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A Review of Semiconductor Metal Oxides for Hydrogen Sulfide and Methane Gas Detection

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Abstract

This review contains comprehensive study of semiconductor metal oxide films gas sensors. SMO are smaller in size, simple fabrication technique, sensitivity and selectivity is good. Review study paper offers understanding about the dopants impurity made the deviations in the SMO materials used to fabrication of gas sensor. Reviews study shows SnO_2-WO_3, ZnO, Au composites good for H2S and methane (CH4) gases it has sensitivity in the range 75 % to 90% at operating temperature between 200 0C to 350 0C. Composites of ZnO-Au, SnO_2, TiO_2 shows decent sensitivity in the range between 72 % to 95% at operating temperature between 200 0C to 350 0C towards H2S and methane (CH4) gases. Pure and composite of different SMO films prepared using Sol-Gel, Sputtering, CVD and Screen printing method result shows Screen printing technique is most preferred and it shows increase in sensitivity at lower operating temperature. This review provides summary of the advance and performance of pure and composites of ZnO and SnO2 based films sensors for H_2S and CH_4 detection, focusing on key parameters such as response time, operating temperature, sensitivity and fabrication technologies. It provides detail analysis of the pure and composite of SnO2 &ZnO towards H_2S and CH_4 and shows future perspective towards increase in sensitivity, lower operating temperature, response time and stability of gas sensor.

Keywords: Semiconductor Metal Oxides, Pure and Composite of ZnO, SnO2, Thin Films, Thick Films, Hydrogen Sulfide, Methane, Gas Sensors.

1. Introduction

Last few decades, rapidly increase in populations needs to increase the industrial development and urbanization. Air pollution is a main issue that the world is currently dealing with the introduction of various hazardous and polluting gases into the atmosphere. Some of these contaminants contain poisonous gases like H2S, SO2, NO2, CO and Volatile organic compounds (VOC) such as ethanol, toluene, benzene etc., such a gases and VOC are harmful to human being and the atmosphere [1-3]. Air pollutions affected nearly 3.8 million people per year face various illnesses, also near 20% of death due to stroke and 20% death of the cardiovascular [4]. In addition, toxic gases seriously affect the breathing tract and deleterious to immune and nervous system [5, 6]. Hydrogen Sulfide (H2S) is a toxic and inflammable gas, it mostly originated from industrial and natural processes such as coal, gas manufacturing, natural gases, garbage, sewage. H2S is deleterious gas which highly effect on human nervous system at higher concentration more than 250 ppm cause death. Consequently, controlling and monitoring of H2S gas is crucial in laboratories and industrial areas [7, 8]. NO2 is a common and hazardous gas that is generated by both man-made activities like power generation, vehicles and industrial boilers, as well as natural activity such as lightning and volcanic eruptions [9]. NO2 effects the damage of ozone layer which cause acid rain which are hazardous to people as well as atmosphere [10]. NO2 causes respiratory and cardiovascular diseases for long exposure to low concentration [11]. Methane (CH4) is second richest colorless and odorless greenhouse gas next to CO2. Methane used as fuel for electric power production all over the world as compared to coal. After combustion it is less toxic and hazardous substances. It is important source for low-carbon economy [12]. CH4 is extremely combustible so that monitoring is extremely important for safety purpose. It is released during mining and coal extractions [13-15]. CH4 gas detection is important for environmental and industrial safety in regard to transportation, gas production and consumption. Gupta & Singh et. al., identifying gases like hydrogen sulfide (H2S) and methane (CH4) (Singh & Gupta,2019) [16]. semiconductor metal oxides (SMOs) are commonly utilized in gas sensor applications (Kumar et al., 2020). In past two-decade large advancement has been achieved in gas sensing technology. SMOs in thin and thick film forms, particularly for detecting H2S and CH4 (Jain et al., 2021) [17]. Semiconductor metal oxides (SMOs) are increasingly used as the main material in gas sensor development, particularly for identifying hazardous and combustible gases.

2. Need of Gas sensor

In environment, air we inhale contains several artificial and natural chemical species placed in which some gases are essential for life and some of them are harmful [18]. Now a day, industrial productions of hazardous ingredients, mostly dangerous and flammable gases. Unavoidable, sometime leakage of the gas occurs, which produce hazards to the industries and societies living close to industries. Globally several such an incidents of gas leakage, bursts and loss of life are the notice of this difficulty. Hence, gas sensor plays key solution to above stated problems, which show an important part in different recent technology processes, wherever control and detection of gases is required [19]. In 1984, Indian has witnessed that Bhopal gas tragedy, methyl isocyanides leakage from union carbide company. This incident resulted about 8000 people death and 5,00,000 people affected. Increasing air pollution, industrialization, urbanization, use of motor vehicles, are the major sources of increase in gas pollutants above threshold level as a result several diseases from pollution [20]. According to report, seven million deaths occur due to air pollution [21]. Air we live contains 20.9% oxygen, 78% nitrogen, 1% remaining gases. Overexposure of nitrogen leads to shortfall of oxygen and suffocation occurs. When concentration of oxygen drops below 16% it becomes inadequate for animals and humans for respiration. Hence, to protect from dangerous effects need to develop effective gas sensor [22].

3. Experimental Techniques for Thin and Thick Films

Several experimental methods have been employed to fabricate pure and composite SnO_2 thin and thick films, each influencing the morphology, thickness, and gas-sensing performance:

3.1 Sol-Gel Method: This sol-gel technique is broadly used for preparing SnO_2 thin films due to its simplicity (Rani et al., 2020) [23]. Studies indicated this method used for fabricating ZnO thin films due to its simplicity (Kumar et al., 2019).

3.2 Chemical Vapor Deposition (CVD): Jin et al., and patil et al., studied Chemical Vapor Deposition is good technique for depositing SnO_2 thin films with excellent homogeneity it provides excellent crystalline structure (Jin et al., 2019, Patil et al., 2020).

3.3 Sputtering: Sputtering is widely used for creating both thin and thick SnO_2 films (Patil et al., 2020). A versatile technique used for both thin and thick films, allowing the deposition of ZnO and composite films with precise thickness control (Singh et al., 2018) [24].

3.4 Screen Printing: It is a common method for fabricating thick films, allowing the deposition of SnO_2 pastes onto substrates to create sensors with high surface area (Goswami&Goswami, 2020) [25].

4. Semiconductor Metal Oxide Materials for Gas Sensing

Semiconductor metal oxide sensors are easier for fabrication and minor production cost for gas detection. Sensor changes their resistance when it comes in contact with concentrated gas [26]. SMOs materials such as ZnO, SnO_2, TiO_2, WO_3, and In_2O_3 have been usually used for gas sensing applications. These materials are preferred due to their chemical stability, electronic properties and high surface area which are crucial for detecting trace amounts of gases.

4.1 SnO_2-Based Thin and Thick Films for Gas Sensing

Tin dioxide (SnO_2) is a n-type semiconductor metal oxide, widely used due to its excellent sensitivity to reducing gases effective in gas sensor applications (Singh et al., 2020) [27]. Yadav et al., studies development of SnO_2 in the form of thin and thick films particularly for improving gas sensor performance (Yadav et al., 2019) [28]. Tin oxide has been a cornerstone in gas sensor technology due to its excellent electrical conductivity and stability (Okamoto et al., 2003) [29]. Early studies focused on optimizing SnO_2-based sensors through adjustments in operating temperatures and surface properties (Tian et al., 2005) [30]. More recent research has explored doping SnO_2 with transition metals such as Pt, Pd, and Au to enhance its gas-sensing capabilities (Zhao et al., 2007) [31]. These dopants alter the electronic properties of SnO_2, leading to improved sensitivity and selectivity (Liu et al., 2010) [32].

4.1.1 Pure SnO_2 Thin and Thick Films

Pure SnO_2 thin and thick films are commonly used in gas sensors because of larger ratio of surface-to-volume and ability to change electrical resistance upon exposure to target gases (Kumar &Ramaprabhu, 2020). The sensing process primarily includes the adsorption of oxygen molecules on SnO_2 surface, which trap electrons and create a depletion layer, increasing the film's resistance (Gupta & Sharma, 2017) [33]. reducing gases like H_2S and CH_4, these gases react with the adsorbed oxygen changes in resistance in films (Wang et al., 2019) [34].

4.1.2 Composite SnO_2 Thin and Thick Films

For enhancing performance of gas sensor like response time, selectivity, sensitivity and reducing the operating temperature needs to Compositing SnO_2 with other materials such as Au, Pd, and other metal oxides (e.g., ZnO, WO_3) or carbon-based materials (Zhang et al., 2020) [35]. Liu et al., studies shows the improve in surface properties of SnO_2 for making them more responsive for H_2S and Methane gas (Liu et al., 2018) [36].

Table 1: Sensitivity, Response time of Pure and Composite SnO_2Films

Sr. No	**Year**	**Material**	**Experimental Technology**	**Gas Detected**	**Sensitivity**	**Operating Temperature (°C)**	**Response Time (s)**	**Reference**
1	2012	Pure SnO_2 Thin Film	Sol-Gel	H_2S	80% (5 ppm)	300	25	Gupta & Sharma, 2017 [33]
2	2016	Pure SnO_2 Thick Film	Screen Printing	CH_4	75% (100 ppm)	350	30	Yadav et al., 2019 [28]
3	2018	SnO_2-Au Composite Thin Film	CVD	H_2S	92% (3 ppm)	200	20	Jin et al., 2019 [37]
4	2020	SnO_2-ZnO Composite Thick Film	Sputtering	CH_4	88% (50 ppm)	250	22	Patil et al., 2020 [38]
5	2021	SnO_2-Graphene Composite Thin Film	Sputtering	H_2S	95% (2 ppm)	150	15	Zhang et al., 2020 [35]
6	2019	SnO_2-WO_3 Composite Thick Film	Screen Printing	CH_4	90% (30 ppm)	200	18	Liu et al., 2018 [36]

In above table pure SnO2 thin film prepared using Sol-Gel method shows 80% sensitivity towards H2S gas at 300 0C operating temperature (Gupta & Sharma, 2017). Pure SnO2 thick film prepared using Screen Printing method shows 75 % sensitivity for 100 ppm methane gas at 350 0C operating temperature (Yadav et al., 2019). Liu et al., studies shows SnO_2-WO_3 Composite Thick Film shows 90% Sensitivity towards 30 ppm of methane gas at 200 0C operating temperature film prepared using screen printing method. Jin et al., Zhang et al., Composite of SnO_2-Au and SnO_2-Graphene Thin Film shows 92 % sensitivity for 3 ppm and 95 % sensitivity for 30 ppm towards H2S gas at 300 0C operating temperature.

4.2 ZnO-Based Thin and Thick Films for Gas Sensing

Zinc oxide (ZnO) is n-type semiconductor having bandgap of 3.37 eV to 3.44 eV. For the identification of gases like hydrogen sulfide (H_2S) and methane (CH_4) Zinc oxide used (Kumar et al., 2020) [39]. The change in electrical properties developed in past two decade ZnO to interact with these gases (Sharma & Gupta, 2019) [40]. The films of ZnO has been effective and offering high surface area and smaller sensor devices (Mishra et al., 2018) [41]. Zinc oxide has been recognized towards both reducing and oxidizing gases shows good sensitivity for gases (Liu et al., 2008) [42]. Performance of ZnO's has been significantly improved by employing nanostructures such as nanorods (Kang et al., 2012) [43]. Thin film such enhances structural properties nanostructures to improve sensor response (Wang et al., 2010) [44]. Additionally, doping of ZnO with elements like Ga, In, and Al led to further developments in its gas-sensing properties (Gao et al., 2009) [45].

4.2.1 Pure ZnO Thin and Thick Films

Detecting Reducing gases such as H_2S and CH_4 Pure ZnO thin and thick films have been extensively used due to its excellent response (Jain et al., 2021) [46]. ZnO surface interact with the adsorbed oxygen species, leading to changes in the electrical resistance of the film (Chen et al., 2019) [47]. Different techniques such as Screen printing, sol-gel deposition and chemical vapor deposition (CVD), used for fabrication of ZnO thin and thick films. (Patel et al., 2020) [48].

4.2.2 Composite ZnO Thick and Thin Films

To enhance performance of the ZnO sensors, composite materials have been developed by combining ZnO with other materials such Au, Ag and metal oxides (e.g., SnO_2, TiO_2), or carbon-based materials (Rajput et al., 2020) [49]. These composites of ZnO have been shown to improve parameters selectivity, sensitivity, and response time often at lower operating temperatures of ZnO-based sensors, (Das et al., 2021) [50]. The modification in the electronic structure of ZnO, leading to more efficient gas detection (Kumar & Kumar, 2019) [51].

4.2.3 Comparative Analysis

The table below summarizes the performance metrics of pure and composite ZnO-based thin and thick film sensors for H_2S and methane detection over the last two decades, highlighting the sensitivity, operating temperature, materials used, year of study, experimental technology, and response times.

Table 2: Sensitivity, Response time of Pure and Composite ZnO Films

Sr. No	**Year**	**Material**	**Experimental Technology**	**Gas Detected**	**Sensitivity**	**Operating Temperature (°C)**	**Response Time (s)**	**Reference**
1	2018	Pure ZnO Thin Film	Sol-Gel	H_2S	78% (10 ppm)	300	20	Chen et al., 2019 [47]
	2019	Pure ZnO	Screen	CH_4	72% (100	350	25	Gupta et

Sr. No	**Year**	**Material**	**Experimental Technology**	**Gas Detected**	**Sensitivity**	**Operating Temperature (°C)**	**Response Time (s)**	**Reference**
1	2018	Pure ZnO Thin Film	Sol-Gel	H_2S	78% (10 ppm)	300	20	Chen et al., 2019 [47]
2		Thick Film	Printing		ppm)			al., 2019 [40]
3	2020	ZnO-Au Composite Thin Film	CVD	H_2S	90% (5 ppm)	200	15	Patel et al., 2020 [48]
4	2019	ZnO-SnO_2 Composite Thick Film	Sputtering	CH_4	85% (50 ppm)	250	22	Rajput et al., 2020 [49]
5	2021	ZnO-Graphene Composite Thin Film	Sputtering	H_2S	95% (3 ppm)	150	10	Das et al., 2021 [50]
6	2020	ZnO-TiO_2 Composite Thick Film	Screen Printing	CH_4	88% (30 ppm)	200	18	Singh et al., 2018 [24]

In above table Pure ZnO thin film shows 78% sensitivity for H2S gas at operating temperature 300 0C film prepared using Sol-Gel techniques (Chen et al.,2019). Pure ZnO thick film shows 72% sensitivity towards CH4 gas at operating temperature 350 0C film prepared using Screen Printing techniques (Gupta et al., 2019). Composite film of ZnO-Au shows 90% sensitivity at 5 ppm H2S gas at operating temperature 200 0C thin film prepared using CVD method (Patel et al., 2020). Composite film of ZnO doped SnO2 shows 85 % sensitivity for 50 ppm methane gas at operating temperature 250 0C thick film prepared using sputtering techniques (Rajput et al., 2020). ZnO-TiO2 composite film shows 88 % sensitivity towards 30 ppm methane gas at operating temperature 200 0C thick film prepared by Screen Printing techniques (Singh et al., 2018).

4.3 Pure and Composite SMO Thin and Thick Films for Gas Sensing

4.3.1 Pure SMO Thin and Thick Films

Sharma & Gupta et. al., studies use of pure SMOs, such as ZnO, SnO_2, TiO_2, and WO_3 in gas sensors (Gupta & Sharma, 2019) [52]. Sputtering, sol-gel and chemical vapor deposition techniques applied for the preparation of thin films (Patel et al., 2020) [53]. Screen printing or drop-casting methods used for Thick films preparation which creates higher surface area to enhancing gas sensitivity (Das et al., 2021) [50].

4.3.2 Composite SMO Thin and Thick Films

Composite SMO films such as Au, Ag and other metal oxides (e.g., SnO_2-ZnO, TiO_2-WO_3), to improve gas sensor performance (Sharma & Gupta, 2019). These composites are designed to enhance properties such as surface area and electrical properties to enhance sensitivity and selectivity at lower operating temperatures (Kumar et al., 2020) [54]. The integration of these materials into thin and thick films has shown significant improvements in detecting H_2S and CH_4 gases (Jain et al., 2021) [55].

4.3.4 Comparative Analysis

The table below summarizes the performance metrics of pure and composite SMO-based thin and thick film sensors for H_2S and methane detection over the last two decades.

Table 3: Pure and Composite SMO Films

Sr. No	**Year**	**Material**	**Experimental Technology**	**Gas Detected**	**Sensitivity**	**Operating Temperature (°C)**	**Response Time (s)**	**Reference**
1	2018	Pure ZnO Thin Film	Sol-Gel	H_2S	78% (10 ppm)	300	20	Chen et al., 2019 [47]
2	2019	Pure ZnO Thick Film	Screen Printing	CH_4	72% (100 ppm)	350	25	Gupta et al., 2019 [40]
3	2020	ZnO-Au Composite Thin Film	CVD	H_2S	90% (5 ppm)	200	15	Patel et al., 2020 [48]
4	2019	ZnO-SnO_2 Composite Thick Film	Sputtering	CH_4	85% (50 ppm)	250	22	Rajput et al., 2020 [49]
5	2021	ZnO-Graphene Composite Thin Film	Sputtering	H_2S	95% (3 ppm)	150	10	Das et al., 2021 [50]
6	2020	ZnO-TiO_2 Composite Thick Film	Screen Printing	CH_4	88% (30 ppm)	200	18	Singh et al., 2018 [24]

Above tables results Pure ZnO Thin Film prepared using Sol-Gel technique shows 78 % sensitivity for 10 ppm H_2S gas at 300 0C operating temperature (Chen et al., 2019). ZnO-Au and TiO_2-WO_3 Composites shows 72 % sensitivity for 100 ppm at 350 0C operating temperature, 95 % sensitivity for 3 ppm at 150 0C operating temperature towards H_2S gas (Patel et al., 2020, Das et al., 2021). SnO_2-ZnO,

SnO_2-TiO_2 Composite Thick Film shows 85 % for 5 ppm, 88% for 30 ppm sensitivity towards methane gas at 250 0C, 200 0C operating temperature (Rajput et al., 2020, Singh et al., 2018).

5. Applications

1. **Environmental Monitoring:**
   Su et al., studies WO3 materials films effective for detect low concentrations of NO2. Thick films gas sensor used to detect pollutants like CO2, NO2, and SO2. (Su et al., 2003). Fine et al., Studies have shown that WO3 and SnO2 are particularly effective for detecting pollutants.
2. **Industrial Safety:**
   For Industrial safety detection of combustible gases such as hydrogen sulfide (H2S), LPG is critical. Composite of metal oxides thick film sensors shown high sensitivity to these gases (Chaudhari et al., 2006).
3. **Automotive Applications:**
   Thick film gas sensors are used in vehicles to monitor air quality and exhaust gases. For instance, sensors in air cleaning systems detect and control the levels of pollutants within the vehicle cabin (Kim et al., 2006).

6. Challenges and Future Directions

Despite significant advancements, several challenges remain:

1. **Optimization of Doping Techniques**
   Doping has been widely used to improve the selectivity and sensitivity of SMO films doping used mostly. For example, SnO_2-ZnO Composite films have shown improved gas sensing properties Rajput et al., 2020 [49]. Due to the lack of comprehensive studies on the long-term stability and reproducibility of doped SMO films. More research is needed to understand the effects of different dopants on the microstructural changes and their impact on gas sensing performance over time.

2. **Sensitivity:** Both Zinc oxide and SnO2 has shown promising results in gas detection due to its sensitivity to both reducing and oxidizing gases still need to achieve higher sensitivity by making different composite films.
3. **Selectivity Enhancement:** Various approaches, including the use of noble metal catalysts and heterojunctions, have been investigated to enhance selectivity (Patial&Deshwal, 2021), (Colmenares et al., 2020). A significant gap exists in developing universally applicable methods for improving selectivity across various gases and operational environments. Research should focus on exploring new materials and surface modifications that can provide broad-spectrum selectivity improvements.
4. **Stability and Durability:** More work is needed to ensure that SMO sensors maintain consistent performance in real-world environments. This includes understanding the effects of long-term exhibition to different gases and the potential degradation mechanisms that may affect sensor longevity.

5. **Low Power Consumption:** Reducing the power consumption of sensors is essential for portable and wearable applications. Research into low-power materials and efficient sensor designs is ongoing (Williams & Rahman, 2014).
6. **Response Time:**ZnO sensors typically have faster response times compared to SnO_2, often ranging from 10 to 30 seconds (Wang et al., 2010). Work is needed to improve the gas sensors response time by trying novel materials and their composites.
7. **Operating Temperature:** Achieving room temperature operation for SMO gas sensors has been a major focus, with some success using techniques like UV activation and advanced materials like reduced graphene oxide (Wang et al., 2012), (Filipovic&Selberherr, 2022). There is still a need for robust, scalable solutions that enable room temperature gas sensing without compromising sensitivity and selectivity. Integrating these solutions with existing CMOS technology for mass production remains a challenge.

7. Conclusion

From the past two decades, significant development has been made in the improvement of SnO_2-based thick and thin film sensors for detecting H_2S and methane gases. Pure SnO_2 thin films shows 80% sensitivity towards H2S gas at 300 0C operating temperature film prepared using sol-gel techniques (Gupta & Sharma, 2017). but the introduction of composite materials has led to substantial improvements in sensitivity, selectivity, and lower operating temperatures. SnO_2-WO_3 Composite has 90% sensitivity towards methane gas at 200 0C operating temperature thick film fabricated using Screen printing method (Liu et al., 2018). Thick film fabricated using Screen printing method of pure ZnO thick films have 72% sensitivity towards methane gas at 350 0C operating temperature (Gupta et al., 2019). ZnO-Au Composite Thin Film prepared using CVD shows 90% sensitivity towards H_2S gas at operating temperature 200 0C (Patel et al., 2020). The development of pure and composite semiconductor metal oxides thin and thick films has significantly advanced gas sensing technology for H_2S and methane detection over the last two decades. Pure SMOs have demonstrated effective gas sensing capabilities, while composite materials have led to substantial improvements in sensitivity, selectivity, and response times. Future research should continue to explore novel composite materials and advanced fabrication techniques to additional improvement and performance of these sensors for industrial and environmental applications.

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