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A Review of Gas Sensors Based on Semiconducting Metal Oxide

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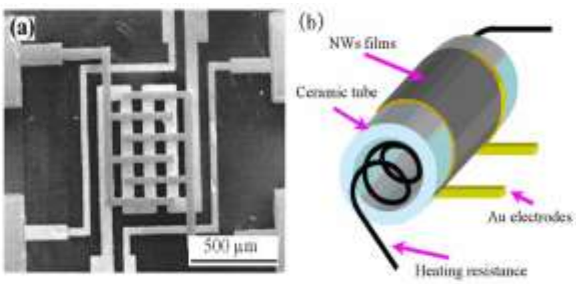
Abstract: *This study intends to provide a review of recent progress in gas sensors based on several theoretical and empirical investigations regarding semiconducting metal oxide nanostructures. Modified or doped oxide nano-wires, device structures such as electronic noses and low power consumption self-heated gas sensors, and their gas sensing performance has also been evaluate. Finally, the researcher also point out some challenges for prospect investigation and practical application.*
Keywords: *Gas sensors, semiconducting oxides.*

I. Introduction

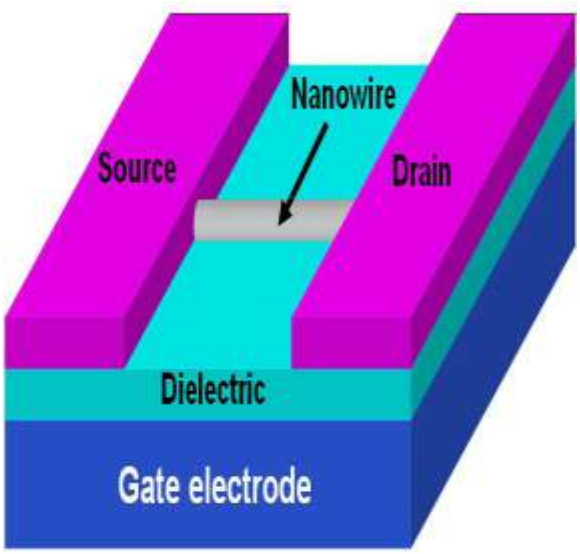
Yamazoe (1991) in his work revealed that the Semiconducting metal oxides have been decades to good gas sensing materials. Ethanol sensors based on SnO2 thick films have been commercialized for years. The researcher also demonstrated the reduction in crystal size would significantly increase the sensor performance [1]. This is because nano sized grains of metal oxides exhausted of carriers and exhibits much inferior conductivity than micro sized grain in ambient air. Hence, when exposed to target gases, they display greater conductance changes as more carriers from their attentive states to the conduction band than with micro sized grains. Thus, the technological challenge moved to the invention of materials with nano-crystals which maintained their constancy over long-term operation at high temperature [2].

According to Pan and Others (2001) oxide nanostructures has been inspired and facilitated by the convenience of obtaining large amounts of single crystalline nano wires the vapor transport [3] and vapor-liquid-solids (VLS) methods [4]. Sberveglieri [5] and Yang [6] groups initiated the investigation of gas sensing properties of SnO2 nano-belts. Sberveglieri demonstrated the use of SnO2 nano-wires as sensor materials showing prominent current changes towards ethanol and CO respectively, in a synthetic air environment while Yang *et al.* demonstrated the first photochemical NO2 nanosensors operating at room temperature. In 2004, our group reported high performance ZnO nano wire sensors with a low detection limit of 1 ppm ethanol at 300 °C [7]. The gas sensor configurations and measurements, performance parameters, as well as theoretical fundamentals of gas sensor, *Fabrication and Characterization of Gas Sensors* metal oxide nanostructures sensors have been characterized in three ways: conductometric , field effect transistor (FET) and impedometric ones. Conductometric sensors are based on resistance changes caused by publicity of the sensor surface to a mark species. So far, two types of conduct metric nanowire gas sensors have been mainly fabricated: one is the film type, in which a film collected of nanowires is contacted by pairs of metal electrodes on a substrate (Figure 1a) or a ceramic tube (Figure 1b); the other is the particular nanowire type in which a lone nanowire bridges two metal electrodes on a deeply doped silicon substrate covered with SiO2 acting as insulating layer between the nanowire/electrode combination and the conducting silicon (Figure 2). In fabrication of film type nanosensors, nanowires products are pulverized to a pulp state and either directly painted or screen-painted [16] onto the substrates or tubes. But other approaches are report. Sometimes nanowire growth is included into device fabrication [17-20]: SiO2/Si substrates with patterned metal coatings were used to catalyze the growth of the metal oxide was coating also acts as electrodes contacting the sensing material. This type of sensor has lower resistance compared to the previous one because the nanowire expansion course is integrated into device fabrication. Well-aligned nanowire arrays have been fabricated into nanosensors to explore benefits in order.

Figure 1(a) shows MEMS structures with inter-digitized electrode [7] and Figure 1(b) gives the Schematics of nanowire gas sensors on ceramic tube [26].



The following figure 2 provides the schematic of the single nanowire field effect transistor.



For instance, Figure 3 shows the sensor device structure that has been adopted in exploring gas sensing properties of ZnO nanorods [21], in which ZnO nanorod arrays are sandwiched between the silicon substrate supporting their growth and an indium thin film that forms ohmic contact with the nanorods and the copper electrode. Another interesting approach to accumulate involves combining two vertically aligned CuO nanowire in which two pieces of nanowire arrays were attached to the copper plate and the micromanipulator tip (copper wire) and the distance between the two arrays can be adjusted. The sensor built in this style was reported to be capable of detecting air-diluted H2S at the parts per billion level [22]. However, these aligned nanowire arrays are grown by “bottom up” methods, and the orderliness is not as good as those fabricated *via* “top to bottom” approaches. Francioso and Son *et al.* employed microelectronics processes such as photolithography and plasma etching (demonstrated in Figure 4) to fabricate TiO2 [23] and ZnO [24] parallel nanowire arrays, respectively, and investigated their gas sensor behaviors. Son *et al.* [25] developed an alternative technique to construct well-aligned nanowire arrays, which takes advantage of the two facts: 1) the step edges of terraces are energetically positive for the nucleation of adatoms; step edges of terraces. They prepared uniform terraces on (0001) sapphire substrates byannealing a miscut sapphire substrate and minimized the ZnO evidence rate by using low laser pulse repetition rate as well as a shadow mask which blocks straight ZnO plume generated by laser ablationpulsed laser deposition

Figure 3. The schematic of the ZnO nanorod array sensor.

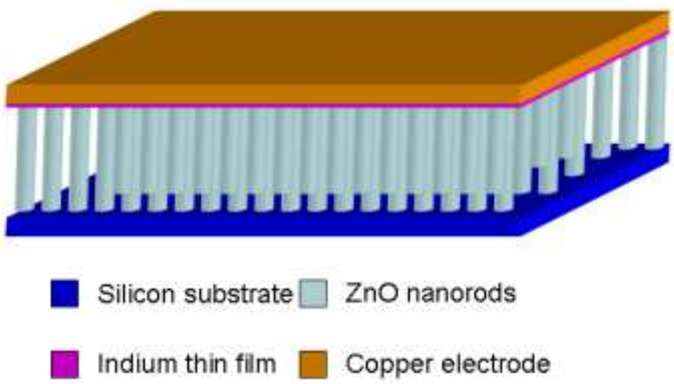
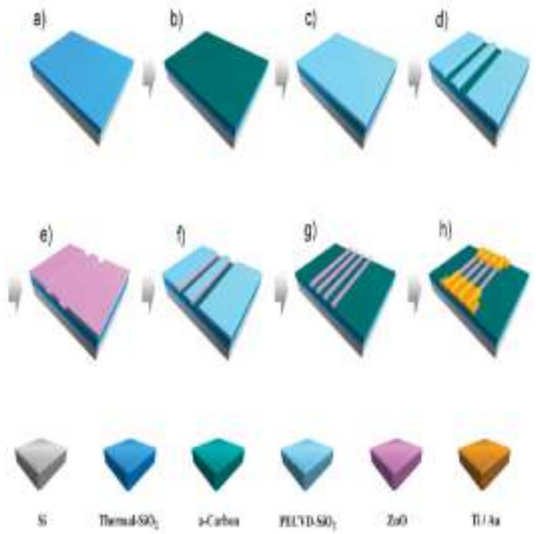


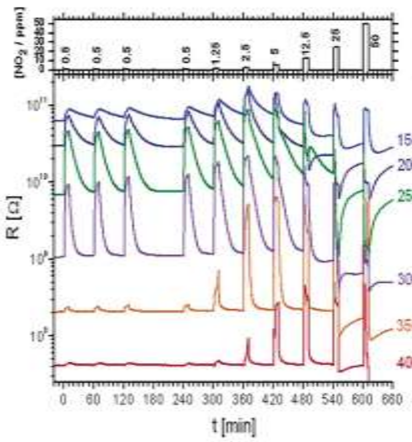
Figure 4 Schematic diagram illustrating the fabrication processes of the ZnO nanowire
device based on nanoscale spacer lithography (NSL): (a) thermal growth of SiO2 layer, (b) deposition of a-carbon thin film (act as etch-stop layer in subsequent process), (c) plasmon enhanced chemical vapor position (PECVD) of SiO2 thin film (sacrificial layer), (d) sacrificial layer patterning, (e) atomic layer eposition (ALD) of ZnO thin film, (f) top view of the chip after plasma etching of ZnO, (g) sacrificial layermoval, and (h) top view of the ZnO nanowire device after metal electrode deposition [24].



Basically, the working principle of a FET type sensor is that the species adsorbed onto the channel surface can work as an extra virtual gate bias and hence cause changes in the apparent thresholdvoltage. However, the FET configuration of a sensor does not guarantee it work in the simple ideal way. For example, Andrei *et al.* [27] discovered that their SnO2 nanobelt FET can be modeled as two Schottky diodes connected back-to-back with a series resistance from the nanobelt separating thediodes and only work as a FET in the presence of hydrogen. Another interesting phenomenon observed by Zhang *et al.* is that the gate effect typical of a FET was substantially weakened when their In2O3nanowire transistors were exposed to high concentrations of NH3 (10%) [28]. They proposed that these NH3 molecules residing on the nanowire surface can be charged and discharged by sweeping the gatebias and hence effectively work as charge traps screening the electric field induced by the gate bias. Impedometric sensors are based on impedance changes and are operated under alternating voltage upon exposure to target species. Like conductometric sensors, there are two types of impedometric ones: film type [29] and single nanowire type [30]. But this group of sensors has not attracted as much attention as the conductometric sensor yet. The rest of Section 2 will be mainly focused on conductometric sensors.

Surface Reactions for Gas Sensors:

Semiconducting oxides generally owe their conductivity to their departure from stoichiometry .Defects such as interstitial cation or anion vacancies also play an important role in their conductivity.Target species can be confidential into two groups: oxidizing gases or electron acceptors such as NO2, which produce a decrease in the conductance of n-type semiconducting materials (*i.e.*, electrons are the major carriers, such as ZnO, SnO2,In2O3) and an increase in the conductance of p-typesemiconducting materials reducing gases or electron donors such as H2S, CO, H2 and water vapor, which act in a reverse manner. Interestingly, sensors based on TiO2 nano fibers have recently been reported not to undergo conductance decrease towardsoxidizing gas NO2 as they normally do [31].Figure 5 highlights the resistance response during cyclic exposure to 10 min pulses with increasing concentrations of NO2 mixed in dry air at various operating temperatures [31].



As shown in Figure 5, at higher meditation ranges the device resistance plummet before NO_2 stimulus was detached. The authors attributed such an irregular response behavior to the conduction type inversion (n-to-p) of the sensing material whose transmission is surface-trap limited, owing to thehigh surface-to-volume ratio of this materialThere are two types of adsorption: physisorption, the first step of the relationship of the gas specieswith the sensor surface, and chemisorptions, which involves exchange of electrons between the adsorbed variety and the material surface. The major difference among these two processes is thatphysisorption is exothermic even as chemisorption is endothermic, precisely an activated process whose activation energy can be complete by thermal or non-balance ones such as illumination [2].

This leads to the fact that physisorption predominates in low temperature range whereas chemisorption dominates in advanced temperature variety. The sensing characteristics of metal oxides are widelyconsidered to be related with chemisorbed oxygen and water, which can act as intermediates catalyzing the charge transfer process between gas species and the mass and which complicate the study of gas sensing mechanisms. The major way they interfere with the gas sensing process is through fluctuations in the concentration and the charges of oxygen vacancies. Ahn *et al.* [32] investigate the effect of oxygen-situation-related defects on gas-sensing properties of their single ZnO nanowire gassensors and found that the gas sensitivity towards NO_2 was linearly proportional to the photoluminescence intensity of oxygen-vacancy related defect. Their work proves evidence of the role that oxygen vacancies play in gas sensing. Recent development in the synthesis of single crystalline nanowires or nanobelts has inspired research into their gas sensing properties, which divulge important information about the reactionsbetween target species and metal oxide surfaces free from complications caused by grain margins. For example, single crystalline SnO_2 nanobelts provided Moskovits *et al.* the opportunity to studysurface reaction kinetics between the individual nanobelt surface and CO and fit the investigational data to the analytical model they derived [33]. Another example is that single crystalline SnO_2 nanobeltswith well-defined facets and also give Yang *et al.* the model toverify their results from numerical investigation into surface connections between SnO_2 with NO_2species: through first principle bulk functional theory DFT calculations. They establish unexpectedly that most stable adsorbed species involve an unexpected NO_3 group doubly bonded to Sn centers,which was confirmed by their X-ray absorption spectroscopy studies on nanoribbons [34]. Nanowire/nanobelt diameter is usually on the order of several nanometers and is comparable to the Debye length and this often results in much larger sensitivity than their thin film or bulk counterparts.The size dependent characteristic has also been studied by some researchers. Liao *et al.* found that thin nanorods have a significantly better sensing performance than thick nanorods in the detection ofC_2H_5OH and H_2S (100 ppm) in air [21]. The gas performance of film type gas sensors can be limited not just by surface reaction processes, but also by the morphology and microstructure of the films.

Contact barriers among nanowires can also affect the gas sensing properties via affecting the resistanceof the bulk material [35]. Generally, researchers use the power law, $S = a + bc\rho$, to fit the concentration-sensitivity curves of film type nanosensors, and Langmuir adsorption isotherm to fit the sensitivity-concentration curves of single nanowire gas sensors [36].

Performance Parameters

Sensitivity, response and recovery time, linear range, as well as limit of detection (LOD) are important performance parameters for gas sensors. The sensitivity of conductometric sensors is defined as the ratio of the device's resistance when exposed to target species to that in ambient air, exactly R_g/R_a (where R represents resistance, the subscript 'g' represents target gas, and 'a' represents ambient air) if the target gas is an oxidizing one, or G_g/G_a (G represents conductance) if it is areductive one. Response (recovery) time is defined as the time period needed for the device to undergo resistance changing from 10% (90%) to 90% (10%) of the value in equilibrium upon exposure to an oxidizing (reducing) analyze. According to its definition, the estimation of LOD is done via extrapolating the R_g/R_a versus concentration curve to $3\sigma/R_a$ (σ is the standard deviation of R_a), but very few references mention to have done it in this way [31,37]. This is mainly because of the morphological complexity of the porous sensor surface and lack of efficient model to fit the sensitivity-concentration curves. There are few problems in this field of study which can severely hinder real applications of metal oxide 1D nanostructures. First, the researchers in this community do not abide by a unified LOD when claiming the detection limit of their gas sensor reaches some ppb or ppm level. Generally they justlabel the lowest concentration of the analyze used in their test as the detection limit of their gas sensors. The second is lack of uniformity in the working temperature selected. Almost half the publications report a working temperature setting of 300 °C, and one quarter at 400 °C. The optimumworking temperature is not always explored. Third, as water vapor produces resistance changes for metal oxides, it is important to present humidity information. Fourth, very few researchers (except [30,38-40]) have worked outside the linear range or selectivity to facilitate industrial applications. Up to now, various metal oxides 1D nanostructures (SnO_2 nano whiskers, In_2O_3 nanowires, ZnOnanorods, WO_3 nanowires, TeO_2 nanowires, CuO nanoribbons, CdO nanowires *etc.*) have been fabricated into film type nanosensors. As shown in Table 1, the most widely studied substances are

SnO2 and ZnO, probably due to the convenience of obtaining large quantities of SnO2[7] or ZnO nanowires [41] via thermal evaporation or a vapor-liquid-solid method. Actually the Table 1 serves as supplementary to another one in reference [42], which provides additional information on reported gas sensor properties. It's noteworthy that, in agreement with intuition, gas sensitivities of single nano wire gas sensors are invariably far less than those of nanowire film gas sensors, but the significance of single nanowire gas sensors is their potential application in microarray electronic noses [43].The metrics of humidity senor involves more complicated procedures than sensors for other targetspecies in order to obtain reliable data. In the latter, the ambient gas is usually switched between airand a target gas diluted in air, which simulates real applications; in the former, the target gas (water vapor) has to be injected into a highly dry environment which requires pretreatment such as evacuation to remove water adsorbent in the chamber [44]. When testing is conducted under high vacuum, the usual concept, the concentration, used to represent the quantity of gas species present is replaced by an alternative one, the gas pressure of target species.
Figure 6 views(a) SEM images of ZnO brushes [67]. (b) SnO2 brushes [28]. (c) ZnO dendrites [68]. (d) ZnO nanoflowers [69].

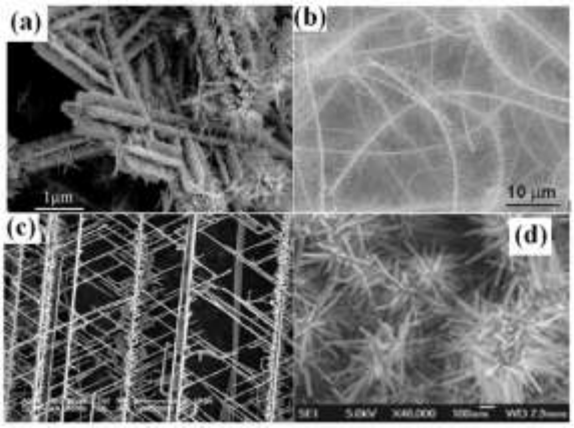


Table -1: Gas sensing properties of metal oxide nanostructure special morphology.

Material	Gas species	Sensitivity	Response	Reference
ZnO brushes	Ethanol	3 (5 ppm)	<10 s/<10 s (10 ppm)	[67]
SnO2 brushes	Ethanol	2.3 (0.5 ppm)	4 s	[26]
ZnO dendrites	H2S	3.3 (10 ppm)	15–20 s/30–50 s	[68]
ZnO Nanoflowers	Ethanol	4.1 (1 ppm)	1–2 s/1–2 s	[69]

II. Conclusion

In a nutshell, numerous publications have reported on the gas sensor behaviors of metal oxide. Since the lowest detection concentration is a very important performance index for sensors, it is necessary to make a compile on the level achieved towards common target species (CO, NO2, NH3, ethanol, H2, H2S) of sensors fabricated from different metal oxide. Table 3 brings the result of such effort. This study pertains to refer to those who are all wish to publish new data in this area.

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