Nano metal oxides for sensing the VOC based gaseous pollutants

Farida A. Ali 1*, Rasmita Nayak 2*, Binita Nanda 2*

1Department of Electronics and Instrumentation Engineering, 2 Department of Chemistry, Institute of Technical Education and Research, Siksha ‘O’ Anusandhan (Deemed to be University), Bhubaneswar, Odisha, India

*corresponding author e-mail address: binitananda@soa.ac.in | Scopus ID 53980125400

ABSTRACT

The detection and monitoring of harmful toxic volatile organic compound (VOC) gases liberated from various domestic activities is necessary for environmental concern. In this regard, VOC sensors have taken greater demand in domestic environmental monitoring because of their low operating cost, cheap, reliable, low power consumption, improved energy efficiency and reduced emission. In this regard, metal oxide materials in their nano form show growing strength in many research laboratories and public health and so on with their effective chemical, physical, and electronic properties. Therefore, an attempt has been made to study the nano metal oxides for different toxic gases basing on overall research trend like, a good correlation between sensing materials and sensing properties to improve the sensitivity and selectivity of VOC sensors. This paper tries to accumulate the findings of different metal oxide semiconductor nanomaterials used in various types of VOC sensors.

Keywords: Nano materials; metal oxide semiconductors; gaseous pollutants; sensor.

1. INTRODUCTION

The word “sensor” finds its origin from the Latin word “sentire” which basically means ‘to identify’ [15]. A sensor is a device that is capable of identifying a physical quantity and then converting it into a signal type that can be read by an observer or by an instrument. For accuracy purpose sensors are calibrated against known standards. Nowadays sensors are used everywhere like inside cars, machines, aeroplanes, homes, offices, robotics etc. A sensor is a device, which generates a functionally related output usually in the form of an electrical or optical signal corresponding to an input quantity [16, 17].

The sensitivity indicates how much the sensor's output changes when there is a small change measured in input quantity. Sensors that are capable of measuring very small changes have very high sensitivities. Demand for the variety of sensors and the capabilities of technological advancements allowing more and more sensors to be manufactured in different areas. Among all, gas sensing technology has received increasing attention and has become more significant because of its widespread and common applications on the detection and monitoring of flammable and toxic VOCs in domestic as well as in commercial and industrial environment. Thus, the detection of VOC gas molecules with conversion of their concentration information into electrical signals needs the use of sensing technologies in different fields. Among the different active sensing materials, metal oxides have shown effective results towards VOC sensing application. A lot of research studies have been done on VOC sensing applications with the help of metal oxides.

Therefore, in this paper effort has been made to gather findings of different researchers about different metal oxide semiconductor nano-materials used to detect few popular VOCs that are toxic in nature and the most popular pollutants available in and around human surroundings.
2. ROLE OF NANO METAL OXIDES IN VOC SENSING.

Nano metal oxides play a significant role in the gas sensing properties. Mostly the transition metals and their oxides from periodic table III-VI with electronic configuration (d0-d10) mainly exhibit the sensing property. The synthetic procedure used in the material science shows a remarkable change in the properties of nanomaterials and shows a better performance in different applications towards gas sensing. The properties are i) high surface-to-volume ratio facilitates more gas to adsorb onto the surface of nanomaterial for better sensing, ii) introducing defects in the nanomaterial, functionalization are more beneficial in the absorption of gas molecule, iii) proper charge transfer property because of semiconducting nature, iv) catalytic combustion or resistance modulation make nanomaterial suitable candidate for gas sensing.

![Scheme 1. Schematic diagram VOC sensing mechanism.](image)

3. DIFFERENT POLLUTANTS AND THEIR DETECTION

3.1. Formaldehyde (CH₃O).

Presence of CH₃O in indoor air within the range (0.2-50 ppm) may cause a serious health problem. The maximum exposure level of CH₃O is 80 ppb per 30 minute as the direction of world health organization (WHO). So, the early detection of CH₃O is highly necessary. In this regard, the development of nano based material plays a key role in the selective detection of CH₃O. Till date a handful of nano based metal oxides are used in the detection of CH₃O. The mechanism of CH₃O mainly depends on chemisorbed oxygen on the surface of the catalyst. The oxygen molecule present in air, first chemisorbed on the catalyst. Again this oxygen species (O₂⁻) react with CH₃O and simultaneously releases of electrons to the catalyst. This leads to a decrease in electron depletion layer (ELD) thickness and thereby increasing the sensing efficiency. The mechanism was shown as follows:

O₂ + 2e⁻ → 2O⁻
CH₃O + 2O⁻(ads) → CO₂ + H₂O + 2e⁻
CH₃O + O₂⁻(ads) → CH₂O₂ + e⁻

Zhang *et al.* synthesized NiO doped SnO₂ nanosphere and utilized the material for better sensing for CH₃O at different temperature range 50°C to 400°C than neat SnO₂ and NiO. The selectivity of the material NiO doped SnO₂ was again compared with ethanol, acetone, methylbenzene, chloroform and 2-chlorophenol. It was seen that 50 ppm CH₃O showed enhanced sensing (26.03) at optimum temperature (100°C). Chemisorbed oxygen plays an important role in sensing work.

In case of SnO₂ nanoparticle, only half-backed monolayer of oxygen was chemisorbed whereas in case of NiO-SnO₂, doping of NiO on the surface of SnO₂ easily aggregate and chemisorbed the full monolayer oxygen ions and oxidised to higher oxidation state [18]. Kim and his group prepared multi-heterogeneous oxides NiO/Fe₂O₃ loaded SnO₂ for formaldehyde detection.

Catalytic hetero-junction effect plays a crucial role in high gas sensing. Apart from hetero-junction property, loading of catalyst and operating temperature also plays an important role in high sensing of formaldehyde. Herein, the establishment of multiple hetero-junction between NiO, SnO₂ and Fe₂O₃ (p-n junction between NiO and SnO₂ and n-n hetero-junction between SnO₂ and Fe₂O₃). The migration of e’s from NiO to SnO₂ and again to Fe₂O₃ decreased the EDL and increases the metal oxide sensing of NiO/Fe₂O₃-SnO₂ composite [19].

Zhou and his team fabricated 3D hierarchical Co₃O₄ nanostructure by calcining Co₅-based MOF microcrystals. As prepared Co₃O₄-350 shows the best sensing towards formaldehyde at low temperature (170°C), the low detection limit of 10 ppm and 30 day stability.

Here, 3D nanostructure, high surface area, more active active sites, high porosity, shorter ionic diffusion lengths made the material more effective for formaldehyde sensing [20].

3.2. Methane (CH₄).

Methane is one of the most significant constituents of natural gas. It shows hazardous because it considered a
greenhouse and also flammable. Therefore it is necessary to detect CH₄ at lower concentration for warning purposes when it leaks and also to prevent hazards. Nowadays a handful of works have been done by the scientists and researchers. Nasrfsahani et al. was synthesized Pd-doped SnO₂/rGO and Pd-doped SnO₂/rGO and compared the methane sensing. The sensing performance was evaluated from the concentration of methane gas which ranges between 800-16000 ppm at room temperature. In this regard Pd doped SnO₂/rGO shows the better result (9.8 % sensitivity) among all synthesized materials. This is because rGO nanosheets provide a favourable surface for nucleation and growth of nanoparticles and a conducting network that facilitate transfer of electron. High catalytic activities of Pd also play as an additive role which improves the methane sensitivity at a very low working temperature [21].

Microwave assisted spinel Co₃O₄ nanoparticles were fabricated by Shaalan and his team for methane sensing. Co₃O₄ is a p-type semiconductor. As the holes are the major charge carriers in the p-type semiconductor, it dominates the passage of current through the boundary of a semiconductor. Thus, there is a decrease in resistance occurs due to the formation of oxygen ions on the surface of the material by the trapping electrons. Again, during the mixing of CH₄ and O₂, the electrons come back to the material and increase the resistance because of electron-hole recombinaton [22].

3.3. Ethanol.

Ethanol is a reducing gas that used as the raw material for colors, antifreeze and formaldehyde. Direct methanol fuel cells (DMFC) are eco-friendly and considered as the important alternative fuel for automotive manufactures. However, 10 mL intake of methanol may cause total blindness and more than 30 mL, which may cause fatal diseases. So, the early detection of low concentration of methanol at low operating temperature is highly necessary.

Yang et al. adopted the hydrothermal method to synthesize SnO₂/Zn₂SnO₄ porous nanosphere for excellent ethanol gas sensing. The two main responsible factor for ethanol sensing are: i) porous structure and ii) the hetero-junction attained by SnO₂ and Zn₂SnO₄. High surface area provides a large active sites which are essential for gas adsorption and diffusion. More is the surface active sites more is the adsorption between the adsorbed oxygen and methanol and more is the sensing. The hetero-junction play a significant role in sensing performance because of different band gap energy and work function of SnO₂ and Zn₂SnO₄. Herein the conduction band edge potential of Zn₂SnO₄ is higher than the conduction band edge potential of SnO₂ [23].

Cheng et al. were synthesized pure and Pd-doped α-Fe₂O₃ porous nanotubes (PNTs) by a facile electrospinning approach. The results reveal that, compared with pure α-Fe₂O₃, the 3.0 wt% Pd-doped α-Fe₂O₃ PNTs sensors exhibit superior performance in ethanol detection (at 240 °C). The highest sensitivity reaches 65.4 for 50 ppm ethanol and the detection limit is as low as 0.1 ppm. It is excellent selectivity, and good sensor towards ethanol [24].

3.4. Methanol.

Methanol is an essential substance in daily life and in the industry. It is also considered as a raw material for colors, antifreeze and formaldehyde. Though it has advantages, the dietary intake of 10 mL may result in total blindness and exceeds that quantity may cause fatal diseases. So, this is necessary to quick detect methanol gas of low concentration at low operating temperature. Li and his group fabricated Sm₂O₃ doped ZnO/SmFeO₃ microsphere for the high performance in methanol sensing.

The composite shows excellent behaviour toward very low concentration of methanol (5 ppm) sensing at very low operating temperature (195°C) with 24 sec recovery time. Herein two main factors are responsible for methanol sensing are high surface area and p-n junction between ZnO (p-type) and SmFeO₃ (n-type).

High surface area of SmFeO₃ (24 h) enhances the active sites which adsorb oxygen. This leads to releasing back of electrons to the semiconductor surface and decrease the conductivity. The reaction as follows:

\[
\text{CH}_3\text{OH} + 3\text{O}^\cdot \rightarrow \text{CO}_2 + \text{H}_2\text{O} + 3\text{e}^\cdot
\]

Secondly, the formation of p-n heterojunction between ZnO and SmFeO₃, the transfer of electron and hole takes place through the interface as long they balance the Fermi level. Methanol reacts with the adsorbed oxygen on the surface of ZnO, releasing electron back. CH₃OH + O” → HCHO + H₂O + e”

HCHO + O” → CO₂ + H₂O + e”

Again, on the surface of SmFeO₃, the hole react with the adsorbed oxygen

\[
\text{CH}_3\text{OH} + h^\cdot + O^\cdot \rightarrow \text{HCHO} + \text{H}_2\text{O}
\]

HCHO + h^\cdot + O^\cdot → CO₂ + H₂O + e”

That means the two different type of metal oxide can easily reactive with oxidative and reductive gases leading to high methanol sensing [25].

3.5. Acetone.

Acetone is good for some synthetic fibres and it is water miscible, can be used in common cleaning purpose. Acetone poisoning occurs when there's more acetone in your body than your liver can break down. When exposed to the air, it quickly evaporates and remains highly flammable. Acetone is dangerous to use around an open flame.

He prolonged exposure to acetone i.e. more than 173 ppm is highly toxic and cause serious threat to human health. As per the literature survey, the excelled gas of a healthy man is less than 0.8 ppm and in case of diabetic person is more than 1.8 ppm. So, sensing acetone is the best way to diagnose as compared to other testing. Thus it is highly necessary to prepare a suitable material that can easily sense acetone at a lower concentration. Wu et al. synthesized ZnFe₂O₄/rGO for acetone sensing (10 ppm) at the operating temperature (150°C-225°C).

When adsorbed oxygen on the surface of ZnFe₂O₄ surface capture electrons in the CB to form O₂- and O-. This results in the increase in resistance because of electron depletion layer. When acetone is exposed to the surface of ZnFe₂O₄, O₂- and O” react acetone at the grain boundary of ZnFe₂O₄. At this time again electrons are released back to CB.

\[
\text{O}_2(\text{gas}) \rightarrow \text{O}_2(\text{ads})
\]

\[
\text{ZnFe}_2\text{O}_4(\text{rGO} + \text{O}_2(\text{ads}) + \text{e}^- \rightarrow \text{O}_2^\cdot
\]

\[
\text{ZnFe}_2\text{O}_4(\text{rGO} + \text{O}_2^\cdot + \text{e}^- \rightarrow 2\text{O}^\cdot
\]

\[
\text{CH}_3\text{COCH}_3 + 8\text{O}^\cdot \rightarrow \text{CO}_2 + \text{H}_2\text{O} + 8\text{e}^-
\]

Again, the enhancement of sensing of acetone occurred due to 2D-rGO nanosheet by the improvement of electron mobility
inside the composite, forming a heterojunction structure in the composite. [26]

Xu and his team fabricated hollow Pt functionalized tungsten oxide (W_{18}O_{49}) for acetone sensing. The combined effect of porous shell and properties of Pt enhances the sensing performances 20 times more than pure W_{18}O_{49}. The enhanced sensing properties mainly due to two factors, one is spreading of Pt nanoparticle over the surface of W_{18}O_{49} and the second one is the lower work function of Pt (5.65 eV), the electron flows from W_{18}O_{49} to Pt nanoparticle till the Fermi level equalized, decreasing the electron depletion layer and simultaneously increasing the scottky barrier height at the interface of n-p junction (W_{18}O_{49} and PtO).

This results in increase the resistance and therby the sensing performance. Apart from these properties, the high surface are, hollow sphere provides a better active sites for gas diffuson and inhibits the excellent gas sensing [27].

3.6. Triethylamine (TEA).

TEA is a colourless volatile liquid widely used in chemical industries. It is also highly explosive. The maximum consumption limit of TEA is 10 ppm or less. Higher to this concentration level, it causes headache, lungs problem etc. Various traditional methods are used for TEA detection such as gel chromatography and gas tube detection. But these methods are tedious and also costly. To avoid these problems, sensing is the best way to detect very low concentration of TEA gas at the faster rate. In this regard, porous metal oxides are the best option to use as the sensing material. Highly effective SnO₂ hallow microfiber was prepared by Zhou and his co-workers for TEA sensing.

Sensing mechanism of hollow SnO₂ microfibres is mainly due to the presence of oxidative radicals (O₂⁺, O−, and O²⁻) and formation of electron depletion layer on the surface of the material. Other than that, unique structural characteristics such as the small grain boundary of SnO₂, large active sites, 1D carbon morphology and hollow porous structure contribute the outstanding response to TEA sensing having concentration 100 ppm at operating temperature 270°C [28]. Song et al. synthesized Au decorated porous α-Fe₂O₃ nanorod for very low ppm level detection of Triethyl amine sensor. As selectivity is the most important character for gas sensor, Au/α-Fe₂O₃ exhibit the highest response towards 50 ppm of TEA at 40°C as compared to neat α-Fe₂O₃. High response is mainly based on the bond energy. As bond energy of C-N (305 KJ/mol) is weakest among all other bonds, TEA is most unstable. For this reason the bond breaks easily lead to the high activity of TEA and response increases. When TEA is exposed on the surface of Au/α-Fe₂O₃, there is an oxidation and reduction reaction occur between TEA and O²⁻ asper the reaction:

\[ \text{N}(\text{C}_3\text{H}_7\text{H}) + \text{O}^{2-} (\text{O}^{2-}, \text{O}^{-}, \text{O}_2) \rightarrow \text{N}_2 + \text{H}_2\text{O} + \text{CO}_2 + e^- \]

In addition, the decoration of gold on the surface of α-Fe₂O₃ leads to extraction of more number of electrons from the conduction band of α-Fe₂O₃. High extent of chemisorbed oxygen adsorption capacity on the surface of Au/α-Fe₂O₃ results in the high response by accelerating the reaction between the oxygen species and the reducing gas. This leads to decrease in recovery time of Au/α-Fe₂O₃ sensors [29]. In addition, few other nano materials used in the sensing of various types of VOCs at different concentration and operating temperature are summerised in the Table 1.

![Table 1. Different other materials utilized in the sensing of VOCs.](image)

4. CONCLUSIONS

This paper addresses the detection and monitoring of harmful volatile organic compounds liberated mostly from household activities. Because of the adverse effects of these gaseous pollutants on human health and also on the surrounding environment, VOC sensors have been taken up for greater demand in environmental and also domestic monitoring. This paper is an
attempts to study few well known VOC sensors based on metal oxide semiconductor nano materials.

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