOQ) for formaldehyde determination. (**b**) The overall chemical reactions of the designed ionic microchannels with formaldehyde [100]. (Copyright (2020) American Chemical Society).

3.4.2. QCM Formaldehyde Sensors Based on Different Polymers

Wang et al. [67] designed a novel formaldehyde sensor by depositing nanofiber PEI/PVA films as sensitive coatings on QCM by electrospinning. The morphology of porous 3D PEI/PVA films could be controlled by adjusting the composition of the polymer and solvent in the PEI/PVA solution. The sensor makes use of the reversible interaction between formaldehyde molecules and PEI amino groups to achieve a rapid response to formaldehyde. The response of the sensor to formaldehyde shows good reversibility and reproducibility in the concentration range of 10–255 ppm operated at room temperature. When exposed to 255 ppm formaldehyde, the sensitivity of the QCM sensor with porous 3D PEI/PVA film as the sensitive coating is three times higher than that of the corresponding flat film-coated QCM sensor. The research proves that the sensing performance of gas sensors can be improved by changing the structure of the sensitive coating. Ding et al. [68] made further improvements and optimization based on the research of Wang et al. [67]. PEI functionalized polyamide6 (PA6) nanofiber/net (NFN) films were deposited on QCM for the first time by a simple electrospinning process as a simple room temperature formaldehyde sensing model. The innovation of this research is to obtain the NFN structure substrate of electrospinning nanofibers with large surface area, high porosity, and large bulk density by a simple electrospinning process and to improve the sensing performance of the sensor. When exposed to 100 ppm formaldehyde, the QCM-based PEI-PA6 NFN (30 kV) sensor achieves the maximum response value, which is about three times that of the PEI flat film-coated sensor. The LOD of the QCM sensor is as low as 50 ppb, while the sensor has high response speed, good selectivity, and good reproducibility. Zhang et al. [69] conducted a similar study in which the nanoporous polystyrene (PS) fibers with a large specific surface were deposited on QCM by the electrospinning method. Then, the fibrous PS membranes are functionalized with PEI. The morphology and the specific surface area of the PS membrane can be controlled by adjusting the concentration of the PS solution. The formaldehyde sensor can respond quickly at room temperature with a LOD of 3 ppm and can selectively identify formaldehyde in the presence of other interfering VOC. Huang et al. [101] also took the enlarged surface area of the sensitive coating on the QCM sensor as a starting point and modified the PAN nanofiber membrane with poly vinylamine (PVAm). The PAN nanofibers with rich primary amino groups prepared by electrospinning have a large specific surface area and rich hierarchical structure. Thanks to the above points, the prepared QCM HCHO sensor can detect the low concentration of formaldehyde (500 ppb) in a short time (200 s). When exposed to formaldehyde with a concentration of 500 ppb, the sensitivity of the sensor is 2.5 times higher than that of the sensor based on flat film.

Hydrophilic PDA nanotubes attached to QCM also can be used to detect formaldehyde vapor [83]. Since PDA is a hydrophilic polymer, H_2O will be adsorbed in a humid environment and give an error response. In order to overcome the influence of false responses caused by the increase in environmental humidity, Wang et al. [59] prepared a QCM formaldehyde sensor with a special structure by covering a superhydrophobic polymerized n-octadecyl siloxane (PODS) nanostructure on a hydrophilic PDA membrane. And the preparation process and corresponding workflow of the sensor are shown in Figure 6. The sensor based on PODS-PDA composite sensing material can maintain good stability when the ambient humidity increases. The study provides a new strategy for reducing false responses in a humid environment.

3.4.3. Other Types of Formaldehyde Sensors Based on Composites of Different Polymers

Considering the complex pretreatment steps and high cost of most gas sensing systems, Wang et al. [102] developed a polymer-based colorimetric sensor for core-shell nanoparticles. Uniform spherical polymer core-shell nanoparticles composed of poly (styrene-co-maleic anhydride) (PSMA)/PEI core-shell nanoparticles impregnated with methyl red (MR), bromocresol purple (BCP) or 4-nitrophenol (4-NP). In the presence of formaldehyde, MR, BCP, and 4-NP colorimetric sensors turn yellow, red, and gray, respectively, and the color changes are shown in Figure 7. The colorimetric reaction is the most obvious when

PEI/PSMA ratio is selected as 4:1. The MR colorimetric sensor can effectively detect gaseous formaldehyde in a 30% relative humidity environment. And the method can be used for the rapid detection of 0.5 ppm formaldehyde with the naked eye. The research provides a new development direction for real-time visual detection of gaseous formaldehyde.

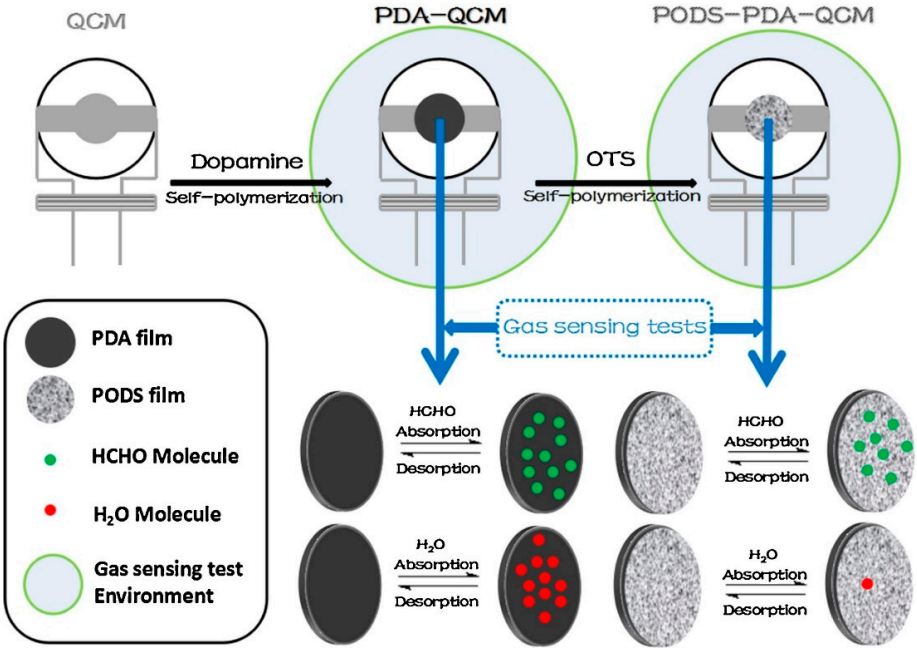


Figure 6. Structural diagram of PODS-covered PDA film-based formaldehyde sensor and the work principle of the sensor [59]. (Copyright (2018) ELSEVIER).

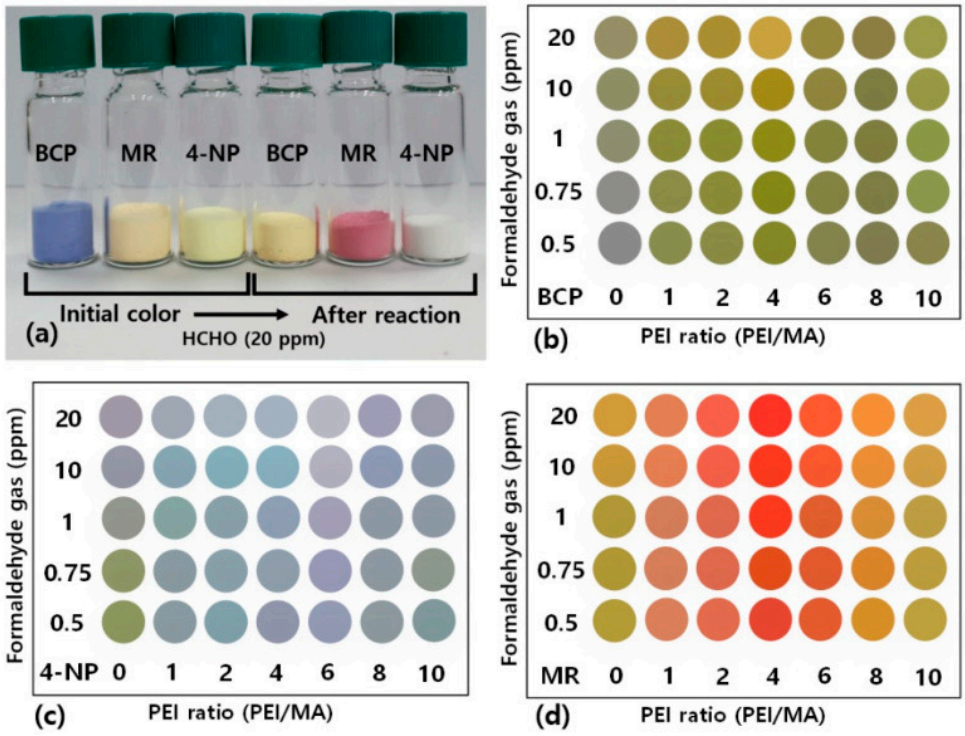


Figure 7. (**a**) Color changes observed by the naked eye for the bromocresol purple (BCP), 4-nitrophenol (4-NP), and methyl red (MR) colorimetric sensors when exposed to formaldehyde gas (20 ppm). Colorimetric responses (ΔRGB values) of (**b**) BCP, (**c**) 4-NP, and (**d**) MR sensors prepared with various PEI/PSMA ratios (0–10) on exposure to 20 ppm formaldehyde gas [102]. (Copyright (2020) Multidisciplinary Digital Publishing Institute).

3.5. Formaldehyde Sensors Based on Polymer/Biomass Material Composites

3.5.1. Formaldehyde Sensors Based on Polymer/Bacterial Cellulose (BC)

BC is a kind of nanoporous 3D fiber membrane with a high specific surface area (103 m^2 g^{-1}) and high porosity [103–105], which is an ideal material for further improving the sensitivity of formaldehyde sensors [106]. Hu et al. [65] combined nanofiber PEI with BC membrane for the first time to prepare a QCM gas sensor for detecting formaldehyde. The nanoporous 3D-PEI/BC films were composed of nanofibers ranging from 30–60 nm. Through the interaction between PEI and formaldehyde molecules, the selective recognition of formaldehyde can be realized. The sensor has good reversibility and repeatability for 1–100 ppm HCHO at room temperature. Wang et al. [107] further studied formaldehyde sensors based on PEI/BC double-layer nanofilm. The PEI/BC double-layer nanofilms were coated on ST-cut quartz substrate by sol-gel method and spin coating process. The filamentous and fibrous network structure of BC provides a large number of attachment sites for PEI, which greatly improves the sensitivity of the sensor and reduces the response and recovery time. Moreover, the surface acoustic sensor is more sensitive than the QCM sensor, and the response time is shorter, which further improves the performance of the sensor. The PEI/BC sensor was coated with three layers of PEI as the sensing layer showed the best sensing performance, and its LOD was 100 ppb.

3.5.2. Formaldehyde Sensors Based on Polymer/Chitosan

Chitosan is the product of natural polysaccharide chitin to remove part of the acetyl group, which has good adsorption capacity, biodegradability, and biocompatibility [108–110]. Wang et al. [111] coated QCM with PEI functionalized chitosan nanofiber network binary structure to afford a proper formaldehyde sensor (Figure 8). Chitosan fiber substrate was prepared by the electrospinning method, which was composed of nanofibers and spider webs. Then the chitosan fiber substrate was functionalized by PEI. The chitosan fiber substrate was composed of nanofibers and spider webs, which greatly improved the sensing performance. And there is a strong adhesion between the chitosan film and the QCM electrode. These characteristics give the formaldehyde sensor the advantages of fast response speed and low LOD (5 ppm) under environmental conditions. Li et al. [112] designed a robust hydrophilic hydrazine naphthalimide-functionalized chitosan (HN-chitosan) polymer probe. The polymer probe relies on the specific chemical reaction between formaldehyde and the grafted hydrazine-naphthalimide groups to trigger the fluorescence opening reaction. Compared with the small molecule analogs, the HN-chitosan polymer probe based on biopolymer chitosan can enrich low-concentration formaldehyde pollutants around the polymer chain and accelerate the chemical reaction between formaldehyde and the grafted hydrazine-naphthalimide groups. At the same time, the chemical reaction between formaldehyde and naphthol hydrazine groups can also be accelerated by the synergistic interaction between the multiple recognition sites of naphthol hydrazine and adjacent hydroxyl groups, allowing the obtained fluorescence probe to have high sensitivity and fast fluorescence response (less than 1 min).

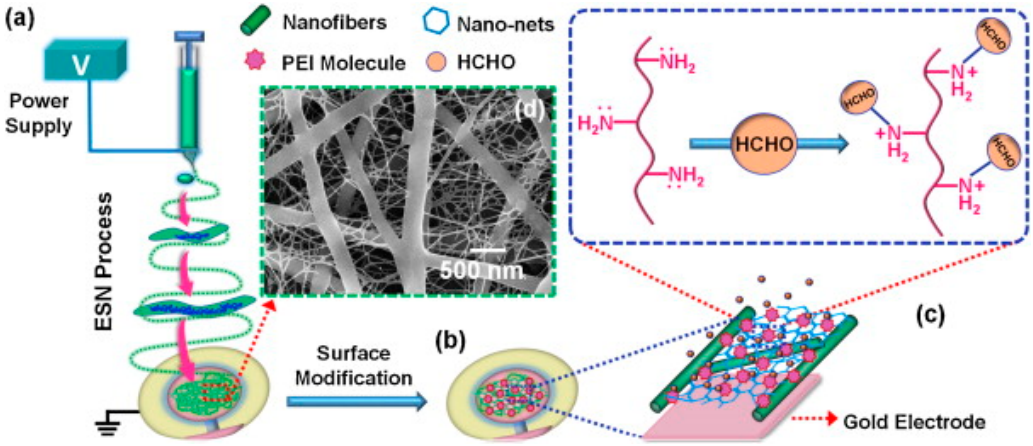


Figure 8. Schematic diagram illustrating the fabrication of sensing layers on QCM. (**a**) ESN deposition of fibrous membranes onto the electrode of QCM. (**b**) Surface modification of NNB with diluted PEI solutions. (**c**) Illustration of PEI-chitosan fibers on the gold electrode and the reaction mechanism between formaldehyde and PEI. (**d**) Typical FE-SEM image of NNB structured chitosan membranes [111]. (Copyright (2014) ELSEVIER).

3.5.3. Formaldehyde Sensors Based on Polymer/Lignin Composite

Lignin is the second largest biomass resource in the plant kingdom, whose storage capacity is only inferior to cellulose and a kind of complex organic polymer with rich aromatic rings and strong fluorescence [113,114]. Ma et al. [115] designed a PVA/cellulolytic enzyme lignin nanoparticles (CEL-NPs) nanocomposite film fluorescence sensor based on the fluorescence properties of lignin. This method of preparing aggregation-induced emission (AIE) nanoparticles CEL-NPs from CEL through simple one-step self-assembly successfully solves the complex problem of most AIE nanomaterials synthesis methods. Compared with the commercial 4′, 6-diamine-2-phenyl indole (DAPI) dyes, CEL-NPs exhibit temperature-dependent fluorescence, better resistance to photobleaching, and good stability in acidic and low-temperature environments. The fluorescence sensor has good sensitivity and can detect formaldehyde vapor at ppm level. Carbon quantum dots are widely used in the sensing field due to their excellent optical properties, good biocompatibility, low cost, and other advantages. Wang et al. [61] obtained alkali lignin (AL) from spruce and used it to prepare nitrogen-doped carbon quantum dots (NCQDs) with good photoluminescence properties. NCQDs with excellent photoluminescence properties are obtained from AL and m-phenylenediamine after a simple one-pot hydrothermal treatment. Under the optimum conditions, these NCQDs exhibit blue-green fluorescence with excitation/emission wavelengths of 390/490 nm. NCQDs and PVA composite films are combined as formaldehyde sensors. When the sensor was exposed to gaseous formaldehyde, the fluorescence intensity of the composite film surface increased, and the color gradually changed from blue-green to blue, showing that the fluorescence sensor has a good response to gaseous formaldehyde.

3.5.4. Formaldehyde Sensors Based on Polymer/B-Cyclodextrin (B-Cd) Composites

Cyclodextrin molecule has a slightly tapered hollow cylinder 3D ring structure, and the outer part is hydrophilic, while the hydrophobic region is formed in the cavity due to the shielding effect of the C-H bond [116,117]. Many functional groups can be chained to cyclodextrin molecules or cross-chained to polymers for chemical modification or polymerization with cyclodextrins as monomers [118,119].

Kadam et al. [120] used electrospinning to prepare nanofiber membranes of PAN and β-CD for capturing aerosols and VOC in the air. The addition of β-CD makes the nanofiber membrane thicker and the pore size larger. By changing the mass ratio of PAN and β-CD, the properties of PAN/β-CD nanofiber membranes are changed. The experimental results show that the pressure drop and mass coefficient of nanofiber films containing 30 and 45 wt.% β-CD are better than those of pure PAN. In general, when the content of β-CD is

30 wt.%, the composite nanofiber membranes have the best performance. The filtration efficiency of particulate aerogel can reach more than 95%, the pressure can be reduced to 112 Pa, and it has good adsorption performance for formaldehyde and xylene. Poly (5-carboxyindole) (P5C) has been used as the sensitive layer of electrochemical sensors to detect methanol, ethanol, acetone, and ether [81]. However, it has never been used to detect formaldehyde. And as a sensing material, P5C cannot detect target analytes with low concentrations (less than 1000 ppm). Hodul et al. [121] firstly applied P5C mixed with β-CD to a resonance mass sensor for detecting low concentrations of formaldehyde in a room. The LOD of the sensor is 25 ppm, and its response time (27 s) and recovery time (16 s) are very short. The study proves that P5C with β-CD can further improve sensor performance.

3.6. Formaldehyde Sensors Based on Polymer/Carbon Material Composites

3.6.1. Formaldehyde Sensors Based on Polymer/Graphene

Graphene is a 2D material with a single honeycomb lattice structure formed by the compact packing of sp^2 hybridized carbon atoms [122]. The unique 2D structure of graphene makes it very sensitive to the surrounding environment, and the sensitivity of graphene chemical detectors can be comparable to the limit of single molecule detection. Therefore, graphene is widely used in the sensing field [123,124].

According to the excellent sensing characteristics of graphene, Alizadeh et al. [20] designed a chemical resistance sensor based on graphene/polymethylmethacrylate (PMMA) composite film to detect formaldehyde vapor. Formaldehyde adsorbed on the sensing film leads to the resistance increase of the sensing film, thus realizing the detection of formaldehyde. The direct interaction between formaldehyde and graphene sheets is the direct cause of the observed reactions. And the graphene/polymer ratio is an important parameter affecting the sensing performance. The sensor with the best performance has a linear relationship with formaldehyde at 0.05–5.0 ppm level, and its lowest LOD is 10 ppb. Kongkaew et al. [125] dispersed ellipsoidal palladium nanoparticles (PdNPs) uniformly on polyacrylic functionalized graphene oxide (PAA-GO/GCE) modified glassy carbon electrodes (PdNPs-PAA-GO/GCE) as sensing materials. Cyclic voltammetry is used to characterize the properties of the sensor material, which proves that it has good electrocatalytic activity. Then the modified electrode is combined with flow injection amperometry (FI-Amp) for formaldehyde detection. The optimal analytical performance is obtained by optimizing the application potential, flow rate, and injection volume of PdNPs-PAA-GO/GCE. The sensor has good repeatability and stability (RSD = 1.5%, n = 500), and its LOD is 480 ppb. Chuang et al. [126] introduced printable technology into the field of formaldehyde sensing and designed a printable rGO/PMMA sensing material. The sensing material uses the energy band distortion caused by graphene and the adsorption of PMMA on formaldehyde to detect formaldehyde. And 2% graphene/10% PMMA is the best ratio for formaldehyde detection. The sensor has high selectivity for formaldehyde and can produce a 30.5% resistance change for 1000 ppm formaldehyde. The minimum LOD is 100 ppm, and the resistance change is 1.51%. This work explored a variety of applications for printable sensing materials.

3.6.2. Formaldehyde Sensors Based on Polymer/Carbon Nanotubes (CNT) Composites

CNT can be regarded as rolled graphene sheets, so they can be divided into single-walled CNT (SW-CNT) and multi-walled CNT (MW-CNT) according to the number of graphene sheets [127–129]. The mechanical properties, electrical conductivity, and flexibility of CNT are also pretty good [130]. CNT are often used to construct conductive structures of sensors due to their excellent electrical conductivity [131]. CNT have very weak physical adsorption of formaldehyde molecules. However, chemical modification can significantly increase their adsorption performance [132]. It is well known that increasing the specific surface area of the sensing layer can improve the performance of the sensor to a certain extent [133,134]. Based on this principle, Tai et al. [66] developed a PEI/MW-CNT nanocomposite thin-film QCM formaldehyde sensor. The sensing principle of the

sensor is that the amine functional group of PEI reacts with formaldehyde. The UV-vis can demonstrate the occurrence of the reaction. Compared with the pure PEI film sensor, the PEI/MW-CNT composite film sensor has a much stronger adsorption capacity. When the volume ratio of the PEI/MW-CNT is chosen as 1:1, the sensor has the best performance. The sensor can realize selective detection of formaldehyde at room temperature, with a LOD of 0.6 ppm, and the detection results have good reproducibility as well. Sachan et al. [135] designed a quantum resistance vapor sensor (vQRS) assembled from polyhedral oligomeric silsesquioxane (POSS) and PMMA or PS and CNT. In order to further improve the sensitivity of the sensor, CNT are introduced into the polymer nanocomposites because the organic-inorganic compounds have a greater specific surface area and flexibility than the organic homopolymers. Both sensors show high sensitivity to formaldehyde and ammonia in dry air, which can detect 300 ppb of formaldehyde and 500 ppb of ammonia. Surprisingly, the sensor can respond to formaldehyde and ammonia within 5 s.

In order to further improve the sensitivity of the sensor, Song et al. [76] chose the thin-film volume acoustic resonator as a sensing platform, which is more sensitive than the QCM sensing platform. SW-CNT with a high specific surface area are selected as sensitive coatings, in which CNT are modified with PEI to improve their selectivity to formaldehyde. The working frequency of the thin-film acoustic resonator is about 4.5 GHz, which can detect the small mass change of the sensitive coating after formaldehyde adsorption. The sensor has a reversible linear response to formaldehyde in the range of 50–400 ppb, with a fast response (less than 1 min) and low LOD (24 ppb). Wang et al. [74] selected PEI-modified MW-CNT to do a similar study. The influence of the spraying process on sensing performance is mainly studied. Nanocomposite films can be coated on the surface of the resonator by a simple spray process. The sensor shows a reversible linear response to formaldehyde in the range of 60–500 ppb, with a minimum LOD of 60 ppb. The performance of the sensor is slightly inferior to that of the thin-film bulk acoustic resonator sensor using PEI-modified SW-CNT as the sensitive coating. Wang et al. [75] made further improvements on the basis of the previous researchers by introducing the concept of layer by layer assembly. The PEI modifies CNT nanofilms were assembled layer by layer on the surface of the resonator as a sensitive coating (Figure 9). The resonant frequency is a function of the number of cycles of CNT/PEI, which decreases almost linearly. Compared with the single-layer film, the multilayer film has a random porous structure, which provides a larger specific surface area for formaldehyde adsorption and diffusion. The minimum LOD of the sensor is 24–38 ppb. And the sensor can respond within 1 min, which can realize rapid detection of formaldehyde vapor.

In order to improve the conductivity of polymer nanocomposites, many researchers introduce CNT into polymer nanocomposites. For example, Dai et al. [136] prepared poly(methacryloyl hydrazide) nanofibers with formaldehyde molecular recognition sites by electrospinning. CNT were introduced into the synthesized nanofibers to improve the conductivity of the nanofibers. Then the prepared composite nanofibers were used to make a biomimetic sensing platform. And the resistance method was used to detect formaldehyde. The biomimetic sensing platform has a good recognition ability for formaldehyde and can quantitatively measure low-concentration formaldehyde. The linear response range of the sensor is 1 ppb–10,000 ppb, and its LOD is 0.8 ppb. Ma et al. [60] used MW-CNT/PEI bilayer as the sensitive layer of ZnO piezoelectric thin-film resonator to detect trace gaseous formaldehyde. Formaldehyde molecules can be adsorbed on the MW-CNT /PEI bilayer structure through the nucleophilic reaction between formaldehyde molecules and amine functional groups on PEI. The addition of CNT has enhanced the conductivity of the sensitive layer and further improved the performance of the sensor. Moreover, the operating frequency of the resonator can reach 3.1 GHz, and the mass sensitivity is extremely high, which can sense the ultra-small mass change. The resonance frequency decreased linearly with the increase in formaldehyde concentration. At room temperature, the formaldehyde sensor showed a detection range of 50–400 ppb. It also has fast response speed, high sensitivity, good reversibility, and good repeatability.

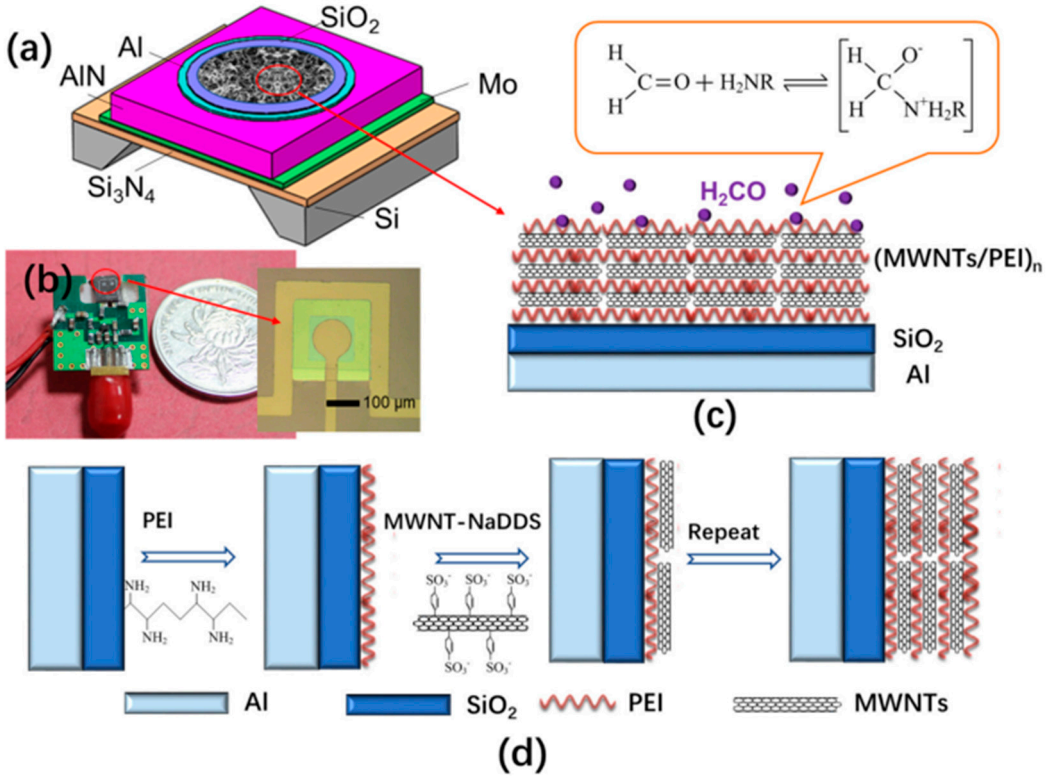


Figure 9. (**a**) Schematic structure of the FBAR sensor, (**b**) photographs of the fabricated device on a PCB, (**c**) (MWNTs/PEI) n multilayer assembled on the top of FBAR, (**d**) LBL assembly process of the (MWNTs/PEI) n multilayer [75]. (Copyright (2018) IOP Publishing Ltd., Bristol, UK).

3.7. Formaldehyde Sensors Based on Polymer Composites with Other Materials

In addition to the more common polymer nanocomposites used in the field of formaldehyde detection, other researchers have developed some novel nanocomposites for the field. For example, Khan et al. [137] prepared conductive PPY-zirconium selenide iodide (PPY/ZSI) cation exchange nanocomposite by sol-gel method. At 130 °C, the DC conductivity of the composite can still maintain good stability. The cation exchange nanocomposite is used as a formaldehyde vapor detection sensor at room temperature. With the increase of formaldehyde concentration, the resistance of the sensor also increases, and the sensor shows a good reversible response to formaldehyde vapor. Gil-Gonzalez et al. [31] prepared a room-temperature benzene and formaldehyde gas sensing system by polymerizing poly(N-isopropylacrylamide) on a gold fork electrode in the presence of 1-ethyl-3-methylimidazolium ethyl sulfate ionic liquid. With N_2 as the carrier gas, the sensor shows high sensitivity to formaldehyde. Both response and recovery time of the sensor are short. This study shows that gel-ion materials can be used for room-temperature VOC sensors. Darder et al. [138] used PMMA 3D-printed fiber as an optical waveguide in the field of formaldehyde sensing for the first time. The surface of the filament was coated with leuco fuchsin (LF)-doped Nafion perfluoro-sulfonated cladding. The performance of the formaldehyde sensing fiber was tested with a special photoelectric device. The detection limit of the optical sensor is 0.03–0.20 ppmv (0.037–0.25 mg m^{-3}). A portable prototype of the sensor has been tested for 4 months at a Spanish industrial paper impregnation process unit. Zong et al. [139] used hollow mesoporous silica spheres modified (HMSS) with PDA as sensing materials for the detection of formaldehyde in food. QCM was selected as the sensor platform to detect formaldehyde. The large surface area of HMSS provides advantages for gas diffusion and enough attachment sites for PDA. Moreover, the available hollow space and radial channels of HMSS make the sensor more sensitive. The sensor has a minimum LOD of 100 ppb and can respond (1.5 s) and reply (3 s) in a short time.

As shown in Table 1, almost all sensors are capable of detecting formaldehyde at ppb levels. From the perspective of the type of sensors, QCM sensors occupy the vast majority of gaseous formaldehyde sensors, while MEMS sensors have the highest sensitivity. From the perspective of the type of sensitive material, the sensing properties of almost all composites are superior to the single polymer.

Table 1. Formaldehyde sensor based on single polymer and polymer composites.

Types	Materials	Sensitive	Respond Time	LOD	References
Resistance	PANI	Not mention	Not mention	400 ppb	[82]
QCM	PDA	1400 Hz/ppm	8.3 s	100 ppb	[83]
MEMS	PEI	1,216,000 Hz/ppm	Not mention	37 ppb	[57]
Resistance	MIP	Not mention	2.5×10^{-5} s	30 ppb	[88]
QCM	MIP	Not mention	Not mention	500 ppb	[89]
QCM	MIP	Not mention	28 s	152 ppb	[86]
Resistance	PET/TiO_2	Not mention	Not mention	25 ppb	[90]
QCM	PEI/PA6	Not mention	<150 s	50 ppb	[68]
Colorimetric	PEI/PSMA	Not mention	60 s	500 ppb	[102]
QCM	PEI/BC	3560 Hz/ppm	Not mention	100 ppb	[107]
QCM	PEI/chitosan	Not mention	Not mention	5 ppm	[111]
Resistance	PMMA/graphene	Not mention	Not mention	10 ppb	[20]
MEMS	PEI/SW-CNT	Not mention	<60 s	24 ppb	[76]
MEMS	PEI/MW-CNT	6,220,000 HZ/ppm	Not mention	60 ppb	[74]

However, most of the polymers that can be used as the sensitive layer of formaldehyde sensors are hydrophilic polymers, which will have a negative impact on the performance of the sensor in a wet environment. But in recent years, few researchers have paid attention to this issue. In the relevant research mentioned in this review, covering the gas-sensitive layer with hydrophobic polymer is the best method to improve the performance of gaseous sensors in humid conditions. However, this method will reduce the sensitivity of the sensor. Therefore, reducing the influence of the hydrophobic layer on the sensing performance or seeking new methods to improve the performance of gaseous formaldehyde sensors in a humid environment can be regarded as a breakthrough point in future research. Liu et al. [140] proposed a small-sample local Gaussian process regression method in the latest research on polyaniline/cerium dioxide ammonia sensors. This method can be used for temperature and humidity compensation of gas sensors, which provides a new way to improve the sensing performance of gaseous formaldehyde sensors in a wet environment.

4. Conclusions

This mini review has discussed the recent progress of gaseous formaldehyde sensors based on single polymer, multi-polymers, MIP, and polymer composites with other materials. Compared with the formaldehyde sensors fabricated with small molecules, this type of polymer-based sensor has higher selectivity but slightly lower sensitivity. In order to improve the sensitivity of the polymer-based gaseous formaldehyde sensor, metal oxide, biomass materials, and carbon materials can be introduced into the polymer to form polymer composites.

Although the problem of sensor sensitivity has been solved, most gaseous formaldehyde sensors perform poorly in humid environments. The polymers that can be used as the sensitive layer of the formaldehyde sensor are mostly hydrophilic polymers, which will have a negative impact on the performance of the sensor in a humid environment. At present, researchers are improving the existing sensors with small-sample local Gaussian regression processes and other methods. In addition, some researchers have used hydrophobic polymers for the development of gas sensors. Gaseous formaldehyde sensors can be used to detect formaldehyde concentration in a living environment and have wide application prospects. It is very important to improve the sensing performance of the

gaseous formaldehyde sensor and its practicability in different environments to improve the quality of human life.

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