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Latest developments in NO_2 gas sensors based on PEDOT: PSS nanocomposites and metal oxides: A comprehensive review

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ABSTRACT

Nitrogen dioxide (NO_2) is toxic atmospheric contaminant having serious impacts on human health and environment. The low concentration detection of NO_2 in high accuracy and high sensitivity is still one of the difficult points in air quality monitoring. Recently, hybrid gas sensors based on the conducting polymer PEDOT:PSS and metal oxide semiconductors have been proposed as potential candidates for high-performance NO_2 sensing. Using the p-type polymer and n-type or p-type metal oxides in nanocomposites can lead to a synergetic effect in terms of improved charge transport, sensitivity, and operation at lower temperatures. Recent developments in the area of PEDOT:PSS/metal oxide nanocomposite-based NO2 sensing are reviewed with a critical look at the structural and electronic nature of PEDOT:PSS, gas-sensing mechanism of conventional metal oxides, and importance of interface engineering for device performance. It also highlights eco-friendly synthesis techniques e.g. water-based processing and green synthesis of metal oxides, contributing to sustainable production practices for the sensors developed. Major previous works are summarized and discussed on the basis of important performance largeness, such as the detection limit, response/recovery time/temperature/humidity and environment-friendly processing conditions. Although these hybrid systems present obvious opportunities compared with pure sensors, the issues of long-term stability, selectivity under mixed gas environments and reproducibility of the fabrication approaches are still challenging. In general, PEDOT:PSS/metal oxide nanocomposites offer a viable and a sustainable platform for the next-generation NO_2 gas sensors with lavish sensing performance, eco-compatibility, and prospects to be scaled-up for cost-effective fabrication.

Introduction

Atmospheric pollution due to poisonous gases like nitrogen dioxide (NO_2) has been one of the most serious environmental & public health problem on global scale. As a reactive and oxidizing pollutant, NO_2 is the main contaminant derived from combustion sources such as vehicle engines (exhaust) and industrial activities. NO_2 exposure even at low levels for extended periods can cause significant respiratory diseases and contribute to environmental pollution[1-4].

To solve this issue, gas sensors for showing high sensitivity, selectivity and stability toward NO_2 have drawn wide attention. The use of conventional sensors for signal transmission, especially those used in heat exchangers such as thermocouples and electrical resistance sensors, is almost always subject to some degree of error or delay in signal transmission; therefore, this technology is being adopted [5,6].

Novel sensing materials such as hybrid ones have been becoming the promising strategy in recent years to improve the performance of NO_2 sensors. Especially, the composite material of conducting polymer with metal oxide semiconductor has exhibited significant cooperative effects for improving gas sensing performances. Poly(3,4-ethylenedioxythiophene): poly (styrene sulfonate) (PEDOT:PSS), a typical p-type conducting polymer, has been considered because of its relatively high electrical conductivity, good environmental stability and flexible characteristics, The synergistic effect of combining CdS nanoparticles with PEDOT:PSS resulted in a 2.2-fold increase in sensitivity compared to pure PEDOT:PSS. Intriguing results indicate that the significance of GO-PEDOT:PSS has a synergetic effect in sensor applications [7-8].

Combined with metal oxide nanostructures, e.g., SnO_2, ZnO, NiO, and In_2O_3 PEDOT:PSS can greatly improve charge collection efficacy in the resulting pp Structures at lowered operating temperatures as well reducing response and recovery time. These features render PEDOT:PSS/metal oxide nanocomposites as promising candidates for the new-fangled NO2 gas sensors [9,10].

Despite the remarkable progress achieved in NO_2 gas sensors based on PEDOT:PSS and metal oxide nanocomposites, several fundamental challenges still limit their large-scale and long-term practical implementation. Among these challenges, long-term stability remains a critical issue, as PEDOT:PSS-based sensing layers are prone to environmental degradation, including humidity-induced swelling, polymer aging, and interfacial instability with metal oxides, which can lead to gradual performance deterioration over time. In addition, achieving high selectivity toward NO_2 is still challenging due to the presence of interfering oxidizing and reducing gases that can induce competing charge transfer processes at the sensing interface. These limitations are further intensified under real operating conditions, where variations in temperature and humidity may cause signal drift and poor reproducibility. Consequently, recent research efforts have focused on engineering PEDOT:PSS/metal oxide nanocomposites to improve interfacial charge transport, enhance chemical robustness, and promote selective interaction with NO_2 molecules, thereby addressing these persistent challenges [11-13].

In addition, more attention are devoted to the eco-friendly and sustainable fabrication of such hybrids. Green synthetic methods, for example, water-based processes, natural plant extracts and low-temperature routes, are increasingly being employed in the synthesis of sensors to comply with environmental safety and sustainability during the production process [14–15].

The purpose of this review is to summarize recent enhancements of NO2 gas sensors constructed by PEDOT:PSS and metal oxide nanocomposites. This review communicates a detailed overview of it covering the sensing mechanisms involved, various techniques used for its fabrication (including green approaches), parameters affecting performance and current issues besides literature dealing with development toward efficient and eco-friendly NO_2 sensing devices.

2- Fundamentals of NO_2 Gas Sensing

Nitrogen dioxide (NO_2) is one of highly reactive, toxic and oxidizing gases that are harmful for human health and environment system. From car exhaust and industrial processes to fossil fuel combustion, NO_2 is a major pollutant in urban air that also contributes to acid rain and photochemical smog. Regulators like the World Health Organization (WHO) have established a very stringent exposure limits, being detrimental to human health even down to 40ppb [16-19]. Therefore, on-site, accurate and real-time detection of NO_2 gas has been a high-priority goal in environmental monitoring. Chemiresistive gas sensors are one of the most promising sensing techniques based on simplicity, cost effectiveness, scalability, and compatibility to miniaturization among various sensing technologies [20,21].

The chemiresistive GSSs detect the resistance change caused by the interaction between sensing material and target gas. No2's role as strong electron acceptor is reflected in its adsorption on the surface of a semiconducting material by withdrawing electrons from conduction band, which increases resistance in n-type SiCH film or decreases that in p-type SS(SH)based PEDOT:PSS [22-24].

Sensitivity (i.e., the magnitude of response to a given gas concentration), selectivity (i.e., discriminating NO_2 from other species in mixed gas) are among key efficiency measurements for gas sensors along with the response and recovery times (time-related aspects of signal changes during exposure and removal of target gases), LOD, operating temperature, and long-term stability [23]. It is still a challenge to obtain high performance with respect to these performances, especially for NO_2 because of its low concentration and reactiveness [25, 26].

To cope with such challenges, emerging materials including metal oxide semiconductors and conducting polymers (particular the hybrid ones) have been broadly studies. These nanesheets provide adjustable surface chemistry, high surface−volume ratios, and efficient charge pathways, thereby improving sensing performance. The basic operation of NO 2 detection based on chemiresistive sensors is shown as a model above in Figure 1 with different response between n-type and p-type semiconductors [27-29].

3. PEDOT:PSS: Structure, Properties, and Role in Gas Sensing

One most studied conducting polymer in the field of organic electronics and gas sensor sensing is poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate), generally PEDOT:PSS. It consists of conjugated PEDOT, which gives it electrical conductivity and polyanion PSS (provides water dispersibility and film forming). This special PEC leads to a colloidal gel which can be simply prepared in aqueous solutions, and therefore it is very amenable for low-cost and large-scale process also as spin-coating, inkjet printing, drop casting [30-33].

PEDOT:PSS possesses a repertoire of desirable features (i.e., high electrical conductivity, up to 1000 S/cm after treatment, mechanical flexibility, optical transparency and thermal stability). It could be an excellent candidate for flexible and wearable gas sensors with such properties. In addition, PEDOT:PSS is also p-type semiconductor thus sensitive to the oxidizing gases like NO_2, as it interacts with electron accepting molecules especially those which can change the carrier density of charges and so the conductivity of polymer [34.35.36].

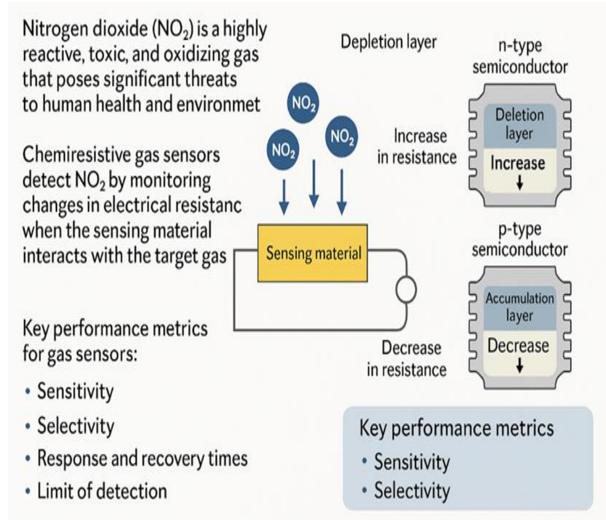

Figure 1: NO_2 sensing mechanism in n-type and p-type semiconductors.

For gas sensing, the behavior of PEDOT:PSS is controlled by the modulation of its hole concentration when contacting with analyte gases. NO_2 molecules attract electrons from the polymer chains and, in turn increasing hole concentration, thereby improving conductivity of the PEDOT:PSS surface. Such a behavior is different from that of n-type metal oxides, which often experience the resistance increase in response to NO_2 [37-39].

Yet, pure PEDOT:PSS has certain shortcomings, like poor selectivity and humidity sensitivity and a relatively low specific surface area that have an impact on the gas adsorption ability. To address these problems, PEDOT:PSS has been hybridized with nanostructured materials such as metal oxides [10–20], carbon nanotubes (CNTs) [21–27], and graphene [28–30]. These composites combine the large surface area and gas adsorption ability of inorganic units with processability and electronic conductivity from polymer, which can bring the enhanced sensitivity, stability, selectivity [40-42].

Thus, PEDOT:PSS functions not only as an active sensing material but also as conductive matrix or charge transport channel for hybrid sensor constructs. Especially, its tunability (by doping, post-treatment, as well as nanocomposite formation) offers a flexible base for the production of novel NO_2 gas sensors. The chemical structure, principal properties, sensing properties, and hybridization possibilities of PEDOT:PSS are described in Figure 2[43-45].

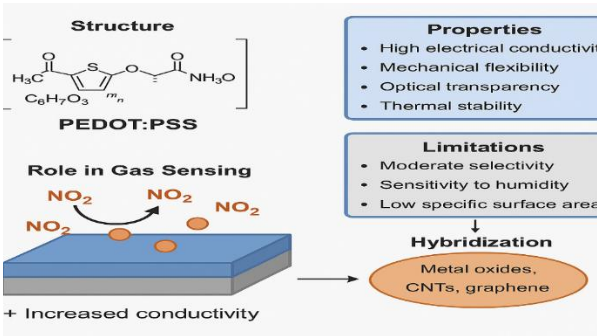

Figure 2: PEDOT:PSS in NO_2 Sensing.

3. Metal Oxide Semiconductors in NO_2 Detection

Metal oxide semiconductors (MOS) are among the most important families of materials in gas sensing since they present great chemical stability, adjustable electronic properties and abundant surface reactivity with gas molecules. Typically used MOS materials for the NO2 sensing are tin oxide (SnO_2), zinc oxide (ZnO), nickel oxide (NiO), tungsten oxide (WO_3), and indium oxide (In_2O_3) which exhibit different band structures, surface chemistries, as well as response characteristics based on their stoichiometry, morphology and growth parameters [46,47].

For MOS, the gas sensing mechanism is based usually on surface adsorption and charge transfer process. Exposure to air/MOS oxygen molecules adsorb on the MOS surface and trap electrons from conduction band, resulting chemisorbed species as O^-, O_2^- or O^{2-}. This results in an electron-depletion layer formed near the surface, and so increases the resistance of material (in n-type MOS) or decreases it (in p-type MOS) [48,49].

When the device is exposed to a strong oxidant gas such as NO_2, molecules of this type react either with the adsorbed oxygen or directly with the solid surface in order to capture electrons and broaden the depleted region on n typesemiconductors like SnO^2 or ZnO. This leads to an appreciable rise in electric resistance. In contrast, in p-type MOS (e.g., NiO and CuO), NO_2 is inclined to extract electrons from valence band, which enhances the hole concentration, thereby lowering the resistance [50,51].

The performance of MOS based NO_2 sensors is dominated by several issues [52-54]:

Morphology and surface size: Nano structures (nanorods, nanowires, hollow spheres) possess large surface-to-volume ratio leading to gas adsorption.

Doping and defect control (malification): The introducing of the dopant (Sb, Al, or rare metal) can effectively tune band structures to enhance charge carrier mobility/selectivity.

Operating temperature: There are many MOS materials that require high operating temperatures (150–300 ° C) for maximum sensor sensitivity because of thermal activation of surface reactions, which will restrict the practical integration into low-power and flexible applications.

However, the MOS-based sensors are still hampered by some deficiencies including high operation temperature, influence of humidity and longer response/recovery times although they display high sensitivity and tunable selectivity. These limitations have stimulated the development of hybrid materials that combine the MOS with other semiconductors, such as conducting polymers or carbon nanostructures (CNs), to take advantage of synergism between chemical–physical properties and increase the overall sensing capability [55,56].

Accordingly, metal oxide semiconductors are a good candidate for NO_2 detection and they can be combined with organic components as PEDOT:PSS, which material paves the way to future sensors whose performance is enhanced under environment conditions. The basic properties and sensing principle of the metal oxide semiconductor for NO_2 detection are outlined in Figure 3 [57,58].

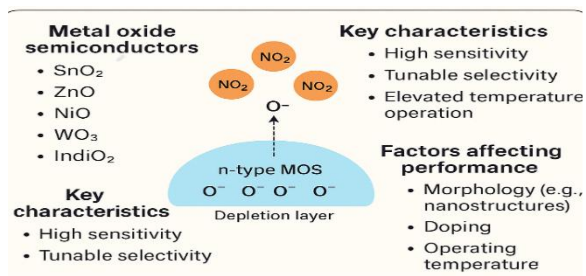

Figure 3: Metal Oxide Semiconductors for NO_2 Sensing.

4. Synergistic PEDOT:PSS/Metal Oxide Nan-ocomposites for NO_2 Sensing

Conducting polymers such as PEDOT:PSS integrated with metal oxide semiconductors resulted hybrid nanocomposites for enhanced performance of gas sensors. These improvements are due to the synergistic effect caused by the polymer-metal oxide interaction, in which their mutually reinforcing properties help overcome each other's limitations [59,60].

4.1 Mechanism of Enhancement

Pathways: PEDOT:PSS acts as a conducting medium that assists rapid carrier exchange between the sensing sites and the electrodes, and reduces the response time The enhancement of NO_2 sensitive performance is driven by several related effects [61-64]:

Heterojunction formation: When p-type PEDOT:PSS is interfaced with n-type metal oxide (SnO_2, ZnO), a p–n heterojunction is established. To yield increased separation of charge and energy barriers due to gas exposure, causing enhanced electrical sensitivity.

Charge transport and recovery times.

Low operating temperature: A conductive polymer provides a reduction in the thermal activation energy for gas interaction, so high-temperature oxides can be operated at room temperature.

Flexibility: The hybrid configuration is compatible with flexible, wearable sensor designs.

4.2 Synthesis Strategies and Their Impact

Table 1: Various synthetic approaches have been developed to prepare PEDOT:PSS/metal oxide nanocomposites, each with advantages and limitations:

Method	Description	Advantages	Limitations
In-situ polymerization [65]	PEDOT polymerized in presence of oxide particles	Strong interfacial adhesion, uniform coating	Complex chemical control
Physical blending [66]	Pre-synthesized PEDOT:PSS and oxide powders mixed	Simplicity, scalability	May suffer from poor dispersion
Layer-by-layer (LbL) [67]	Sequential deposition of polymer and oxide layers	Precise thickness control, reproducibility	Time-consuming process
Sol-gel incorporation [68]	PEDOT:PSS added to oxide precursor solution	Low-temperature processing, good morphology control	Risk of polymer degradation during gelation

The choice of synthesis technique influences [69-71]:

• Particle dispersion within the polymer matrix.
• Interfacial contact area.
• Pore structure and surface area.
• The overall sensor sensitivity, selectivity, and stability.

4.3 Case Examples and Observations

The efficacy of such combination has been recently emphasized in several studies:

• Hydrothermal mixture of PEDOT:PSS/ZnO nanocomposites demonstrated a fast response (<20 s) and high humidity resistance [72].
• SnO_2/PEDOT:PSS hybrids with stable room temperature operation were selective toward NO_2 [72].
• NiO/PEDOT:PSS composites obtained with a green precursor route presented mechanical flexibility as well as reproducibility in several cycles [74].

These cases attest that judicious modulation of the composite interface and morphology can play a key role in exploring excellent sensing servers. The schematic diagram of structure, synthesis approach and functional complementarity of PEDOT:PSS/metal oxide nanocomposite for NO≤₂ detection is presented in Figure 4.

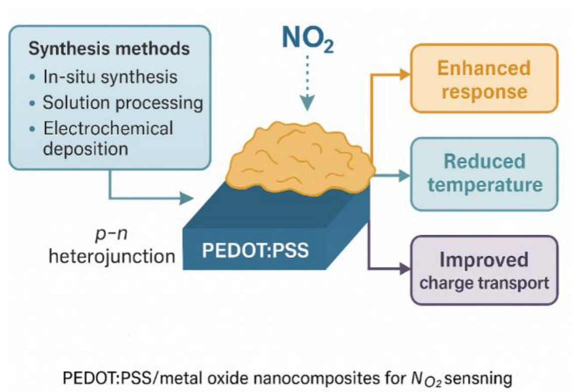

Figure 4: PEDOT:PSS with Metal Oxides.

5. Eco-Friendly Approaches in Fabricating PEDOT:PSS/Metal Oxide Sensors

Due to increasing environmental issues and the demand for sustainable technologies, research related to environmentally friendly fabrication methods of the gas sensing materials has been developed. Particularly, application of green synthesis approaches to PEDOT:PSS/metal oxide nanocomposites turned out to be in good agreement with the principles of green chemistry as it provided pathways for the reduction in energy consumption and hazardous chemicals usage, which contributing to better biocompatibility [75]. Since it is water-processable and dispersible in aqueous solutions, PEDOT:PSS is intrinsically much more environmentally friendly than other conducting polymers that are often processed from organic solvents. Its compatibility with low-temperature processing, such as ink-jet printing, spray coating or solution casting, also helps its implementation in green electronics [76-78]. Meanwhile, the synthesis of metal oxides formerly driven by high temperatures and toxic precursors has been dramatically moving to greener routes. These include: Plant-extract-assisted synthesis: in this method bioactive compounds present in the natural extracts (e.g., tea, leaves or flowers) serve as reducing and capping agents [79].

Sol–gel approaches in aqueous media provide good control of morphology at low temperature [80]. Room-temperature precipitation or hydrothermal method without the calcination process, lowed carbon footprints [81]. These methods can also be used together successfully in hybrid systems. For example, metal oxide nanoparticles produced by green chemistry methods can be incorporated in PEDOT:PSS matrices without subsequent harsh treatment.

These systems not only achieve superior performance as gas sensors but pave the way toward safe, scalable and sustainable preparation methods [82,83]. These results potentially open up an exciting direction for the future research of NO2 gas sensors towards cost, power and environmentally friendly sensor fabrication which is essential for wide applications in urban air monitoring and wearable sensing devices. Schematic illustration of the greentechnology fabrication process for PEDOT:PSS/metal oxide hybrids is shown in Figure 5, which presents natural steps regarding organic and inorganic components to realize high performance gas sensors [84-86].

Observations and Trends Several general trends can be drawn from the studies reviewed:Room temperature operation is already under popular use, the conductivity and softiness of PEDOT:PSS mostly are members to this.ZnO and SnO_2 are still the most in-depth studied metal oxides, while NiO and In_2O_3 form new group of attention because they have high stability and are p-type. Sol–gel, hydrothermal and electrodeposition techniques are more frequently used in fabrication efforts, particularly for eco-friendly low-energy routes of synthesis. Superior performance figures are invariably accompanied by well-dispersed nano-architectures and closer contacts between the polymer and oxide components.

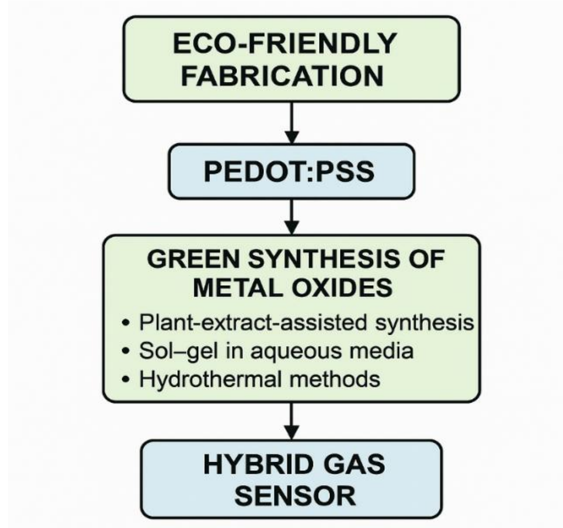

Figure 5: Eco-friendly PEDOT:PSS/Metal Oxide Sensor Design.

Figure 6: PEDOT:PSS/metal oxide nanocomposites for NO_2 sensing.

Conclusions and Future Perspectives

PEDOT:PSS has been found to have a lot of potential in improving the performance of NO_2 gas sensors when inserted with metal oxide semiconductor. These hybrid nanocomposites retain high electrical conductivity, flexibility and room temperature processability of PEDOT:PSS and the strong gas adsorption capacity and surface reactivity of metal oxide. This cooperation led to a remarkable performance of gas sensing properties in terms of high sensitivity, fast response and recovery time with low detection limit. We have summarized the recent progress in material design, fabrication methods and sensing performance. More importantly, more and more researches have been focused on green synthesis owing to the policy of green chemistry. Such work is vital to turn the science laboratory-based gas sensor devices to be practical and sustainable environmental monitoring instrument.

However, although promising, there are a number of outstanding challenges including long term operational stability, reproducibility in fabrication, response to humidity and selectivity under complex gas mixtures. To address these challenges, a better understanding of the charge transfer process at polymer/oxide interfaces and new materials synthesis and device engineering will be needed. Further studies From the prospective, future research should concentrate on :

Multi-gas discrimination using sensor arrays.

Using data-driven methods like machine-learning for signal inference.

Beyond sensitive: flexible and wearable gas sensor with low power consumption.

Bio-based and recyclable materials in sustainable manufacturing. In summary, PEDOT:PSS/metal oxide nanocomposites offer a versatile and promising platform for next generation NO_2 gas sensors that link high performance functionality with environmental consciousness.

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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.

Credit Author Statement

Muatazbullah Ibrahim Abdullah: Conceptualization, Methodology, Investigation, Formal analysis, Data curation, Validation, Visualization, Writing – original draft. Hiyam Majeed Ahmed: Methodology, Investigation, Validation, Writing – review & editing. Abdullah Alaliaan: Methodology, Investigation, Formal analysis. Ahmed H. Ahmed: Investigation, Resources, Data curation. Khalid Saleh: Supervision, Writing – review & editing, Resources. Faiz A. Mohammed: Conceptualization, Supervision, Project administration, Funding acquisition, Writing – review & editing.

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