Journal of Science: Advanced Materials and Devices 11 (2026) 101164

Contents lists available at ScienceDirect

Journal of Science: Advanced Materials and Devices

journal homepage: www.elsevier.com/locate/jsamd

Review Article

A comprehensive survey of NO_2 gas sensors: Functional materials, recent advances, present challenges, and future directions

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ARTICLE INFO

Keywords:
NO_2 gas sensors
Smart sensors
Heterostructures
Nanocomposite hybrids

ABSTRACT

Nitrogen dioxide (NO_2), a major atmospheric pollutant, poses significant risks to human health even at low concentrations and exerts similarly harmful effects on plants and terrestrial ecosystems. Therefore, the real-time, accurate and reliable monitoring of NO_2 concentrations in the atmosphere is very important. NO_2 gas sensors translate the concentration of NO_2 at any given time into analysable signals. So, different materials/technologies have been explored to develop NO_2 sensors. However, there is an increasing global awareness about the possible connections between technological advances, waste generation, and environmental pollution. Moreover, research and technological advancements have recently tilted towards the utilisation of artificial intelligence to develop smart devices. Therefore, this paper reviews the different functional materials in NO_2 sensors, including their advantages and limitations. Likewise, the techniques/technologies for the fabrication of NO_2 gas sensors are discussed. Moreover, recent developments that support sustainability, circularity, or end-of-life composability are presented. Furthermore, the contemporary trend of research towards fast and seamless integration of new generation NO_2 gas sensors with AI and the use of IoT for remote data collection is included in this review. Finally, future perspectives on the manufacture of smart, durable, mechanical, and environmentally robust NO_2 gas sensors are discussed.

1. Introduction

Nitrogen dioxide (NO_2) is one of the primary environmental pollutants that poses potentially significant deleterious effects on human and terrestrial life. Long-term exposure to NO_2 can lead to grave respiratory complications, even at very low concentrations [1,2]. Gas sensors used to monitor and measure NO_2 concentrations may be developed from different materials such as metal oxide semiconductors (MOS), conducting polymers, carbon-based materials, and hybrid materials. MOS-based NO_2 sensors have the advantages of high stability, wide application range, low fabrication costs, and good compatibility with portable devices [3]. However, most MOS-based NO_2 sensors are limited by high operating temperature, poor recovery, and insufficient response [4], thereby necessitating the development of room-temperature NO_2 gas sensors.

Carbon-based materials, on the other hand, are notable for their large surface area, high electrical conductivity, great chemical stability, high carrier mobility, and excellent optical properties [5]. However, it is usually a challenge to coat their surface with functional or organic moieties necessary to improve their molecular sensitivity [6]. Nevertheless, when used as an active sensing material in NO_2 sensors, carbon-based materials and their combination with other materials boast several advantages [7].

Conducting polymers (CPs) are another class of materials that have been widely explored for the development of NO_2 gas sensors. Generally, the use of CPs relies on the π-electron framework in their structure, which makes them electrically conductive. Different approaches have been explored to produce CP-based NO_2 sensors with different performance levels, but the most desirable outcomes are obtained by combining CPs with other sensing materials such as carbon materials or metal oxide semiconductors [8]. The development of hybrid material-based NO_2 sensors often aims at creating heterostructures ($p-n$, $p-p$, or $n-n$) that help to synergise the salient properties of individual materials to fabricate high-performance sensors. The basic differences between p−type and n−type materials are summarised in Table 1.

Generally, there is a growing interest in NO_2 gas sensors, with most of the ongoing research efforts tailored around the selection of new/

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https://doi.org/10.1016/j.jsamd.2026.101164
Received 18 December 2025; Received in revised form 9 April 2026; Accepted 13 April 2026

Table 1
Comparison of p−type and n−type materials as they relate to gas sensing applications.

	p−type functional materials	n−type functional materials
Definition	A p−type material is defined as a semiconductor doped with trivalent impurity atoms, creating holes as the majority charge carriers.	An n−type material is defined as a type of semiconductor that has been doped with pentavalent impurities to increase its conductivity by adding free electrons.
Examples	Nickel oxide, cobalt oxide.	Zinc oxide, tin oxide, titanium oxide, iron (III) oxide.
Role of Impurity Atoms	Trivalent impurities, like boron, have three valence electrons that form bonds with the semiconductor atoms, leaving one incomplete bond or hole.	Doping involves adding impurities like antimony, arsenic, or phosphorus to a pure semiconductor, which increases the number of free electrons.
Hole Movement	Holes move within the crystal when neighbouring electrons fill these vacancies, creating a new hole in the previous electron's place.	
Majority and Minority Carriers	In a p−type semiconductor, holes are the majority carriers, while electrons, generated by thermal excitation, are the minority carriers.	In n−type semiconductors, free electrons are the majority carriers, meaning they primarily carry the electrical charge. Holes, created when covalent bonds break, are the minority carriers in n−type semiconductors.
Electrical Neutrality		n−type semiconductors, despite having many free electrons, remain electrically neutral because the total number of protons equals the total number of electrons.
Sensing mechanism	Decrease in resistance. NO_2 reacts with adsorbed O^- as well as adsorbs directly on the material surface. The resistance of p−type material surface decreases because extracted electrons result in the generation of holes in the valence band.	Increase in resistance. NO_2 reacts with adsorbed O^- as well as adsorbs directly on the material surface. The decrease in the majority carriers results in decreased conductivity.

improved materials and technologies that enhance sensitivity and room temperature selectivity. However, with the increasing global interest in sustainability, there has been a paradigm shift towards the integration of new generation NO_2 sensors with contemporary technological advancements, without sacrificing the integrity of the environment. Therefore, this article reviews the different materials and technologies that have been explored for the development of NO_2 gas sensors. In addition, the recent advances and contemporary technological innovations for the fabrication of new generation NO_2 sensor devices are summarised, particularly those within the past few years. Furthermore, the present challenges and future perspectives are discussed in this review.

2. Development of NO_2 gas sensors

The development of NO_2 gas sensors has evolved over the years, leading to the exploration of different materials to obtain specific outcomes. Generally, the performance of NO_2 gas sensors, particularly in terms of response, recovery, and selectivity, is based on the characteristics of the sensing material. The notable material features that determine the performance of NO_2 gas sensors include surface area, donor density, agglomeration, porosity, acid-base properties, presence of catalysts, sensing temperature, and sensor thickness. Therefore, sensor materials are selected to combine some of these features. The commonly used materials to produce NO_2 gas sensors are described in the following subsections, and the advantages/limitations of each material is summarised in Table 2.

2.1. *Metal oxide-based NO_2 gas sensors*

Metal oxides (MO) such as SnO_2, NiO, In_2O_3, CuO, ZnO, and WO have been extensively used to develop gas sensors, taking advantage of their low cost, facile manufacturing, high sensitivity [17], good thermal stability and excellent electrical properties. These sensors utilise resistance changes in the sensitive material to depict the sensing performance. When atmospheric oxygen is adsorbed on the surface of the MO, it extracts electrons from the conduction band of the MO [18], and becomes adsorbed oxygen in the form of ionic species (O_2^-, O^- and O^{2-}) as described by the following reactions:

$$O_2(\text{gas}) \rightarrow O_2(\text{ads}) \tag{1}$$

$$O_2(\text{ads}) + e^- \rightarrow O_2^-(\text{ads}) \tag{2}$$

$$O_2^-(\text{ads}) + e^- \rightarrow 2O^- \tag{3}$$

$$O^- + e^- \rightarrow O^{2-} \tag{4}$$

The stability of the oxygen ions in reactions (2), (3) and (4) is temperature dependent. For example, O_2^- is stable below 100 °C [18], O^- is stable from 100 to 300 °C, while O^{2-} is stable above 300 °C [19,20]. The sensing mechanism in an MO-based NO_2 gas sensor is determined by the material type. In n−type materials, electron transfer from the conduction band produces an increased resistance [21], whereas the resistance of p−type material decreases as presented in Table 1.

When the surface of MO is exposed to NO_2, the gas reacts with the O^- adsorbed on the MO surface, as well as directly adsorbed on the MO surface according to the following reactions [22]:

$$NO_2(\text{gas}) + e^- \rightarrow + NO^{2-}(\text{ads}) \tag{5}$$

$$NO^{2-}(\text{ads}) + O^-(\text{ads}) + 2e^- \rightarrow + NO(\text{gas}) + 2O^{2-}(\text{ads}) \tag{6}$$

The adsorption of oxygen ions (O^-), is an important step in the sensing performance of MO as this helps the adsorbed oxidizing ions, NO^{2-} (ads) to extract electrons from the surface of the MO.

Table 2
Summary of the advantages and limitations of materials for the development of NO_2 gas sensors.

Materials	Advantages	Limitations	Ref.
1D Carbon-based materials - CNT	Peculiar electrical, optical and mechanical properties	Often require modification for good selectivity	[5,9]
Two-dimensional (2D) materials - graphene	Tuneable electronic properties	Requires modification for good selectivity. Low gas-sensing response	[7]
Metal oxide semiconductors (MOS)	Most used, possesses large surface area for gas adsorption	Requires high operating temperature	[1,4,10]
Conducting polymers	Low operating temperature, highly flexibility, and high surface area	Limited by environmental factors such as humidity	[11,12]
Mesoporous materials	Well-defined pore structure High surface area Interconnected conductive network	Poor selectivity Humidity sensitivity Thermal/structural instability	[13,14]
Hybrid materials	High selectivity, high sensitivity, high recovery, excellent response, and high stability	Limited scope of selectivity	[15,16]

Generally, the use of bare metal oxides such as SnO_2 and NiO as NO_2 gas sensors is often limited by the high operating temperatures [23], poor sensitivity and low selectivity towards NO_2 [24]. Therefore, incorporation of a second phase is generally embraced by researchers. A novel study prepared in-situ grown PPy on mesoporous NiO nanosheets and at the optimal nanocomposite ratio, the sensor displayed a very low detection limit of 49 ppb, and very high selectivity towards NO_2 compared to other gases, as shown in Fig. 1 [24]. Likewise, Sharma et al. synthesised a SnO_2 coated boron nitride nanotubes (BNNTs) NO_2 gas sensor which demonstrated high selectivity and stability towards low concentration (500 ppb) of NO_2 gas, as illustrated in Fig. 2 [23]. However, despite the sensing performance of MO based NO_2 sensors, they are generally disadvantaged by the high operating temperature.

2.2. *Transition metal dichalcogenides*

Transition-metal dichalcogenides (TMDs), such as molybdenum disulfide (MoS_2), tungsten disulfide (WS_2) [25], molybdenum diselenide ($MoSe_2$), and molybdenum ditelluride ($MoTe_2$) [26] have emerged as promising candidates for NO_2 sensing. The interest in this class of materials is based on their salient properties, such as high surface-to-volume ratio, tunable electronic properties, and strong interaction with NO_2 [26,27]. The two-dimensional morphology of these materials helps to facilitate charge-transfer interactions, attributed to their abundant adsorption sites and as such have been used to develop NO_2 sensors for room temperature applications [28]. To overcome the stacking tendencies of TMDs, most of the studies on TMD-based NO_2 sensors focus on device optimization through the generation of heterostructures, morphology engineering, defect modifications and so on [29,30].

Among the available TMDs, MoS_2 has been extensively explored for the development of NO_2 sensors [31]. However, WS_2-based sensors have equally demonstrated intriguing low-temperature NO_2 sensing performance [32]. Aside from these, other TMDs have been explored for NO_2 detection, including $MoSe_2$, $MoTe_2$, and VS_2 [29]. While selectivity remains a major challenge for NO_2 gas sensors, the recent crop of studies has demonstrated that TMDs can be engineered towards high NO_2 selectivity [33,34].

2.3. *Third generation NO_2 sensors*

Third-generation NO_2 sensors represent a class of advanced electrochemical sensors whose sensing performance is based on direct electron transfer (DET) between NO_2 and the sensing electrode [35]. Advances in NO_2 gas sensors birthed the development of third-generation sensors that combine durability, selectivity, and sensitivity towards NO_2 under extreme conditions. In this regard, Gallium Nitride (GaN) and Silicon Carbide (SiC) have been explored for their peculiar properties, such as wide band gaps (3.4 eV for GaN and ≈3.26 eV for SiC) [36,37], high thermal stability, and chemical inertness. These properties allow them to be useable in environments where conventional sensors would ordinarily fail [38].

GaN-based NO_2 sensors are built, taking advantage of the high electron mobility and strong surface polarisation effects, which enhance the adsorption of oxidizing gases like NO_2 [39]. Among the recent developments, Han et al. developed a Pt-loaded nanoporous GaN sensor, which showed remarkable NO_2 sensing performance at room temperature, including low concentration sensitivity (100 ppb), rapid response (≈22 s) and quick recovery times (≈170 s) [37]. To address the high detection lower limit (LOD) issues of GaN, Han et al. designed a GaN quantum dots (QDs) film to enhance the gas interaction capability [40]. On the other hand, power consumption, which is a significant issue in conventional sensors, was addressed by Rambeloson et al. through photoactivation of GaN nanowire to produce NO_2 sensors that may be integrated with Internet of Things (IoT) platforms [41].

Silicon carbide (SiC) is also notable for its high thermal and chemical stability [42] and has been used to develop sensors for harsh environments such as combustion systems and aerospace applications. The adsorption of NO_2 on the surface of SiC was theoretically investigated by Yadav et al. [43]. The strong adsorption confirmed high sensitivity, with the material showing stable signal transduction even under fluctuating environmental conditions such as varying humidity and pressure [43]. Although SiC sensors are limited by high operating temperature (>400 °C), catalytic functionalization could be used to lower its operating temperature without negatively affecting its performance [38].

2.4. *Conducting polymers-NO_2 gas sensors*

Conducting polymers (CPs) have high electron affinity and the π-electron framework makes them electrically conductive. The commonly used conducting polymers, such as polyaniline (PANI), polypyrrole (PPy), poly (3,4-ethylenedioxythiophene) (PEDOT), and poly (phenylene vinylene) (PPV) are highly sensitive, and they have shown excellent performance as gas sensors [44]. They also have the added advantage of being cost-effective, with high functionalities, a large surface area for gas adsorption, good recovery, quick response, and facile fabrication methods. The chemical structure of CPs contains a sp^2 configuration, which favours delocalised charge carrier transport. As gas sensors, CPs combine the electronic, magnetic, and optical properties of metal and semiconductors with the processability and mechanical robustness of polymers. Owing to the conjugated backbones in CPs, oxidizing (electron acceptor) gases such as NO_2 can abstract electrons from it as follows:

$$NO_{2(gas)} + e^{-} \rightarrow NO_{2(ads)}^{-} \tag{7}$$

As a result of the above reaction, the concentration of electrons on the CP surface reduces, resulting in increased resistance. This is followed by a subsequent reaction between $NO_{2(ads)}^{-}$ and $O_{2(ads)}^{-}$ as follows:

$$NO_{2(ads)}^{-} + O_{(ads)}^{-} \rightarrow 2O_{(ads)}^{2-} + NO_2 \tag{8}$$

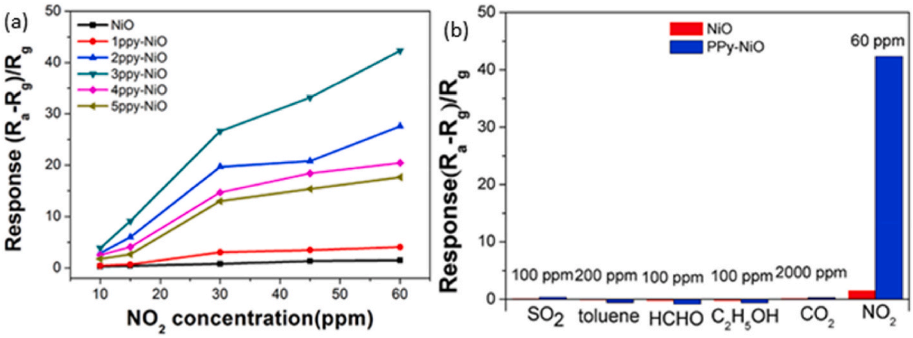

Fig. 1. (a) Response of PPy-NiO composites to different NO_2 concentrations at varying concentrations of PPy, and (b) selectivity of the optimum PPy-NiO composite concentration to NO_2, compared to other interfering gases [24].

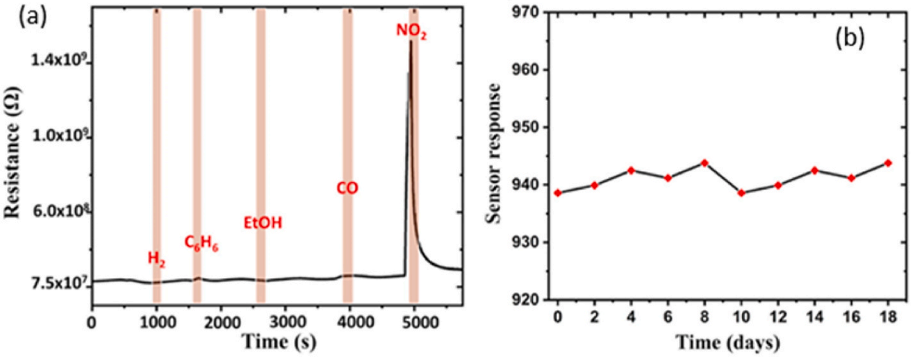

Fig. 2. (a) selectivity of BNNTs-SnO_2 sensor towards NO_2 in the presence of interfering gases, and (b) stability of the sensor at low concentration (500 ppb) of NO_2 [23].

The reaction continues as NO_2 continues to abstract electrons from the polymer surface, acting as a dopant in the process, increasing the surface area of the polymer and, invariably, its response towards NO_2 [45].

Generally, the combination of CPs with other active materials offers additional advantages to the sensing performance as demonstrated by Karmakar et al., who showed that the addition of silver nanoparticles in PPy and subsequent doping with *p*-toluenesulfonic acid (pTSA) produced a remarkable improvement in its NO_2 sensing performance (Fig. 3) [46]. Likewise, Pasupuleti et al. produced a GO-PEDOT:PSS nanocomposite and compared it to PEDOT:PSS or GO alone, the GO-PEDOT:PSS sensor exhibits highly improved NO_2 sensing performance [47].

2.5. *Carbon-materials-based NO_2 gas sensors*

The carbon-based materials used for the fabrication of gas sensors are graphene, carbon nanotubes (CNTs), graphene oxide, reduced graphene oxide and their derivatives [48–50]. The desirable features of CNTs include high electrical conductivity, large aspect ratio, excellent chemical and optical properties, and high specific surface area for gas adsorption [49]. To enhance the performance of CNT-based gas sensors, CNTs are often functionalised either through covalent approaches or through non-covalent functionalization [48,51]. These create defects in CNTs by removing the carbon atoms on the sidewalls and ends of CNTs, thereby facilitating preferential gas adsorption [52,53].

Graphene-based gas sensors rely on the excellent gas adsorption capacity of graphene, in addition to its desirable mechanical robustness, large surface area and high electron mobility [54,55]. There is only a possibility of van der Waals interaction between raw graphene and gas molecules, which often results in gas sensors with very low gas-sensing responses [56]. Therefore, researchers mostly adopt different approaches to modify the raw graphene for the development of gas sensors. Shaik et al. developed a chemiresistive NO_2 gas sensor using nitrogen-doped graphene nanosheets (NGS) [57], which showed excellent sensitivity and selectivity towards NO_2 and against common interfering gases such as NH_3, H_2S, CO, SO_2, and VOCs, as shown in Fig. 4. Nitrogen doping helped to improve the gas sensitivity through enhanced gas adsorption sites [57]. Generally, graphene-based sensors have been developed by several researchers, with different levels of performance. However, the main challenge is the difficulties in coating the active surface of graphene with functional organic moieties [6].

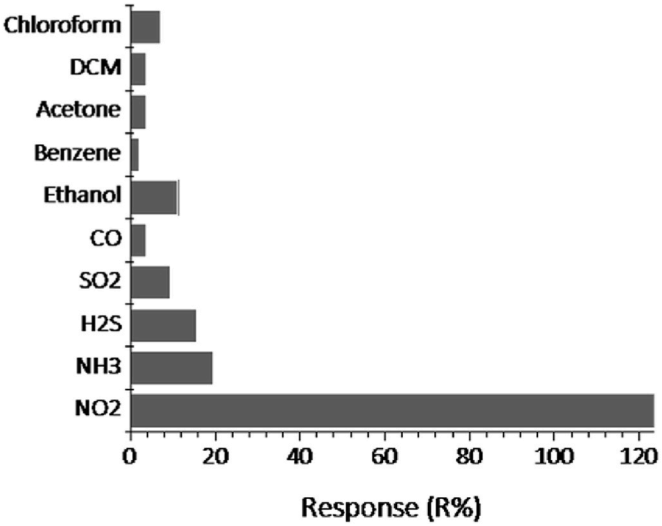

Fig. 4. Selectivity of graphene-based NO_2 gas sensors towards NO_2 compared to common interfering gases and VOCs [57].

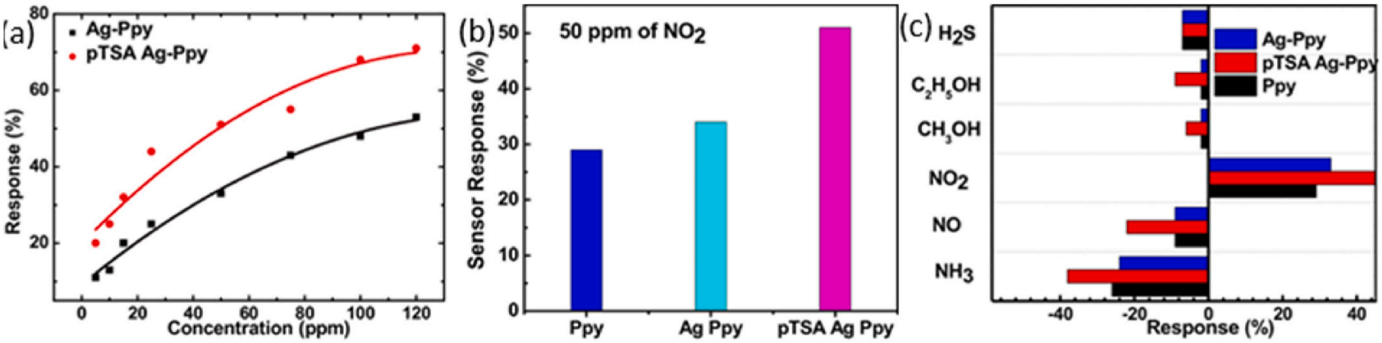

Fig. 3. Effect of *p*-toluenesulfonic acid (pTSA) doping on the sensor response of Ag-PPy nanocomposite film at different NO_2 concentrations (a and b), and selectivity of the films towards NO_2 (c) [46].

2.6. Organic/inorganic nanohybrid-based NO_2 sensors

Hybrid materials offer a synergised approach to material improvement by combining the salient properties of different materials [58]. When used as gas sensors, hybrids can detect gases at very low concentrations [50], as shown by Bai et al., who developed a sensor using rGO nanosheets modified with CuO nanoflakes. The sensing performance was remarkably improved by the synergistic effects of the CuO (flake-like nanostructure) and rGO (extensive conductive channels), which facilitated faster redox reactions and created sufficient active sites for gas adsorption as shown in Fig. 5 [59].

Similarly, Ma et al. used a combined room-temperature oxygen plasma treatment and hydrothermal reactions to produce various Co_3O_4-RGO nanohybrids for NO_2 detection [60]. The generated oxygen vacancies (OVs) helped to modify the nanocrystal structure by increasing the active sites for gas adsorption (Fig. 6a) [60]. Furthermore, the presence of p−p homotypic heterojunctions between Co_3O_4 and RGO was found to support NO_2 adsorption through the creation of p−p heterointerface, which produces a space charge region for electron transfer as illustrated in Fig. 6b and c [60].

Generally, the formation of heterojunctions can help to improve gas sensitivity through increased carrier mobility or increased active sites [21]. In addition, it can help to facilitate faster adsorption/desorption (response/recovery).

3. Technologies

Different technologies have been adopted for the development of NO_2 gas sensors as discussed in the following subsections. For example, Shaik et el., constructed a gas sensing assembly to assess the sensing performance of graphene-based NO_2 gas sensors prepared through a drop-drying method [57]. Likewise, Zhang et al. developed a SiNWs/WO_3 composite nanowires sensor for room temperature detection of NO_2 (Fig. 7) [61]. The sensing performance of the sensor was notably enhanced by the composite structure, compared with the individual components due to the sensor fabrication method which helped to create an increased number of active sites on the sensor for gas adsorption [61].

3.1. Interdigitated electrodes (IDEs)

Interdigitated electrodes (IDEs) act as planar transducers that convert adsorbate-induced changes in a sensing film into measurable electrical signals by concentrating fringing electric fields within micron scale gaps between interleaved "fingers." The use of IDEs for NO_2 sensing is generally associated with its suitable features that include high sensitivity, simple fabrication, and good compatibility with conventional sensing materials [62].

Compared to conventional metal oxide sensors which usually require 200–500 °C to activate surface reactions, IDEs can perform effectively at room temperature or with minimal heating [63]. Patil et al. printed Ag doped ZnO on Ag IDEs, with the sensor demonstrating room temperature NO_2 sensing and a strong response attributed to optimized charge transfer [62]. Different other materials and fabrication techniques have been explored with IDEs NO_2 sensing, particularly at room temperature. For example, Farae et al. applied a solution of CdS/PEDOT:PSS onto IDEs through a drop-casting method [12], while Umar et al. constructed a PANI/Ag_2O/GO composite sensor on IDEs made up of platinum microelectrodes on alumina chips (Fig. 8), through a drop and dry method [64].

The recent trend of research is to miniaturise the IDE-based NO_2 sensor, make it cost-effective and produce it on flexible substrates that support wearable applications. This is partly because the flexibility and bendability of IDE devices makes it compatible with fabric and polymer substrates [63]. Zhang et al. created an IDE (Cu/Au) structure on a polyimide (PI) substrate, spraying the sensing material on the IDE as shown in Fig. 9, thereby making it suitable for wearable applications [63].

The selectivity and response of IDE based NO_2 sensors may be improved through the incorporation of functional groups into the sensing material as demonstrated by Lee et al., through a miniaturised NO_2 sensor made from polymer intercalated graphene/reduced graphene oxide [65]. Overall, there has been a lot of work on the fabrication of IDE-based NO_2 sensors. However, the present crop of research largely focuses on improving signal processing and enhancing device integration [66].

3.2. Surface acoustic wave systems

Surface acoustic waves (SAW) have been explored for the rapid and selective detection of NO_2 gas under different conditions, using various materials because SAW devices are inexpensive, highly reliable, and accurate [6]. Studies on SAWs for NO_2 sensing generally exploit the energy concentration in SAW devices on or near the surface of the developed sensing device. The resonant energy from the sensor is then wirelessly transmitted to recording devices. Various sensing materials may be used to develop SAW devices for NO_2 detection. However, the combination of different materials generally offers better performance than their individual components as demonstrated by a high-performance NO_2 gas sensor based on GO-PEDOT:PSS nanocomposite, which was cast on lanthanum gallium silicate (langasite, LGS) [47]. Similarly, a room temperature SAW-based NO_2 sensor was developed by Xiong et al., using a 3D porous architecture of silver-decorated reduced graphene oxide-polypyrrole hybrid aerogels (rGO-PPy/Ag) (Fig. 10). The sensor showed high sensitivity (127.68 Hz/ppm), high selectivity towards NO_2, and a fast response/recovery time (36.7 s/58.5 s) [67].

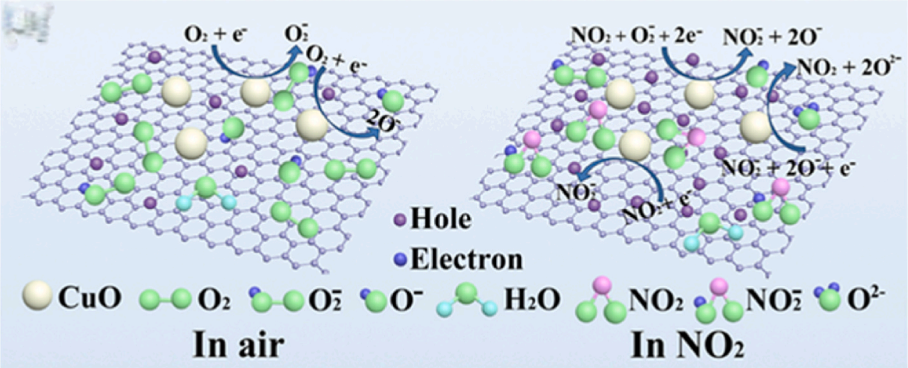

Fig. 5. Schematic illustration of the gas-sensing mechanism of NO_2 gas sensor based on rGO nanosheet modified with CuO nanoflakes [59].

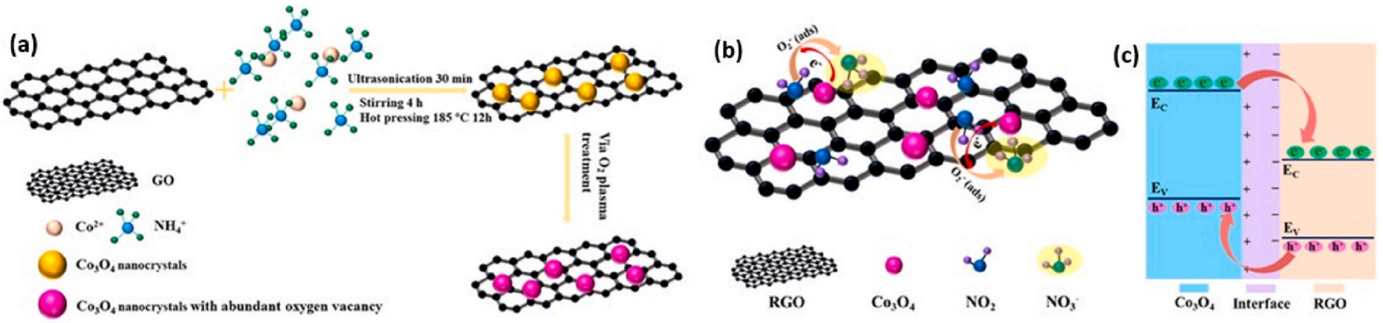

Fig. 6. (a) Synthesis process for Co_3O_4 -RGO-OVs, (b) surface reactions of Co_3O_4 -RGO in the presence of NO_2 gas, and (c) schematic illustration of the electron transfer process [60].

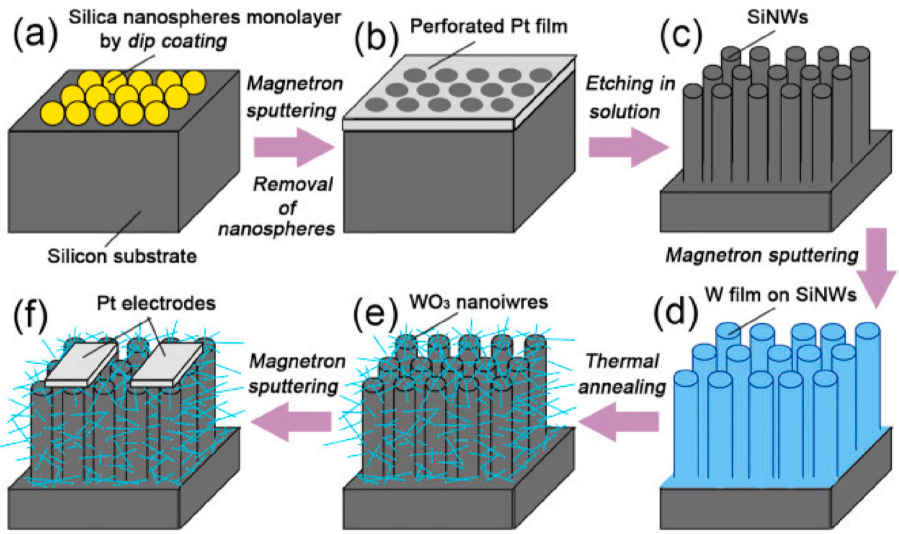

Fig. 7. Fabrication process of $SiNWs/WO_3$ nanowire composite sensors [61].

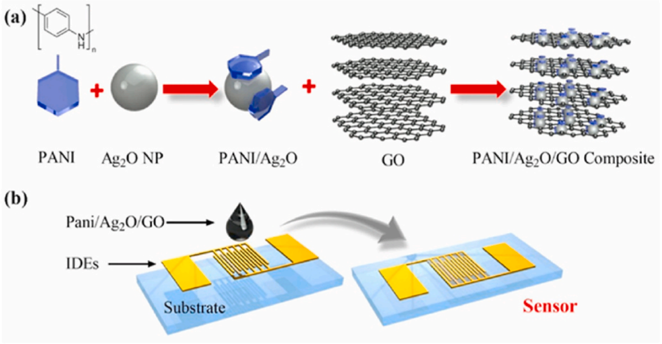

Fig. 8. (a) Synthesis of $PANI/Ag_2O/GO$ composite, and (b) fabrication of $PANI/Ag_2O/GO$ NO_2 sensors [64].

3.3. Flexible and wearable sensors

Emerging flexible sensors have recently gained increased attention among researchers for the detection of NO_2 gas. The conventional wearable NO_2 sensors are based on composites of MOS and other conducting materials that offer high conductivity and sensitivity [63].

The recent trend of research focuses on the fabrication of NO_2 sensors whose performance extends beyond mere sensitivity, towards durability, comfort, and portability. Chemical, electrochemical, and ex-situ approaches such as drop casting, extrusion printing, screen printing, inkjet printing, spin coating, and dip coating may be used to develop wearable NO_2 sensors. However, the electrochemical methods are disadvantaged by the need for electrically conductive substrates, while the chemical methods are generally limited by the inhomogeneity of the components. In contrast, ex-situ approaches are simple, support homogeneity, and can be applied on a variety of substrates, including insulating substrates [11]. Notable among the recently fabricated wearable NO_2 sensors is a watch-type flexible and wearable NO_2 sensor developed by Chen et al. (Fig. 11) [68], and the wearable flexible room temperature NO_2 gas sensor fabricated by Khan et al. [69].

Generally, the creation of porous and breathable structures through technique modification may be used to develop flexible sensors with improved gas diffusion/adsorption, improved comfort in wearable applications, and enhanced mechanical robustness. For example, a highly sensitive, flexible, and breathable NO_2 sensor was fabricated by Yang et al. (Fig. 12) [70]. Asides these, other inorganic and hybrid materials like flexible ceramic sponges and halide perovskites are currently being explored for better durability, sensitivity, flexibility, and room temperature gas sensing performance [71].

3.4. Eco-friendly disposable sensors

The development of eco-friendly and disposable NO_2 gas sensors holds great potential as sustainable alternatives to their non-environmentally friendly counterparts. These sensors are often fabricated on paper or cellulose-based nano fibre substrates, relying heavily on the environmental friendliness and biodegradability of this class of

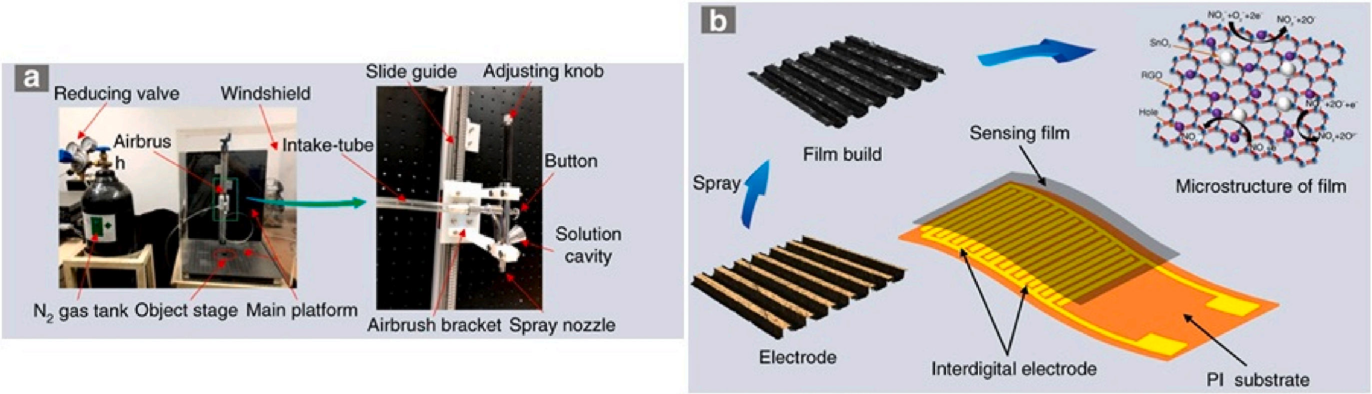

Fig. 9. (a) spraying set up, and (b) rGO/SnO_2-based NO_2 sensor constructed on (Cu/Au) IDEs placed on polyimide substrate [63].

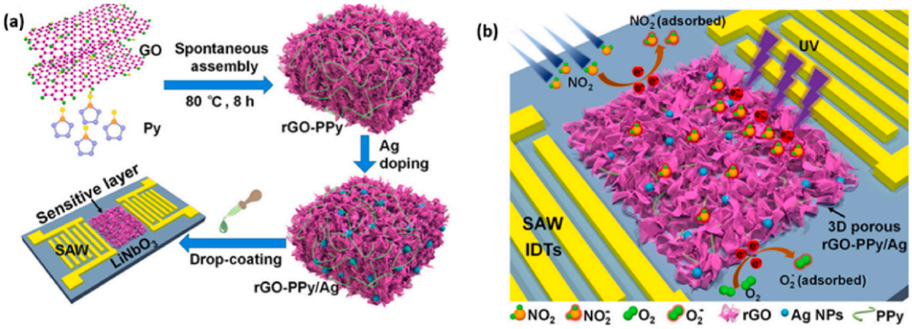

Fig. 10. (a) Synthesis of 3D porous architecture rGO-PPy/Ag composite, and (b) Illustration of the gas sensing mechanism in the presence of NO_2 gas [67].

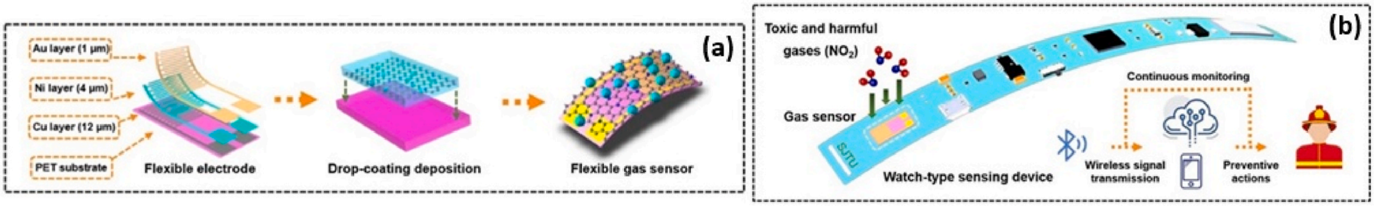

Fig. 11. (a) Illustration of the flexible electrode structure, and (b) schematic illustration of the flexible wearable sensor device, its wireless transmission, and concept of cloud system [68].

materials [72].

A paper-based disposable NO_2 detection device was constructed by Koga et al., which combined zinc oxide (ZnO), low-cost graphite electrodes, and wood-cellulose-based biodegradable nanofibre paper substrate (Fig. 13) [73]. In another study, Beniwal et al. produced a PEDOT: PSS based flexible and disposable NO_2 gas sensor [74], depositing the sensing material on graphene-carbon ink interdigitated electrodes (IDEs), which were screen-printed on a paper substrate. Similarly, Matatagui et al. developed a low-cost, low-environmental impact disposable NO_2 gas sensor with high sensitivity and room temperature selectivity towards NO_2, using a biodegradable paper substrate [75]. Aside from cellulose paper substrate based disposable NO_2 sensors, other supports have also been explored, such as algae sheets and flexible organic substrates [76,77].

3.5. Chipless RFID NO_2 sensors

The development of chipless radio frequency identification (RFID) NO_2 sensors offers low-cost, wireless, and battery-free detection of NO_2. These sensors rely heavily on the RFID principles and the sensing properties of conventional conductive materials to facilitate real-time remote detection of NO_2 gas. In addition, they incorporate IoT, for environmental monitoring [78]. The mechanism of RFID NO_2 sensors is based on the change in electrical properties of conducting materials when exposed to NO_2. As the electrical properties change, the sensor's response to radio frequency is altered thereby supporting the detection of low NO_2 concentration [78,79]. The RFID NO_2 sensor fabricated by Mullomi et al. showed a fast response to NO_2 gas, but the sensor recovery is low, thereby making it more suitable for non-continuous threshold monitoring. However, the recovery can be improved through different approaches, such as UV exposure [79].

Generally, the wireless nature and non-battery requirement of RFID NO_2 sensors place them in a good position for cost-effectiveness and high suitability for remote sensing of NO_2 [78,79]. While it is more common to find RFID sensors for other gases such as CO_2 and NH_3, there is an emerging interest in RFID-based NO_2 sensors [80].

3.6. Smart sensors – AI and IoT

Conventional NO_2 sensors are sometimes disadvantaged by factors such as poor selectivity, high power consumption, and inability to make independent decisions. As such, smart technologies that incorporate

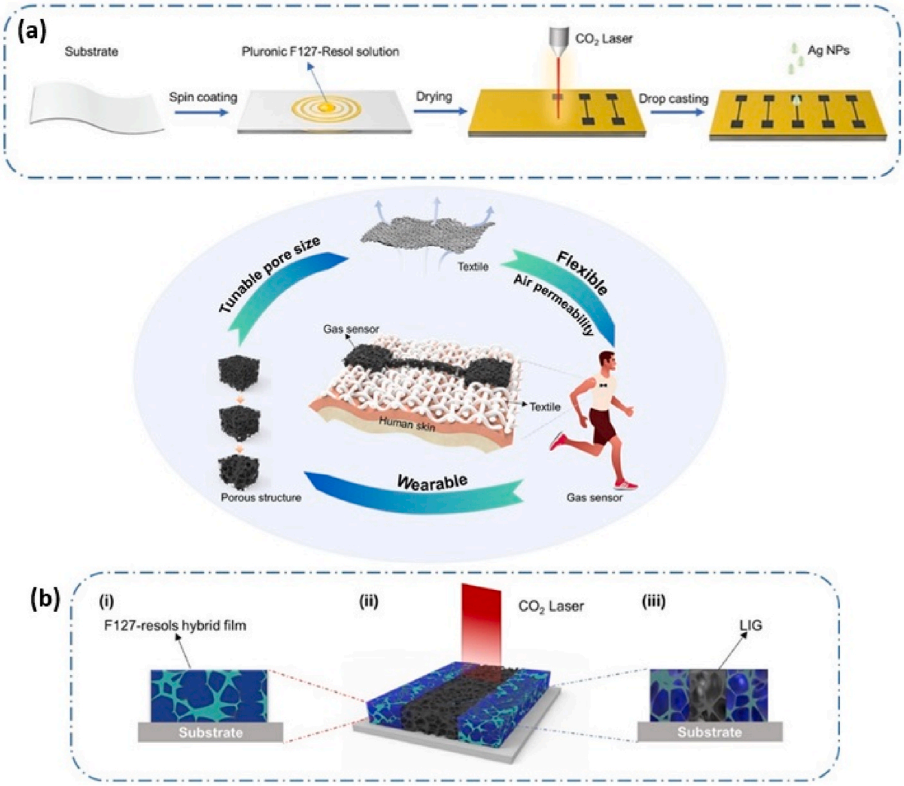

Fig. 12. (a) Fabrication process of Ag/LIG-based breathable NO_2 gas sensor, utilising (b) transformation of F127/resolves into porous LIG with transient heating by a CO_2 laser [70].

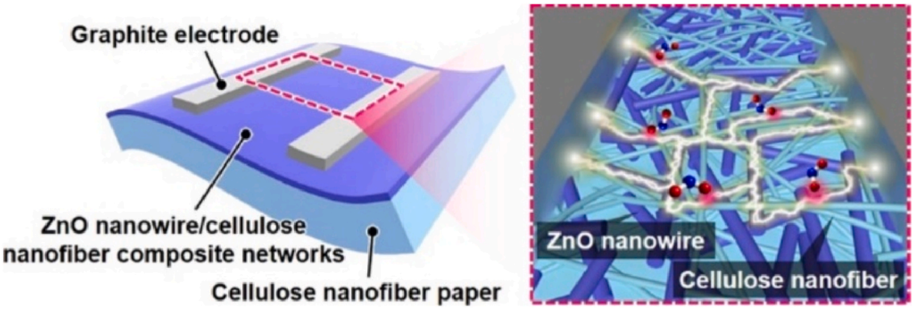

Fig. 13. Disposable molecular NO_2 sensor device made from a cellulose nanofibre paper substrate, ZnO nanowire, and a graphite electrode [73].

artificial intelligence (AI) and IoT into sensor fabrication have recently gained increased attention as demonstrated by Khan et al. [81]. IoT enables fast, real-time, and reliable remote environmental monitoring and data collection from sensors [80]. One of the emerging areas of this technology is the construction of self-powered devices. However, self-powered gas sensors are sometimes limited by high detection limits and more recent studies have focused on overcoming this challenge [82]. Nevertheless, the main benefit of this class of sensors is their ability to ensure real-time monitoring, facilitate fast data analysis and sometimes predict future air quality [83].

Presently, AI is influencing different aspects of material and technological developments. In particular, the selection of functional materials for varying applications may be achieved with significant contributions from AI models. For example, diffusion-based generative models like Microsoft's MatterGen can identify stable inorganic crystals across the periodic table [84]. This could enhance targeted searches for desired properties such as defect formation energies and adsorption energetics, which are important descriptors for gas sensing.

With respect to the development of NO_2 gas sensors, machine learning, which is a branch of AI, is helping to expedite the discovery of new materials and technologies for the fabrication of contemporary NO_2 gas sensors, as well as next-generation sensors. This is largely dependent on the ability of machine learning models to swiftly analyse large databases and come up with sensing and electrode materials with high potential for fabricating high-performance NO_2 sensors [85,86]. As such, machine learning techniques may be used to analyse sensor signals and mitigate the cross-sensitivity, otherwise known as low-selectivity issues of conventional NO_2 gas sensors. For example, Koziel et al. introduced an advanced technique to calibrate low-cost NO_2 sensors by combining machine learning with artificial neural networks (ANNs) [87] while Pietrenko-Dabrowska et al. used machine learning methods to develop a low-cost autonomous NO_2 sensor with high sensing accuracy [88]. An AI integration workflow proposed by Chowdhury et al. is presented in Fig. 14a [89].

Aside from the real-time monitoring of NO_2, machine learning techniques can also be used to predict future concentrations of NO_2 in the atmosphere. For example, a weather-independent NO_2 sensor was fabricated by Lee et al. through the integration of a convolutional neural

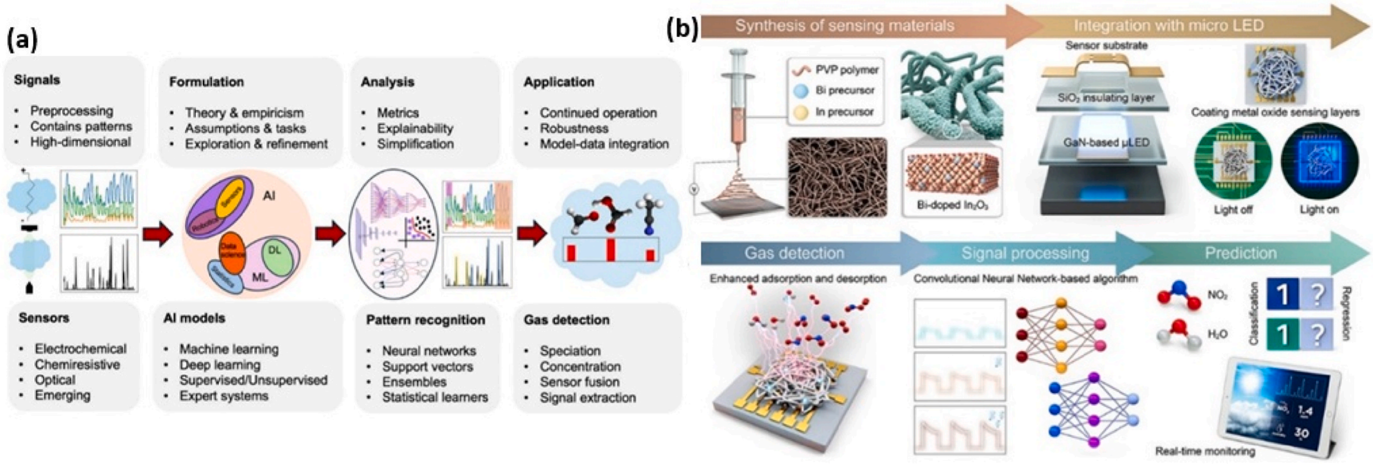

Fig. 14. (a) An illustrative workflow for the integration of AI in gas sensing [89] and (b) illustration of AI-supported NO_2 sensing showing an overview of processes from material synthesis to gas prediction [90].

network-based deep learning algorithm, which demonstrated 99% prediction ability (Fig. 14b) [90].

4. Recent advances in the development of NO_2 gas sensors

The recent advances in the fabrication of NO_2 gas sensors have largely concerted efforts towards the development of room temperature NO_2 gas sensors and those that can detect trace amounts of NO_2. In addition, there is an increasing amount of research aimed at improving the stability, sensitivity, and selectivity of NO_2 gas sensors. Furthermore, there is growing interest in NO_2 sensing materials not only for environmental monitoring but also for health evaluation [91]. Therefore, different approaches are presently being explored, including the use of advanced nanomaterials, the development of hybrid composites, and the engineering of innovative sensor structures for the real-time, reliable, practical, and efficient detection of NO_2.

4.1. *Tailored performance*

The new generation NO_2 sensors are built to combine sensitivity, selectivity, durability, and response/recovery in single sensors. This is mostly achieved by synergising the desirable gas-sensing properties of different materials to generate hybrids and composites for improved performance. For example, there are reports of significant improvements in the sensing performance of materials such as hybrid carbon/metal oxide, carbon/conductive polymer, MXene/metal oxide, and metal oxide/conductive polymer, compared to the single materials [14, 92].

In another vein, the surface of sensing materials is currently being tailored either through defect engineering, incorporation of dopants, and development of heterostructures to facilitate gas adsorption and enhanced NO_2 sensing performance [13,17,92]. One of such approach was explored by Jiangbin et al. who strategically integrated atomic defects of palladium nanoparticles on tungsten oxide (WO_3) through atomic layer deposition (ALD), thereby enhancing the oxygen vacancy concentration, while also optimising the electronic structure of the material [93]. Generally, oxygen vacancy helps to improve the sensing performance of MOS-based sensors and has remained a significant approach being explored in the development of contemporary NO_2 gas sensors [94].

The use of dopants has also produced excellent results for NO_2 sensing through increased NO_2 adsorption and improved sensor response/recovery. For example, the antimony (Sb) doped SnO_2 sensor produced by Xiao et al. showed satisfactory sensing performance [95]. Likewise, Zhang et al. demonstrated the potential of ammonia pyrolysis-treatment of $Ti_3C_2T_x$ Mxene to produce nitrogen-doped $Ti_3C_2T_x$, which invariably enhanced the selectivity and sensitivity of the sensor [96].

4.2. *Materials engineering*

Material-wise, advanced nanomaterials containing carbon nanomaterials such as carbon nanotubes, graphene, and laser-induced graphene (LIG) are currently being used, taking advantage of their impeccable electrical properties. The combination of these materials with other materials (conducting polymers and MOS) for the development of advanced composites produce sensors with improved selectivity, increased sensitivity, and better sensing performance [97]. Recently, Li et al., produced a high performance NO_2 sensor from functionalised graphene nanofibers, using a conducting polymer (PEDOT:PSS) and silver nanoparticles (AgNPs), through supramolecular assembly and oriented lyophilisation (Fig. 15) [91]. Similarly, Soydan et al. fabricated a novel room temperature flexible NO_2 sensor through a low-cost and facile laser scribing of highly porous laser-induced graphene (LIG)/SnO_2 heterostructures [97].

As part of the recent advances in the development of NO_2 sensors, metal oxides and hybrids are also currently being engineered to exhibit specific morphologies, comprise dopants and generate composites with improved stability and sensitivity [17]. Guo et al. synthesised an optoelectronic NO_2 sensor based on ZnS/SnS_2 heterostructures [98], with its performance attributed to the engineered material surface properties and accelerated electron transfer at the heterojunction interface. Asides these, two-dimensional materials such as MXenes and molybdenum disulfide (MoS_2) have also recently emerged as potential candidates for the development of room temperature NO_2 sensors [13,14].

4.3. *Heterostructure design*

Heterostructure engineering helps to improve the sensing performance of NO_2 sensors, but it sometimes comes at a cost that sacrifices the sensitivity and room temperature operation of the sensor [99]. Therefore, the recent trend in the fabrication of NO_2 sensors is the engineering of 2D or mixed-dimensional heterojunctions that amplify charge transfer and adsorption while maintaining room-temperature operation [100]. Yi et al. synthesised a 2D/2D Bi_2Se_3/$SnSe_2$ stack which achieved 25 ppb LOD with 15 s response (10 ppm NO_2) at room temperature [101]. This performance was attributed to built-in band bending (Fig. 16) and abundant edge/surface states that accelerate electron withdrawal by the oxidizing NO_2. Similarly, Dhariwal et al. fabricated a sensor based on rGO/BiOCl heterostructure which showed

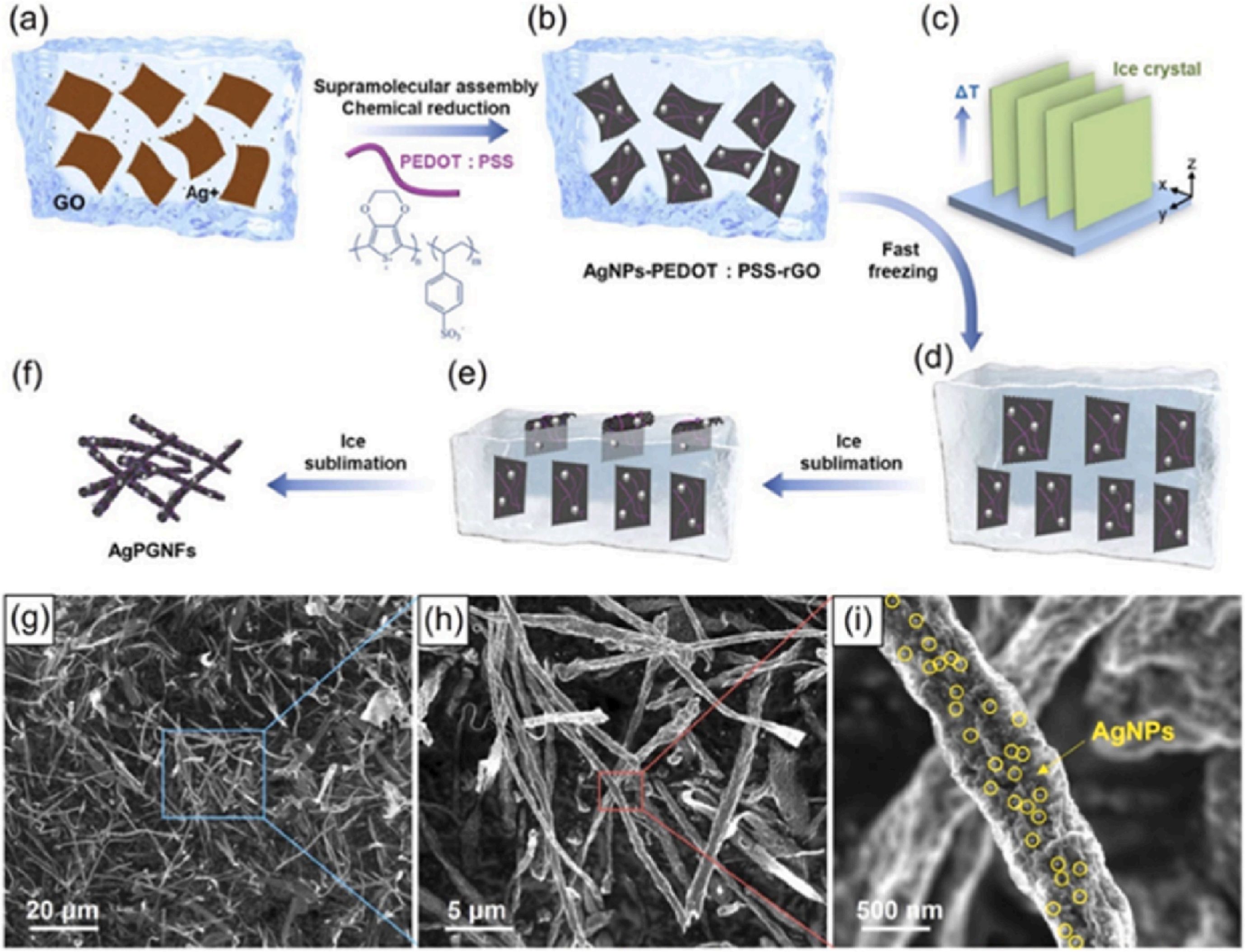

Fig. 15. Schematic illustration of the fabrication of AgPGNFs by supramolecular assembly combined oriented lyophilisation (a-f), and (g) Low-, (h) medium-, (i) high-magnification SEM images of the AgPGNFs [91].

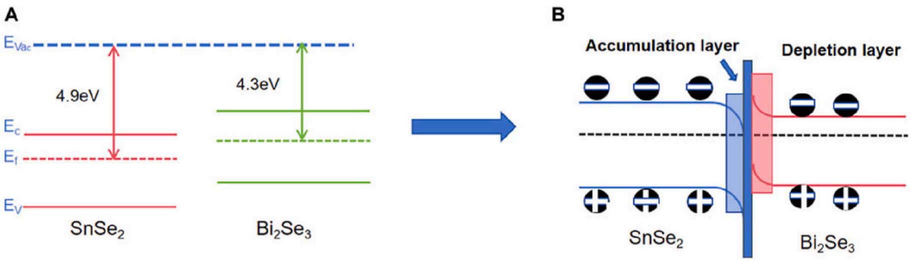

Fig. 16. Description of the energy band of $SnSe_2/Bi_2Se_3$ heterostructure (a) before equilibrium and (b) after equilibrium [101].

LOD of about 0.06 ppb, and a quick response/recovery of ~9 s/21 s (100 ppb) at 25 °C [102]. In another study, an edge enriched CeO_2/MoS_2 heterostructure demonstrated a >10 × response enhancement at room temperature to 5 ppm NO_2 [103].

Heterostructures based on MXene hybrids have also emerged recently, such as the iodine-functionalised $Ti_3C_2T_X$ films produced by Hilal et al. [104], and MXene–Cu_2O composites produced by Ren et al. [105].

4.4. *Oxygen vacancy engineering*

Defect engineering through the generation of oxygen vacancies (V_o) has recently re-emerged as a reliable approach to room temperature NO_2 sensing. Banking on the knowledge that the oxygen vacancy in metal oxides serves as active sites for NO_2 adsorption, and contributes to gas-solid interfacial charge transfer, Zhang et al. demonstrated the room temperature sensing capabilities of V_o-rich 2D SnO nanosheets with a record response ~136 (to 800 ppb) and LOD ~10 ppb at room temperature [106]. V_o-tailored $WO_{3\text{-}x}$ nanostructures fabricated by Ganesan et al. also demonstrated room temperature NO_2 responsiveness and unusual temperature-dependent semiconductor/insulator transitions [107].

Most times, heterostructures merge V_o with band alignment. For example, the MXene–Cu_2O composite fabricated by Ren et al. explicitly couples interfacial tunneling with V_o-enabled chemisorption, yielding ppb-level detection at room temperature [105].

4.5. Performance metrics and mechanistic insights

The recent advances in NO_2 sensing technologies have produced remarkable improvements in sensitivity, selectivity, energy efficiency, and room temperature operability. These innovations primarily stem from approaches based on materials engineering, heterostructure design [60], and defect manipulation through the generation of oxygen vacancies [65,105].

A compilation of some remarkable breakthroughs in room temperature NO_2 sensing is presented in Table 3. It is evident in Table 3 that most of the innovations point toward a mechanistic consensus of either defect engineering, heterostructure designs, or surface modification. These approaches help to significantly enrich active sites, accelerate charge transfer, and enable room temperature NO_2 detection thereby marking a substantial step toward scalable, high performance air quality monitoring technologies.

5. Current challenges and future perspectives in the development of NO_2 gas sensors

The advancement in the development of NO_2 gas sensors is rapidly evolving. As detailed in this review, different materials have been used to produce NO_2 gas sensors, including metal oxides, conducting polymers, carbon-based materials, and advanced nanomaterial composites. The present crop of literature demonstrates different approaches to improve the room temperature application, improve selectivity and enhance the stability of NO_2 sensors. Nevertheless, there are still ongoing challenges of room temperature selectivity towards NO_2, response/recovery, and good sensitivity to very low NO_2 concentrations. In addition, little success has been achieved for NO_2 gas sensors compatible with changing environmental conditions, such as temperature and humidity.

Most of the ongoing research efforts are concerted towards the exploration of new materials and technologies that enhance sensitivity, room temperature selectivity towards NO_2, sustainability, and integration with contemporary technological advancements. Therefore, it is plausible to state that the future trend of research on NO_2 gas sensors will involve the selection and utilisation of new unexplored or partially explored materials for the development of high-performance NO_2 gas sensors. Approaches such as doping and defect engineering, which are currently being explored, will be helpful for the manipulation of material properties towards desirable performance. The generation of heterostructures will also play an active role in the development and performance boost of next-generation NO_2 sensors.

In another vein, there is a growing global environmental awareness about the possible contributions of technological advances to waste generation. The development of NO_2 gas sensors with high mechanical and environmental robustness can help to mitigate this challenge. Therefore, the selection of materials that support sustainability, circularity, or end-of-life composability can be used as a strategy to overcome waste generation.

Furthermore, with the recent global attention towards artificial intelligence, future perspectives on the development of NO_2 gas sensors will be reliant on the use of technologies and materials that support fast and seamless integration with AI and the use of IoT for remote data collection. This would require the fabrication of more self-powering, low-power, portable, flexible, and wearable sensors. While advanced machine learning algorithms can help in the selection of new materials or material combinations for trials, technologies such as non-dispersive infrared (NDIR) can help to ensure data reliability and precision.

Table 3
Summary of materials, and performance metrics of selected room temperature (≈20–25 °C) NO_2 sensors.

Material/Composite	Response (conc., value, time)	Recovery time	LOD	Ref.
rGO/BiOCl heterostructure	100 ppb → Rg/Ra≈3.78; ~9 s	~21 s	≈0.06 ppb	[102]
Bilayer graphene or S-doped graphene on $LiTaO_3$	Sensitivity 0.29°/ppm (RT)	NA	0.068 ppm (Bl-Gr); 0.140 ppm (S-Gr)	[108]
Laser-induced graphene (LIG) and LIG/SnO_2	50 ppm–10 ppm range; trends reported	NA	Up to 2.5 ppm	[97]
3D MoS_2 network microstructure	12.3 ppm → 15% (75 × vs 2D)	204 s	0.297 ppm	[31]
ZnO–MoS_2–TiO_2 ternary	0.1 ppm → 6%; 4 ppm → 58%; ~30.6 s	37.1 min	NA	[33]
I-terminated $Ti_3C_2T_x$ MXene	50 ppb → 0.2%; 200 ppm → 23%; 90 s	100 s	NA	[104]
$Ti_3C_2T_x$-derived K-titanate nanoribbons	50 ppm → 649% (RT); 1 s	2 s	5 ppb	[109]
Blue μLED-activated SnO_2 NPs (Au/Pd/Pt-decorated)	47 s	49 s	200 ppb	[110]
Graphene oxide from upcycled PET	1026 ppb → ~11% response	Fully reversible at RT	~1.43 ppb	[111]
Pt-loaded nanoporous GaN	100 ppm → response in 22 s; range 200 ppm–100 ppb	170 s	100 ppb	[37]
ZnO/In_2O_2 heterojunction	30 ppm → response 5.25; 267 s	7 s	NA	[112]
In_2O_3–NbS_2 heterojunction	500 ppb →7520%; 8 s	155 s	500 ppb	[100]

CRediT authorship contribution statement

John Olabode Akindoyo: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Xiaowen Yuan:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization, Investigation. **Xue Jun Li:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

This work was funded by the Postdoctoral Research Fellowship from the School of Engineering, Computer and Mathematical Sciences, Auckland University of Technology, New Zealand.

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