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Chapter

Sol-Gel Production of Semiconductor Metal Oxides for Gas Sensor Applications

Walid Belaid, Amina Houimi, Shrouk E. Zaki and Mohamed A. Basyooni

Abstract

As they are widely utilized in industries including the food packaging industry, indoor air quality testing, and real-time monitoring of man-made harmful gas emissions to successfully combat global warming, reliable and affordable gas sensors represent enormous market potential. For environmental monitoring, chemical safety regulation, and many industrial applications, the detection of carbon monoxide (CO), carbon dioxide (CO_2), nitrogen dioxide (NO_2), and methane (CH_4) gases is essential. To reliably and quantitatively detect these gases, much-improved materials and methods that are adaptable to various environmental factors are needed using low-cost fabrication techniques such as sol–gel. The advantages of employing metal oxide nanomaterials-based chemoresistive for creating high-performance gas sensors are shown by key metrics such as selectivity, sensitivity, reaction time, and detection. The primary sensing methods are also grouped and thoroughly covered. In light of the current constraints, anticipated future developments in the field of sol–gel nanomaterial-based chemoresistive gas sensors are also highlighted.

Keywords: gas sensor, sol-gel, nanowires, semiconductor metal oxides, thin film

1. Introduction

The sol-gel method has been a topic of interest since its inception as a chemical method for creating glasses at lower temperatures than traditional melting processes. Achieving homogeneity in complex compositions was a requirement that was met by controlling the hydrolysis and condensation reactions of various precursors using different chemical strategies. The method's versatility and precision in controlling material composition, morphology, and properties have made it highly attractive for various applications, such as catalysts, sensors, optics, and energy conversion devices. A wide variety of materials, including inorganic membranes, monolithic glasses and ceramics, thin films, ultrafine powders, and hybrid materials, are frequently produced using the sol-gel process. The hydrolysis of a precursor solution yields suspended colloidal particles as the foundation of the sol–gel technique. Subsequent condensation of the particles leads to the formation of a gel-like substance, which can be further processed to produce the desired material [1–3]. In recent years, the sol–gel technique has emerged as a promising method for depositing gas sensors. This technique offers several advantages over other deposition methods, including low-temperature processing, high purity of the deposited material, and the ability to control the thickness and porosity of the film. Sol-gel-based gas sensors have been used in a wide range of applications, including environmental monitoring, automotive exhaust detection, and medical diagnostics [4–12].

Gas sensors have emerged as an indispensable technology in various industries due to their ability to detect and measure different gases in different environments. For instance, in the petrochemical industry, the detection of hazardous gases such as CO [13, 14], CO_2 [15–19], NO_2 [20, 21], and CH_4 [22, 23] is critical to maintaining safe working conditions and avoiding catastrophic accidents. Similarly, food processing industries require gas sensors to monitor for hazardous gases, such as carbon dioxide, that can accumulate in confined spaces and pose a serious threat to worker safety. Environmental monitoring also relies on gas sensors to detect pollutants and greenhouse gases, like CO_2 and CH_4, that contribute to climate change [24]. In medical applications, gas sensors are used for monitoring gas concentrations in breath analysis and anesthesia delivery. Every year, approximately 3.8 million people suffer from serious and potentially deadly illnesses caused by air pollution. Additionally, household pollution is responsible for roughly 20% of cardiovascular and stroke-related fatalities [25]. Therefore, the importance of gas sensors in different applications cannot be overstated, as they play a significant role in protecting human health and safety [26–32].

The purpose of this chapter is to offer a broad overview of gas sensors prepared using the sol-gel technique for the detection of CO, CO_2, NO_2, and CH_4 gases based on metal oxides. The chapter will provide an introduction to the fundamental principles of gas sensing, the properties of sol-gel films, and the mechanisms of gas detection. Recent advancements in sol-gel technology for the deposition of gas sensors, including the use of various precursors, dopants, and modifiers, will also be discussed. The references cited in this chapter represent a selective but informative collection of papers that describe the fundamental principles of synthesis choices. The goal of the chapter is to offer readers an essential and comprehensive overview of gas-sensing applications using the sol-gel method.

2. Gas sensor classification

Gas sensors can be broadly categorized based on their gas-sensing materials and response. The primary types include optical, electrochemical, and electrical sensors, as demonstrated in an overview of gas sensor classification depicted in **Figure 1**. Optical gas sensors use light to detect the presence of gases by measuring changes in light absorption or scattering [33, 34]. In contrast, electrochemical gas sensors use a chemical reaction to produce an electrical signal that can be measured and interpreted as the concentration of gas [35]. On the other hand, electrical gas sensors use a change in electrical resistance to indicate the presence of a gas by reacting with it and altering its electrical properties [36].

In addition to the gas sensor types mentioned above, a range of additional sensor technologies exists for gas detection purposes, as summarized in **Table 1** which includes information on the principle of detection and typical gases detected for various gas sensor technologies.

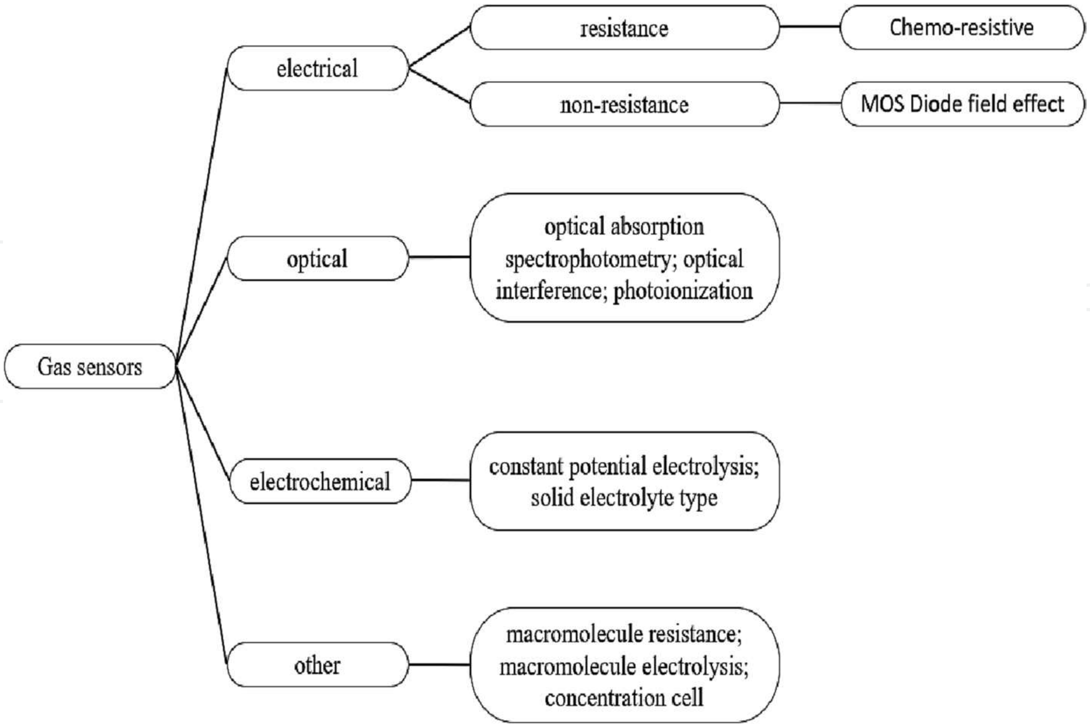

Figure 1.
Overview of gas sensor classification.

Gas sensor technology	Principle of detection	Typical gases detected
Optical	Light absorption or scattering	Carbon monoxide, carbon dioxide, nitrogen dioxide, ozone, sulfur dioxide, volatile organic compounds
Electrochemical	Chemical reaction producing an electrical signal	Carbon monoxide, nitrogen dioxide, hydrogen sulfide, sulfur dioxide, chlorine, ozone
Electrical	Change in electrical resistance	Carbon monoxide, methane, propane, hydrogen, natural gas
Conductometric	Change in electrical conductivity	Flammable and explosive gases
Thermal Conductivity	Change in thermal conductivity	Hydrogen, helium
Piezoelectric	Change in mechanical stress	Volatile organic compounds, other gases
Surface Acoustic Wave	Change in mass or conductivity	Toxic and explosive gases
Photoionization	Ionization of gas molecules by ultraviolet light	Volatile organic compounds, other gases
Solid-state	Change in electrical conductivity or resistance	Carbon monoxide, nitrogen dioxide, methane

Table 1.
Classification of gas sensor technologies based on the principle of detection, with examples of typical gases detected.

Currently, there is an extensive research focused on the development of electrical nanosensors with enhanced performance [37–39]. One type of electrical nanosensor is the resistive gas sensor [40], which is an electronic device that alters its electrical resistance according to the surrounding gas ambient. Resistive gas sensors are categorized into two types based on their operating principle: chemical and physical [41]. Physical resistive gas sensors are capable of detecting changes in the physical properties of the gas, such as temperature, pressure, or mass, while chemoresistive gas sensors [42] employ a sensing layer to interact with the target gas. Although the fabrication and optimization of resistive gas sensors remain challenging due to the complexity of the materials involved and the interactions between the gas and sensing layer, chemoresistive sensors are widely used for gas detection due to their high sensitivity and selectivity. They offer several advantages, such as low cost, low power consumption, and miniaturization capability, making them suitable for various portable and wearable applications. Chemoresistive sensors are also highly sensitive and selective toward specific gases, enabling them to detect even low concentrations of gases [43]. In the next section, we will delve deeper into the mechanisms and applications of chemoresistive gas sensors, which will shed more light on their importance in the field of gas sensing.

3. Chemoresistive gas sensors: mechanisms, advantages, and applications

Chemoresistive gas sensors utilize changes in the electrical conductivity or resistance of the sensing material to detect various chemical or gaseous analytes. Typically, the sensing material is composed of a semiconductor made from a metal oxide, which may be classified as either p-type or n-type.

In n-type gas-sensing materials, the presence of gas molecules leads to the formation of surface states, which capture electrons from the conduction band and reduce the free carrier concentration, increasing resistance. The magnitude of this change in resistance depends on several factors, including the concentration and type of gas, the temperature, and the properties of the sensing material. In p-type gas-sensing materials, the presence of gas molecules leads to the release of holes from the valence band, which increases the free carrier concentration and result in a decrease in resistance [44]. As with n-type sensing materials, the magnitude of this change in resistance depends on several factors, including the concentration and type of gas, the temperature, and the properties of the sensing material [45].

Both n and p types of gas-sensing materials can be used in chemoresistive gas sensors to detect a wide variety of gases and chemical analytes. The performance of these sensors can be affected by factors such as the method of sensing material deposition, the type of sensing material used, and the operating temperature. However, ongoing research in this area continues to improve the sensitivity, selectivity, and other performance characteristics of chemoresistive gas sensors [46]. Additionally, creating a junction between p-type and n-type oxide semiconductors using various contact arrangements can offer innovative approaches to developing gas sensors as reported in an excellent review on nanoscale metal oxide-based heterojunctions for gas sensing by Miller et al. [47]. Chemoresistive gas sensors offer several advantages, such as simplicity, low cost, and potential for miniaturization. Compared to other gas sensors, they do not require reference electrodes, which can complicate the sensor design and increase the production cost. Additionally, chemoresistive gas sensors offer fast response times and can detect gases over a wide concentration range [38, 48]. However, chemoresistive gas sensors may have some limitations compared to other types of sensors. For instance, they may not offer the same level of selectivity as electrochemical sensors, which use a chemical reaction to detect specific gases.

Additionally, chemoresistive gas sensors may not be able to detect certain types of gases that are detectable using optical sensors, which use light to detect changes in the concentration of gases. Despite these limitations, chemoresistive gas sensors are widely used in a range of applications, including medical diagnosis [49], environmental monitoring [50], and industrial safety [51]. Ongoing research continues to improve the sensing performance and capabilities of chemoresistive gas sensors, making them a promising option for gas detection in various fields.

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4. Gas sensing operating temperatures

Temperature is a crucial aspect of gas sensing and has a significant impact on the performance of gas sensors. The sensitivity and selectivity of gas sensors vary depending on the temperature range in which they operate. Operating temperatures that are high can heighten the sensitivity and reaction rates of gas sensors. However, they may lead to thermal drift and instability. On the other hand, low operating temperatures can improve sensor stability but decrease sensitivity and response time. Due to their larger bandgap, metal oxide sensors typically require high operating temperatures between 100 and 450°C for surface redox reactions and reaction kinetics to facilitate sensing measurements [52, 53]. Nevertheless, this high-temperature operation has limitations in terms of energy conservation, application, and potential hazards such as gas explosions and sensor instability. Therefore, operating gas sensors at room temperature is highly desirable to minimize the risk of a gas explosion, decrease energy consumption, increase sensor life, and enable integration into smartphone devices [25].

The two dimensional transition metal dichalcogenides [54, 55] and low-dimensional structures [56] generally operate at lower temperatures compared to metal oxide sensors, typically in the range of 25–200°C. This lower-temperature operation can offer advantages such as reduced power consumption, improved stability, and wider application in portable devices. However, it can also lead to lower sensitivity and slower response times compared to sensors operating at higher temperatures. Therefore, understanding and optimizing the operating temperature of gas sensors is crucial for achieving accurate and reliable gas detection. The gas-sensing performance of sol-gel deposited gas sensors is highly dependent on the operating temperature, which affects the sensor's sensitivity, selectivity, response time, recovery time, and ability to detect specific gases. **Table 2** summarizes the advantages and disadvantages of different gas-sensing operating temperatures.

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5. Metal oxides nanomaterials for gas-sensing applications

The development of gas-sensing materials with high sensitivity and selectivity has been a significant research focus in recent years. Among the various synthesis methods, the sol-gel technique has gained important attention as a promising approach for depositing different types of materials onto various substrates. A schematic view of the sol-gel process for the development of ZnO-based films is given in **Figure 2**. Sol-gel processing offers several advantages, such as low-temperature processing, precise control over the deposited material's composition and morphology, and scalability. By tailoring the deposition parameters and optimizing the material properties, sol-gel-deposited gas-sensing materials have shown superior sensing performance compared to conventionally synthesized materials.

Temperature	Advantages	Disadvantages
Low	• Increases efficiency and lifespan of some electronic and mechanical components • Improves stability and accuracy of some sensors • Reduces risk of thermal runaway and fire hazards in some applications	• Increases viscosity or freezing of some fluids • Reduces battery capacity • Increases susceptibility to condensation and moisture • Some materials become brittle or lose their elasticity
Room Temperature	• Most convenient and cost-effective option • Most components are designed and tested for room-temperature operation • Many sensors and instruments optimized for performance and stability at this range • Comfortable and safe for human operators	• Reduced sensitivity or selectivity of some sensors • Increased noise or drift in some electronic components • Reduced efficiency or performance in some energy conversion systems
High	• Increases efficiency and power density of some energy conversion systems • Improves performance and sensitivity of some sensors • Reduces risk of contamination or corrosion in some applications	• Increases thermal stress and mechanical wear on some components • Reduces the lifetime and stability of some sensors and electronic devices • Increases risk of thermal runaway or fire hazards in some systems • Requires specialized materials and thermal management, which increases the cost and complexity of the system

Table 2.
Advantages and disadvantages of different gas-sensing operating temperatures.

Metal oxide nanomaterials deposited by the sol-gel method have been used for the detection of a wide range of gases, including toxic gases, flammable gases, and environmental pollutants. These sensors have several advantages, including high sensitivity, fast response time, low-power consumption, and the ability to operate at room temperature [57, 58]. Metal oxide-based gas sensors have been extensively studied and are currently the most investigated type of gas sensor. Recently, there has been a growing trend in using these materials with sizes ranging from 1 to 100 nm for gas sensing due to their size-dependent properties [59, 60]. These nanomaterials possess unique mechanical, optical, electrical, catalytic, and magnetic properties, and have high surface area per unit mass with new emerging physical and chemical properties. As the size of the material decreases, the specific surface area and surface-to-volume ratio increase significantly. Additionally, the size and geometry of the semiconductor nanomaterials can affect the movement of electrons and holes. The greenhouse gases sensors are largely studied using metal oxide materials.

5.1 Carbon dioxide

Gas sensors can be broadly categorized based on their gas-sensing materials and response. The primary types include optical, electrochemical, and electrical

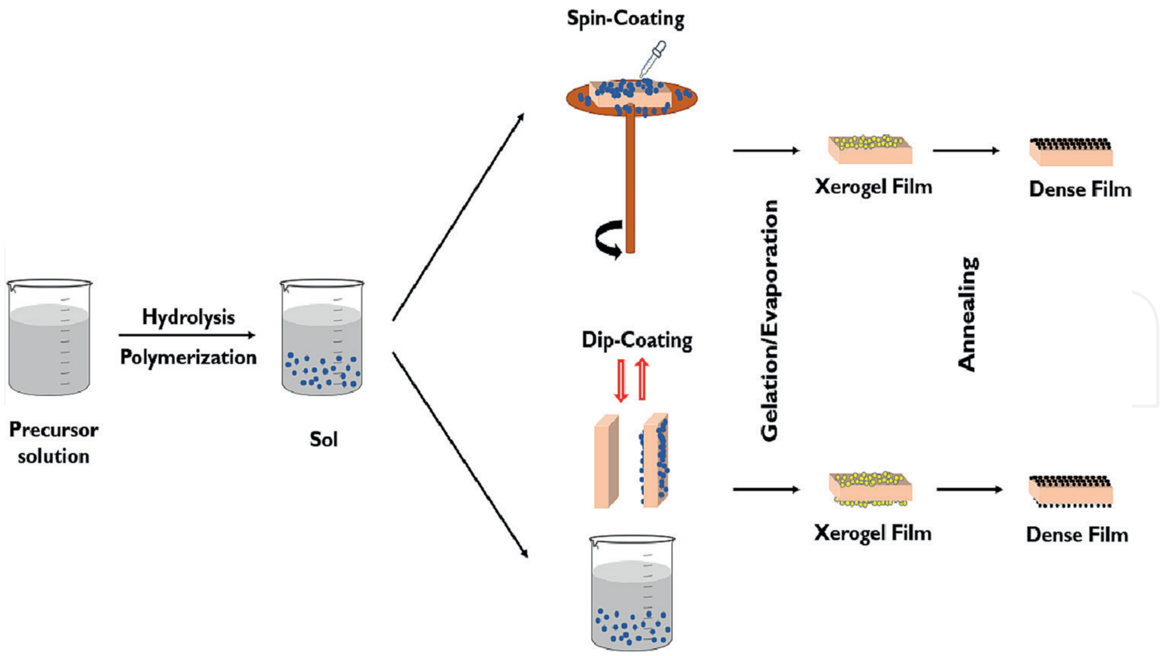

Figure 2.
Schematic of sol-gel process for ZnO thin film deposition.

sensors, as demonstrated in **Table 1**. CO_2 emissions contribute significantly to global warming and climate change, making it crucial to detect and regulate its emission. Metal oxide-based CO_2 sensors can be cost-effective and efficient to measure CO_2 levels even at room temperature (**Table 3**). Nanostructured nickel ferrite ($NiFe_2O_4$) thin films were tested as liquefied CO_2 gas sensors at a low temperature of 27°C [66]. The thin films, deposited using the spin-coating technique, had 1.3 sensitivity at 1000 ppm CO_2 levels with 100 and 400 s as response and recovery time, respectively. Panday et al. [61] tested another metal oxide for CO_2 detection. Sb-doped SnO_2 nanostructured thin films were deposited using the same technique with a 450°C annealing temperature. At room temperature (30°C), the nondoped SnO_2 thin film exhibits an optimum gas response of 78.5%.

Material	Shape	Concentration (ppm)	Response (R)/ Sensitivity (S)	Response/ Recover time	Temperature (°C)	Ref.
SnO_2:Sb	Nanostructured thin films	—	1.78 (R)	2.6/5.8 s	30	[61]
ZnO/Na	Nanostructured thin film	50	81.9 (R)	283/472 s	RT	[62]
TiO_2:PANI	Thin film	1000	53 (R)	9.2/5.7 min	30	[63]
Y@ ZnO:CdO	Thin film	500	9 (R)	4/2 s	RT	[64]
SnO_2:Co_3O_4	Nanocomposite	—	13.68 (R)	2/12 s	30	[18]
$PrFeO_3$	Nanopowder	1000	8.44 (R)	—	160	[65]
$NiFe_2O_4$	Nanostructured thin films	1000	1.3 (S)	100/400 s	27	[66]

Table 3.
Comparison of metal oxides based-CO_2 gas sensing performance.

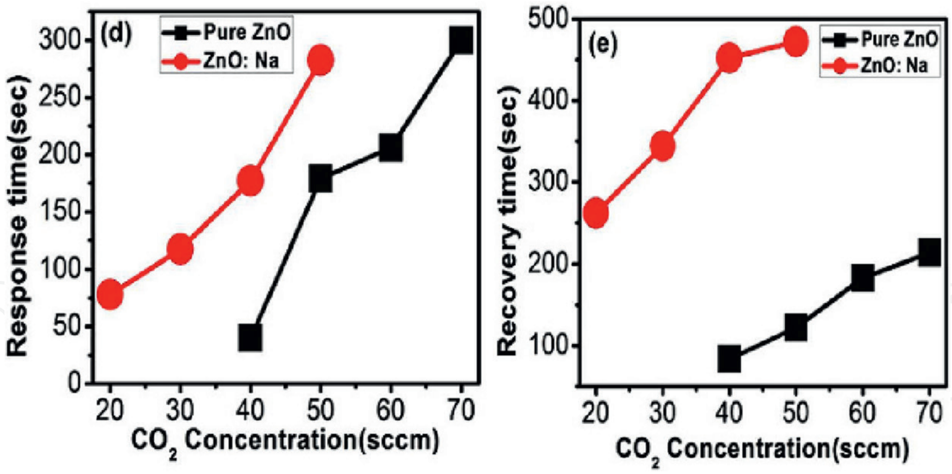

Figure 3.
Response/recovery time of ZnO and Na-ZnO-based CO_2 gas sensors [62].

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Y-doped-ZnO: CdO nanocomposite thin films prepared by sol-gel spin coating technique also proved its efficiency for CO_2 detection [64]. With their cauliflower-like morphology, these metal oxides showed a sensor response of nine at room temperature. A mixture of two metal oxides was also used for the detection of CO_2 gas in the work presented by Joshi et al. [18]. The nanocomposite porous mixture of SnO_2-Co_3O_4 (1:2) thin films showed a high response of 13.68 at a temperature of 30°C compared to the other used ratios. Response/recovery time is an important parameter indicating how fast the sensor can detect the gas, for instance, toward CO_2 as it is the most important greenhouse gas **Figure 3** for ZnO [62].

5.2 Carbon monoxide

CO gas detection is crucial in protecting against the potentially deadly effects of carbon monoxide poisoning, which can be caused by the incomplete combustion of fossil fuels. Early detection can save lives and prevent serious health consequences, making CO gas detection a vital component of any safety plan. Metal oxides synthesized using the sol-gel technique were largely investigated for CO-sensing properties (see **Table 4**).

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Material	Shape	Concentration (ppm)	Response (R)/ Sensitivity (S)	Response/ Recover time	Temperature	Ref.
SnO_2:Sb	WO_3	50–500	1.01–5.67 (S)	200°C	200°C	[67]
ZnO/Na	In/Pd@SnO_2	1	3 (R)	15/22 s	140°C	[68]
TiO_2:PANI	TiO_2:CeO_2	400	16.1 (R)	32/45 s	200°C	[69]
Y@ ZnO:CdO	TiO_2–ZrO_2	100	9.1 (R)	42/48 s	150°C	[70]
SnO_2:Co_3O_4	Au@ In_2O_3	5	104 (S)	130/50 s	RT	[71]
$PrFeO_3$	TiO_2/ perovskite	400	38.41% (R)	—	200°C	[72]

Table 4.
Comparison of metal oxides based-CO gas sensing performance.

Susanti et al. [67] reported in their work the detection of 50 ppm of CO gas at 200°C by WO_3 nanomaterial. The best results were achieved when the thin film's calcination temperature reached 500°C. In another work, CO gas was detected at room temperature by sol-gel-synthesized WO_3 thin films [73]. The deposition of AuNP was ensured by the dip-coating technique which resulted in high coverage of In_2O_3 nanowires treated with SAM (assembled monolayer) layer. As a result, they witnessed a great enhancement in CO oxidation and an improvement in the sensing capabilities of the device at room temperature [71]. Mixtures of Perovskite oxide ($La_{0.8}Sr_{0.2}Co_{0.5}Ni_{0.5}O_3$) with other metal oxides (ZnO: Al [74], ZnO [75], TiO_2 [72], SnO_2 [76]) were reported as well for the detection of carbon monoxide at relatively low temperatures that ranges between 180 and 200°C.

5.3 Nitrogen dioxide

NO_2 gas detection is crucial for identifying and mitigating the harmful effects of nitrogen dioxide on human health and the environment. It allows for prompt action to be taken to reduce emissions, protect public health, and promote clean air. Overall, NO_2 gas detection is vital for safeguarding well-being and ensuring a healthy environment. We present in **Table 5** a performance comparison of NO_2 gas sensors.

Due to their unique effective surface area, WO_3 nanostructures such as nanoparticles have been examined as excellent candidates for gas sensors at lower temperatures. In a study done by Yan et al. [77], WO_3 nanoparticles were synthesized using a sol-gel method onto porous silicon and alumina substrates. Gas sensing tests showed that the WO_3 nanoparticles/porous silicon exhibited improved NO_2-sensing properties at room temperature compared to WO_3 on alumina. In another study, using a sol-gel method graphene-wrapped WO_3 nanosphere composite was synthesized [83]. This composite exhibited p-type gas-sensing behavior, with a linear response to NO_2 concentration at room temperature. Tin dioxide showed a good response toward NO_2 gas in different works [81, 82]. Recently, Kumar et al. [82] reported a remarkably high sensitivity of SnO_2 metal oxide at room temperature toward 2 ppm of NO_2 gas.

Material	Shape	Concentration (ppm)	Response (R)/ Sensitivity(S)	Response/ Recover time	Temperature	Ref.
WO_3	Nanoparticle on porous silicon	2	3.27 (R)	2/>20 min	RT	[77]
Ce @NiO	Nanostructured thin film	40	29% (S)	62/595 s	150°C	[78]
Pt@ZnO	Thin film	1	1.02 (S)	420/660 s	200°C	[79]
WO_3: In_2O_3	Mixed nanocomposites	1	~99% (S)	16/– min	~140°C	[80]
In @SnO_2	Nanoparticles	500	72 (R)	—	150°C	[81]
SnO_2	Nanoparticles	2	8.44 (R)	184/432 s	RT	[82]

Table 5.
Comparison of metal oxides based-NO_2 gas-sensing performance.

5.4 Methane

Natural gas's main component, CH_4 gas, is an odorless, colorless, and extremely combustible gas. It frequently serves as a fuel source for appliances that heat and prepare food, as well as for industrial activities. The dangers posed by methane gas make its detection crucial. In small places, it can potentially replace oxygen, causing asphyxiation. Methane gas is also a strong greenhouse gas that, when released into the atmosphere, accelerates climate change.

Metal oxides synthesized with the sol-gel technique played an important role in improving CH_4 gas sensors (**Table 6**). A large number of works investigated the possibility of applying metal oxides synthesized by the sol-gel method for methane gas sensors (**Table 6**); however, only a few of them are operating at low temperatures (T < 200°C).

Abruzzi et al. [84] SnO_2 manage to obtain very good performance from SnO_2 nanomaterials-based CH_4 sensors using the sol-gel method. At 80°C operation temperature, the response and recovery times were 29 and 47 s, respectively. ZnO thin films were also tested for methane gas sensors using the sol-gel deposition technique [85]. ZnO nanocrystalline thin films were modified using a Pd atom in the work presented by Bhattacharyya et al. [85]. They reported a good response of ZnO nanocrystalline deposited on SiO_2/n-Si. The Pd-modified ZnO thin films showed lower sensing T of 150°C compared to the unmodified thin films (250°C). Also, it displayed lower response and recovery times.

The production of a nanowrinkled $Zn_{0.92}Fe_{0.08}O$ thin film (see **Figure 4**) utilizing the high rpm electro-spin patterning technology via sol-gel approach is discussed in the work presented by Anchal et al. [86]. The film's nanostructure is homogenous and smooth, making it good for gas adsorption and sensing activities. With a rise in gas concentration and operating temperature, the sensor responds more quickly. The maximum response, which outperforms the ZnO flat thin film-based sensor, is 83.4% for 500 ppm of methane at 200°C. Perovskite oxides based on rare earth metals ($LnFeO_3$) are also being researched for use as methane gas sensors [87].

Material	Shape	Concentration	Response (R)/ Sensitivity (S)	Response/ Recover time	Temperature	Ref.
SnO_2	Nanoparticles	20,000 ppm	61 (%)	29/47 s	80°C	[84]
Pd modified ZnO	Nanocrystalline thin films	1%	87.2%	6/33 s	150°C	[85]
$Zn_{0.92}Fe_{0.08}O$	Nano-wrinkled thin film	500 ppm	83.4%	96/107 s	200°C	[86]
$SmFeO_2$	Nano-crystalline	200 ppm	22% (R)	—	200°C	[87]
SiNWs/TiO2	Core-shell nanostructure	120 ppm	182% (R)	75/191 s	RT	[88]
CoO	Thin films	2000 ppm	—	250/250 s	210°C	[89]

Table 6.
Comparison of metal oxides based-CH_4 gas-sensing performance.

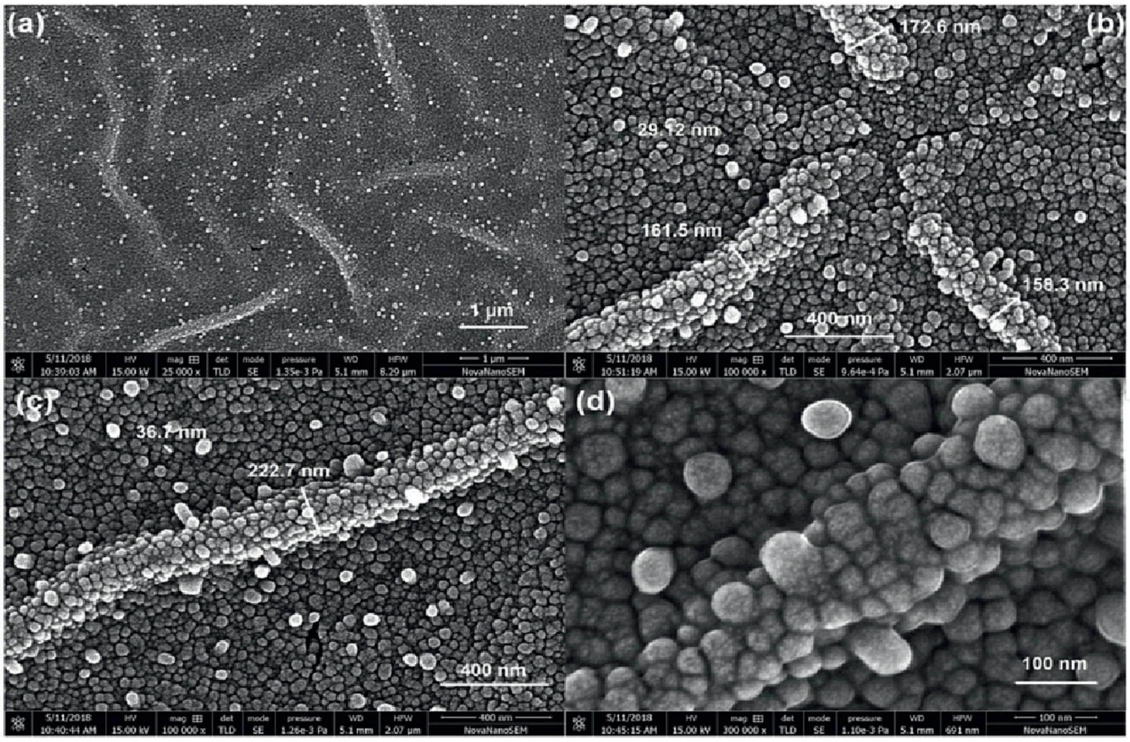

Figure 4.
FE-SEM of nanowrinkled $Zn_{0.92}Fe_{0.08}O$ thin film showing (a) large area view, (b) enlarged structure, (c) grown single wrinkle, (d) wrinkle surface and grain/grain boundary structure [86].

6. Conclusions

The field of sol–gel nanomaterial-based chemoresistive gas sensors especially based on metal oxides has been rapidly evolving in recent years. These sensors have great potential for use in a wide range of applications, including environmental monitoring, medical diagnosis, and industrial process control. One of the current problems in this field is the lack of selectivity of some sensors, which can result in false positives or negatives. It is working to improve the selectivity of these sensors by developing new materials with specific binding sites for target gases. Another challenge is the stability and reproducibility of the sensors, which can be affected by variations in processing conditions and environmental factors such as temperature and humidity. In terms of anticipated future developments, one promising area is the integration of sol-gel nanomaterial-based gas sensors with other technologies, such as microelectronics and wireless communication. This could enable the development of smart sensing systems that can remotely monitor and analyze gas concentrations in real-time. Another area of potential development is the use of sol-gel nanomaterials for the detection of other types of analytes, such as volatile organic compounds and biological molecules. This could open up new applications for these sensors in areas such as food safety, healthcare, and homeland security. Overall, while there are still some challenges to be addressed, the field of sol-gel nanomaterial-based chemoresistive gas sensors holds great promise for the development of highly selective, stable, and versatile sensing systems in the future.

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All authors equally contributed to this study including the conception and design of the work; the acquisition, analysis, and interpretation of data for the work; participating in drafting or revising the work; and approving the final version of the work to be published.

Conflict of interest

The authors declare no conflict of interest.

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