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A Review on Recent Progress, Challenges and Future Perspectives of Indium Oxide-Based Gas Sensors.

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Abstract: *With rapid industrial growth and urbanization, the release of harmful gases has become a serious threat to human health and the environment. To address this, highly sensitive and selective gas sensors are needed. Indium oxide (In_2O_3) has gained significant attention due to its stability and effectiveness in detecting various gases. This review focuses on the latest advancements in In_2O_3-based gas sensors over the past five years, particularly their response to pollutants like VOCs, NO_x, CO, O_3, NH_3, and SO_2.Among the different synthesis methods, the hydrothermal approach has been widely used for fabricating these sensors. However, a major challenge is that most In_2O_3 sensors require high operating temperatures, limiting their long-term stability and commercialization. To overcome this, researchers have explored noble metal-doped In_2O_3 sensors, which show promise for room-temperature operation.Future developments aim to create intelligent, self-powered gas sensors integrated with smartphones and IoT systems. With Bluetooth-enabled In_2O_3 sensors, gas concentrations can be monitored wirelessly without an external power source. This advancement could enhance applications in wireless signal detection, machine learning-based data processing, and smart gas sensing technology*

Keywords: In_2O_3, Gas sensors, Internet of things, Wireless detection Smartphones

I. INTRODUCTION

Air pollution has emerged as the most significant environmental issue, threatening the health of millions of people as well as the world's ecosystem. According to World Health Organization (WHO) statistics, air pollution alone causes approximately 7 million premature deaths peryear [1,2]. Morbidity and mortality rate increases yearly since environmental gases cause respiratory problems and other diseases. Furthermore, about 99% of the world's population breathes air that exceeds the guideline limit and includes high levels of gas exposure, mostly affecting low- and middle-income countries [3]. Around 3 billion people continue to cook and heat their houses using solid fuels (such as wood, agricultural waste, charcoal, and dung) on direct heat and unreliable stoves. Such inefficient cooking fuels and technologies contribute significantly to indoor air pollution, which contains a broad range of harmful contaminants, including microscopic soot particles that penetrate deep into the lungs. One of the contributors to this poor indoor air quality is the high number (i.e., 1.2 billion) of households, which lack access to electricity [4]. Most sensors are developed to detect and monitor the concentration of hazardous and explosive gases. Semiconductor metal oxide (SMO) sensors are favored among many types of sensors due to their higher sensitivity, low limit of detection (LoD), minimal cost, high energy consumption, high reliability, and quick response time [5]. In comparison with other SMOs, an indium oxide (In2O3)-based gas sensor is one of the capable materials in the field of nanoscience. In2O3, compounds of indium and oxygen, are among the most abundant metal oxides on earth. Indium exists in three different oxides, InO, In2O3, and In3O4 generating unique characteristics [6]. It has been most extensively investigated because of its stability in the general environment [7]. In2O3 is an n-type semiconductor, and it consists of bixbyite-type cubic crystals. It has a body-centered cubic (bcc) structure (a = 10.12 A◦) with an indirect band gap of 2.8 eV and a direct band gap ranging from 3.55–3.75 eV [8]. It attracted a lot of attention because of its technological applications in solar cells, thin-film reflectors that are transparent to visible light, memory devices, window heaters, antistatic coatings,

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photocatalysis sensors, gas sensors, and other products [9]. The In2O3 material is capable of detecting various gases, such as NO2, CO, H2, acetone, etc. at room temperature (RT). Indium (III) oxide took on a variety of morphologies including nanoparticles, nanowires, nanorods, nanobelts, and nanotubes [10]. Many attempts have been made to examine the structural, optical, magnetic, and electrical aspects of In2O3 material [11,12]. Thus, in the current epoch, the Internet of Things (IoT), artificially intelligent systems, and self-powered sensing signify the revolutionary technological advancement of civilization and the foundation of the 4th industrial revolution. With the evolution of the devices based on IoT to 41 billion USD in 2027, the global market on IoT is projected to reach 2.4 trillion USD [13,14]. Above and beyond best-performance electronics, and effective technologies need practical gas sensors with unique characteristics to accomplish IoT anticipated components, like affordable, portable, strong, delicate, etc. With all these projections, to the best of our knowledge, there are no review articles on In2O3, including some of the semiconductor metal oxides-based sensors, which are focusing on the advances in In2O3 sensors [14-20], such as (i) self-powered In2O3 sensors, (ii) wireless detection of toxic and flammable gases and (iii) Artificial intelligent systems, (iv) integration of In2O3 sensors on smartphones and (v) internet of things. Additionally, the current technology is driven by the 4th industrial revolution, which demands self-powered sensors, wireless detection leakage, and Artificial intelligence systems. For instance, to protect the life and health of humankind, the wireless detection network, and self-powered sensing systems augmented with artificially intelligent systems signify a route toward the large-scale deployment of the Internet of Things (IoT). Besides, the proposed gas detection approach by making use of IoT to detect the leakage of toxic, harmful, and flammable gases, will provide a better alert to the user and thus prevent any possible leakage. For instance, Shi et al. [15] reported various ways to improve the sensing performance of In2O3 materials by decorating, doping, and forming heterojunctions using various materials for the detection of ethanol concentration. Zhang et al. [16] performed different synthesis techniques for the composite of 1D micro/nanoscale In2O3 surface structures for instance hydrothermal, electrospinning, and thermal evaporation techniques. Moreover, the fascinating electrical, and optical characteristics were examined in various applications such as gas sensors, ultraviolet sensors, and photocatalysts of In2O3 materials. Kumar et al. [17] studied the surface structure of In2O3-based sensors in pristine, doped, loaded, and heterostructure forms for the detection of H2S gas at various optimum operating temperatures. The fabrication procedures, sensing mechanisms, and prospects of In2O3-based H2S sensors were also assessed. Mahajan et al. [18] investigated the formation and the current advancements of In2O3 nanostructures. The primary objective was based on the new techniques for the synthesis of In2O3-based CO sensors to enhance the sensing characteristics such as sensitivity, selectivity, response, and recovery time using different catalysts. Various morphologies of fabricated In2O3 nanostructures were reported especially nanowires, nanopowders, and nanorods. Recent advancements in the composite gas sensors synthesized by combining different metal oxides, such as heterostructure nanomaterials, and 2D materials, such as transition metal dichalcogenides for the detection of H2 gas were reported by Mondal et al. [19]. The reported discoveries concentrate on the development and exploration of new synthesis approaches for achieving different structural morphologies and improving the sensing performance of metal oxides including In2O3 nanostructures. Yoon et al. [20] reported on the development of flexible and wearable In2O3 sensors using different fabrication techniques that control human movement, the pH level of human sweat, human skin, and human breath. The aim was to improve sensing parameters such as sensitivity, rapid response, and recovery times of metal oxide nanomaterials-based sensors in changing environmental health conditions. Therefore, this review article focuses on the influence of environmental pollutants, such as VOCs, NOX, CO, O3, and SO2 on the environment and human life. Additionally, many studies have been conducted on fabricating different nanostructured In2O3-based gas sensors using various techniques, such as hydrothermal, coprecipitation, sol-gel, electrospinning, etc. Furthermore, the effects of the grain size of In2O3, noble metals, lanthanides, transition metals, transition metals dichalcogenides, polymers, MOFs, and the role of heterostructures interface on the In2O3-based gas sensors were studied. The performance of achieving high sensitivity, selectivity, and stability towards a broad range of concentrations of dangerous gases, such as NH3, H2S, NO2, and CH4 at optimal operational temperatures is examined. The next-generation gas sensors are associated with wireless gas detection, their integration on smartphones, artificially intelligent systems, and self-powered sensing technologies by coupling the triboelectric/piezoelectric devices with In2O3-based sensors are also discussed. Lastly, the Internet of Things is progressing to fabricate highly sensitive,

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selective, and robust gas leakage sensing detectors to offer advanced alerts to end users and evade any risks and possible health threats.

Environmental pollutants:

Environmental pollutants are one of the problems that lead to some of the environmental damage such as air, water, and soil pollution that endangers human life. Air pollution is mainly caused by the release of exhaust gases from industry emissions, automobile, household, and forest fires. The release of exhaust gases, such as SO2, NOX, O3, and CO from industries, and automobile, household, and forest fires are the major causes of air pollution. The effects of releasing smog, which is composed of harmful gases, through photochemical reactions caused the formation of acid, global warming, and ozone (O3) layer depletion.

There are two types of pollutants in the air, which are primary and secondary pollutants. Primary pollutants are released from sources including reducing gases such as H2S, NH3, SO2, CO2, and NO, and oxygenic gases such as CO2, NOx, O2, and CH4. Secondary pollutants are released by industries in the environment including explosive gases such as C2H2, C2H4, C2H6, C3H8, C3H6O, CH2O, etc.

2.1. Volatile organic compounds (VOCs) VOCs are organic compounds with high pressure at RT. VOCs consist of aliphatic hydrocarbons, aldehydes, alcohols, esters, and ethers with various functional groups (halogens, sulfur, oxygen, nitrogen, or phosphorus, without carbon oxides and carbonates) . WHO classifies VOCs as chemicals with a boiling point of less than 250 ◦C measured at 101.3 kPa standard atmospheric pressure. This categorization can be regarded as very volatile organic compounds (VVOCs) with boiling points ranging from 0 to 100 ◦C, which are primarily gases. Examples of those gases are propane, butane, methyl chloride, etc. VOCs with boiling points ranging from 100 to 250 ◦C are found on air and water body surfaces or solid surfaces Organic compounds are subdivided into three types based on their volatility which are VOCs, semi-volatile organic compounds (SVOCs), and non-volatile organic compounds (NVOCs). Examples of SVOCs are formaldehyde, BTEX, styrene, etc. The lower the boiling point of the organic compound due to its greater volatility, the more probable it will be ejected into the air from the surface. It is very challenging to measure the very volatile organic compounds since they are so volatile and are entirely detected as gases in the air rather than in solid form. VOCs play an essential role in the tropospheric atmosphere, generating photochemical O3 and other oxidizing agents that enhance the atmospheric oxidizing ability while seriously affecting the quality of air and the health of human beings. VOCs have been examined in terms of both the environment and human health with several being classified as harmful air pollutants.

Formaldehyde (HCHO)

HCHO is a major carbonyl compound found in indoor environments. HCHO is utilized in phenolic, urea-formaldehyde foam to insulate buildings and polyacetal plastics. Exposure to atmospheric HCHO can cause eye or respiratory irritation, inflammation, and pneumonia.

Electrical properties of In2O3:

In2O3 reveals high electrical conductivity, which relies on creating a non-stoichiometry, external doping temperature of the substrate during deposition, post-annealing, oxygen partial pressure, the thickness of the films, etc. In a film form, its electrical resistivity is in the range of 10^{-4} Ωcm. Its optical transmittance is roughly greater than 90% in the visible region. Generally, In2O3 characteristics are strongly reliant on the synthesis methods. Generally, the lower sheet resistance is observed if the thickness of the films is smaller, however, if the thickness is higher, then higher sheet resistance is observed. The pristine In2O3 could behave as an insulator while the same could be transformed into conducting by forming defects in it or creating a non-stoichiometry in the composition. For pristine In2O3 thin films, individual oxygen vacancy (VO) could provide two faintly bonded unrestricted electrons in its nonstoichiometric arrangement. Therefore, free carrier concentration could be increased when the VO is increased. However, the resistivity will be increased, when the carrier concentration a is increased at a higher doping level because of the electron scattering at the grain boundaries and variation in the crystal structure. The In2O3 system's conduction mechanism is often elucidated by its defect chemistry.

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Artificial intelligence systems Wearable self-powered intelligent systems, which are portable are progressively substituting bulky computers as the interface of a new generation of intelligent human-machine interactions. They play a vital role in intelligent identification, intelligent control, and other fields. The sensor component can generate a lot of information for monitoring environmental factors. Sufficient information may be detected in data processing, particularly machine learning (ML) for improving gas sensing, such as principal component analysis (PCA). The information may be examined in combination with the advancement of the 5th-generation mobile communication technology (5 G), with smart gas sensors, such as MOS(i.e. In2O3) advancing into the era of AI and IoT with the performance of next-generation (NG) gas electronics. The IoTs, Al, and big data technology revolutionize our lives daily. AI was anticipated to aid in the advancement of gas sensing, gas networks, and gas mixtures, enabling low-cost and responsive NG gas sensory information. Lee et al. developed a wireless radio frequenc identification (RFID) system for H2 gas detection. They proved that a wireless communication RFID-reader antenna may achieve high sensitivity (1 ppm). Further studies have been made by Lee et al. presenting a multifunctional gas sensor array for gas detection using a triboelectric nanogenerator and the PCA approach. It proved that PCA can accurately identify H2, CO, and NO2. Zhu et al.

Summary, future trends, and challenges:

This review discusses the latest research made over the last five years on In2O3-based gas sensors. The influence of environmental pollutants, such as VOCs, NOX, CO, O3, NH3, and SO2 on the environment and human life are discussed. The advantages associated with the hydrothermal synthesis approach compared to other synthesis methods have continued to make it the most dominant method for the preparation of sensors based on In2O3. Moreover, the drawbacks and challenges associated with the In2O3 sensors for the last 5 years, showed that most of the sensors have been functioning at higher temperatures. This has led to a lack of commercialization because of reliability issues related to longterm stability drift. To date, doped In2O3 sensors, have shown promising advantages for RT operation. The catalytic interactions of the nanostructures that generate the heterostructures, as well as the stability of the two different nanomaterials, must be fully examined. It was revealed that noble metals with highly effective oxidation catalyst activity had a significant influence on the In2O3 sensor due to improved gas detection performance at low operating temperatures. Thus, more efforts should be considered in the future to explore strategies for improving the doping of In2O3 with various noble metals, in a systematic way to increase the number of sensors to operate at RT. However, the results showed that transition metal-doped In2O3 sensors operate at high temperatures since transition metals are less active at low temperatures. If such attempts are realized, this will provide more advantages for reaching the next generation of intelligent gas sensors from micro/nano materials to the era of a self-powered, artificially intelligent system and their integration on smartphones. This would also be useful in wireless signal detection, in data processing, particularly in ML, to aid in gas sensing. ML plays a role in improving ion mobility analyzers by using a triboelectric-based ionizer that can provide excellent selectivity, ion mobility, and detection of different gases. Self-powered systems have shown promising results in sensing, which is rapidly becoming the dominant form of electronic devices in the IoT era and 5 G mobile communication technologies. The formation of a portable and wearable self-powered system represents a new form of human-machine interaction interface. In the future, more materials and structural designs will be developed to increase stability and create standardized methods for portable wearable devices that can be manufactured. Furthermore, In2O3 gas sensors with Bluetooth modules can make it possible to wirelessly monitor the concentration of different gases without the need for an external power source and transmit information via a smartphone.

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.

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