International Journal of Engineering & Technology, 7 (3.34) (2018) 918-923

International Journal of Engineering & Technology

Website: www.sciencepubco.com/index.php/IJET

Research paper

A Survey on Capacitive based CO_2 Gas Sensor

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Abstract

In this current scenario, different sensing mechanisms are investigated and used in detection of hazardous gases. A review of capacitive type gas sensor for detection is presented in this paper. Even though there are very limited numbers of publications on capacitive based gas sensors, detection of CO_2 based on change in dielectric constant it shows good sensing mechanism with sensitivity and high reliability.

Keywords: *Gas Sensor, CO_2, Sensitivity, Selectivity, Metal Oxide*

1. Introduction

In this current world, the substantial increase in automobiles, industries, urbanization, modernization results in more air pollution. This in turn results in health hazardous, green house effect which ultimately is a major threat to mankind and environment. So in this regard there is a need in controlling the pollutant gases in the atmosphere. In order to control the increase of pollutant gases there is a requirement of sensors especially chemical sensors to detect and monitor these hazardous gases.
Detection of hazardous gases is more complex phenomena, because human nose is not able to recognize the dangerous gases like carbon monoxide, carbon di-oxide, hydrogen sulphide, chlorine, hydrogen chloride, hydrogen fluoride, nitric oxide, nitrogen dioxide, sulphur dioxide, ammonia, hydrogen cyanide etc. and are found in a wide variety of situations, varying from industry: chemical, petroleum, electronic, coal, gas mines, to warehouses, enclosed parking areas, vehicles, sewerage, waste disposal, the atmosphere, which affect human life and environment[1-3]. The risks because of these hazardous gases are, i) Risk of explosion by flammable gases. ii) Risk of asphyxiation (suffocation) by oxygen displacement. iii) Risk of poisoning by toxic gases. So it is essential to detect these dangerous gasses by means of appropriate measures called gas detectors or sensors.
By definition, Gas sensor is a transducer which detects the gas molecules by altering its physical properties like, resistance or capacitance and produces electrical signals which in proportional to the gas concentration. Recently, gas sensing technologies become more promising because of its wide verity of applications in environmental pollution monitoring, toxic and combustible gas monitoring, automobile combustion control, medical analysis and energy and drying operations markets. The emerging markets include defense, consumer products, automobiles, wastewater treatment, and food and beverage processing, etc [1].

1.1. Characteristics of the Gas Sensors

On the basis of properties, gas sensors can be used for different applications.

Sensitivity*:* It is the ratio of the change in sensor output in response to the change in sensor input in the presence of target gas. The sensitivity S (a) at point 'a' is sensor output y is related to the input x by the function y = f(x) [4].

$$S(Xa) = \Delta y \backslash \Delta x_{\ x=a} \qquad (1.1)$$

For a resistive gas sensor, the sensitivity S is defined as,

$$S(\%) = \frac{R-R_0}{R_0} \times 100 \qquad (1.2)$$

Where R_0 is the resistance which is in presence of reference gas, R is the resistance which is in presence of target gas, and S is the sensitivity in percentage with respect to resistance.
The sensitivity S of a capacitive sensor is defined as:

$$S(\%) = \frac{C-C_0}{C_0} \times 100 \qquad (1.3)$$

Where C_0 is the capacitance in the flow of reference gas, C is the capacitance in the presence of target gas, and S is the sensitivity in percentage with respect to capacitance.

Response Time: The time for the output of a gas sensor to reach a certain percentage of full scale after being exposed to target gas. For example, T_{90} = 60 seconds means that it takes the sensor 60 seconds to reach 90% of its full scale output after being exposed to a full scale gas concentration.

Recovery Time: The time for the output of a gas sensor to return to its normal states after the removal of the target gas. It is usually specified as time to fall to 10% of steady state value after the removal of the measured gas. An example of gas sensor response time and recovery time is as shown in figure 1.

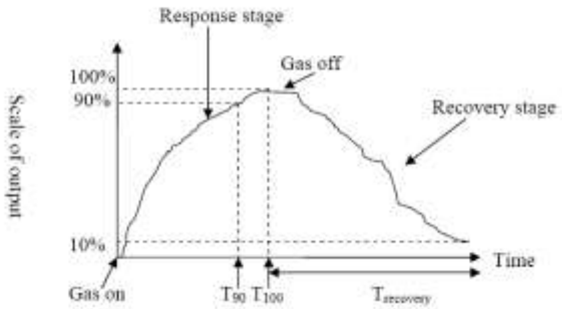

Figure 1: Response and Recovery time of Gas sensor.

Selectivity*:* It is the ability of a gas sensor to detect a target gas without being affected by the presence of other interference gases. Most gas sensors are sensitive to a family of gases and it is difficult to produce a sensor specific to only one gas. Moreover, temperature and humidity may also affect sensor performance.

Repeatability: It is defined as the ability of a sensor to repeat the measurements of gas concentrations when the same measured is applied to it consecutively under the same conditions.

Linearity: Usually the gas sensor shows linearity at initial stages and linearity of a gas sensor reaches saturation as the concentration of target gas increases. A general gas sensor output curve is shown in figure 2, which highlights the responses of sensor in three regions like linear region, nonlinear region and saturation region. On analysis of graph, gas sensor shows a better linearity at lower concentration compared with higher gas concentration.

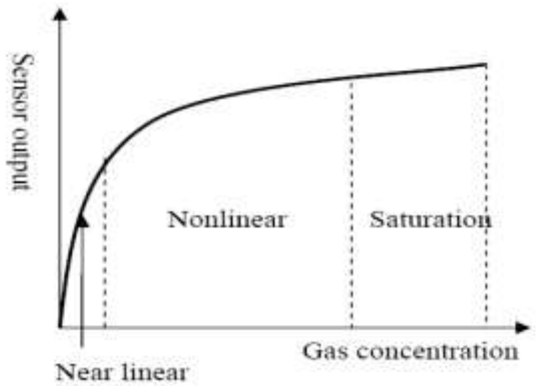

Figure 2: Typical Gas Sensor Output Curve (It Consists Of Near Linear, Nonlinear And Saturation Regions)

Various sensor technologies have been used for gas detection such as optical sensor, electrochemical sensor, catalytic sensors, infrared image sensors, ultrasonic sensors, Infrared point sensors, acoustic sensors. The operating principle and other features of sensor technologies are summarized as in table 1 [5].

Table 1: Sensing characteristics of various types of gas sensors

Characteristics	Semi conductor	Catalytic	Solid electrolyte	Electro chemical	Thermal	Infrared
Sensitivity	Excellent	Good	Excellent	Excellent	Bad	Excellent
Accuracy	Good	Excellent	good	Good	Good	Excellent
Selectivity	poor	Bad	Excellent	Good	Bad	Excellent
Response	Excellent	Good	Excellent	Poor	Good	Good
Stability	Excellent	Good	Good	Bad	Good	Good
Maintenance	Excellent	Good	Excellent	Bad	Good	Poor
Cost	Excellent	Excellent	Good	Good	Good	poor
Detectable concentration	Few ppm	10 ppm	10-10-1 atm	1-1000 ppm	1-100%	1ppm – 100%

On observing table 1, the disadvantages of above said technologies except metal oxide semiconductor are high cost, complex, less reliable, scalability issues so on. Hence a low cost, low power, more reliable, high sensitive and selective gas sensors are desirable.

In this context, many researches are working on metal oxide semiconductor based gas sensors which possess high sensitivity, good reliability, low cost, low power and fast response and recovery time. There are two types of sensing mechanisms to detect gases using metal oxide semiconductor gas sensors. 1. Resistive based gas sensor 2. Capacitive based gas sensor.

In resistive type gas sensor, the mechanism is based on the detection of gas with respect to change in resistance when sensor film is exposed to target gases and in another type gas sensor detection of gas with respect to change in capacitance of sensor film. Capacitance gas sensors measure variation capacitance due to either change of dielectric constant or a change in thickness of the sensor film. However a capacitive type metal oxide semiconductor gas sensor is advantages over resistive type.

1.2. Metal Oxide Semiconductor Sensors

Metal oxide gas sensors detect the target gases through Redox reaction between semiconductor layer and target gases. Steps involved in detection of target gas are, 1. Redox reactions- reaction of O^- which is present on the surface of the semiconductor layer with the target gas tends to change in physical properties of the oxide layer and then 2. This change is converted into an electrical resistance variation of the sensor. The change in resistance can be detected by measuring the any of the parameters like change of capacitance, work function, mass.

Electronic configurations, determines the selection of metal oxide layer for gas sensors, and are categorized as,

1. Transition metal oxides.
2. Non transition metal oxides – a) Pre transition metal oxides b) Post transition metal oxides [6].

Inert metal oxides are called as pre-transition-metal oxides (MgO, *etc.*). In such materials neither electrons nor holes can easily be formed because of their large energy band gaps. Due to their difficulties in electrical conductivity measurements non transition metals are rarely used as gas sensor materials [7].

Transition metals show good catalytic properties and have variable oxidation states which are useful for selecting different metal oxide for gas sensors. Transition metal oxides are used in technologically important catalytic processes like selective oxidation, selective reduction.

Transition metals are more sensitive than pre-transition-metal oxides to target gases. However, structure instability and non-optimality of other parameters limit their selection in gas sensor applications. Only transition-metal oxides with d^0 and d^{10} electronic configurations find their real gas sensor application. The d^0 configuration is found in binary transition-metal oxides such as TiO_2, V_2O_5, and WO_3. The configuration d^{10} is found in post-transition-metal oxides, such as ZnO, SnO_2. On the basis of change in resistance, metal oxides, such as Cr_2O_3, Mn_2O_3, Co_3O_4, NiO, CuO, SrO, In_2O_3, WO_3, TiO_2, V_2O_3, Fe_2O_3, GeO_2, Nb_2O_5, CeO_2 and TiO_2 [8], can be used to detect combustible, reducing, or oxidizing gases.

Sensing of target gas is strongly depends on the surface reactions. Different metal oxide materials show different reaction activation to the target gases. Oxygen which is adsorbed on the surface of metal oxide sensing material which opposes the flow of current in the sensor. In the presence of reducing gases the surface density of adsorbed oxygen decreases as it reacts with the reducing gases. Which allowing current to flow freely through the sensor. By changes in the resistance of sensing material, target gases can be detected.

Seiyama and Taguchi were developed the first metal oxide semiconductor sensor in 1960s [9][10]. It consists of a ceramic supporting tube with a platinum heater coil and a sintered semiconducting SnO_2 coating on the outside as shown in figure 3. To achieve the typical sensor working temperature range of 200-550°C the heater is used and the two gold electrodes are used to measure gas sensitive changes in electrical conductivity. The chemical reaction of target gas with the surface of SnO_2 particles conductivity of sensor changes [11] [12].

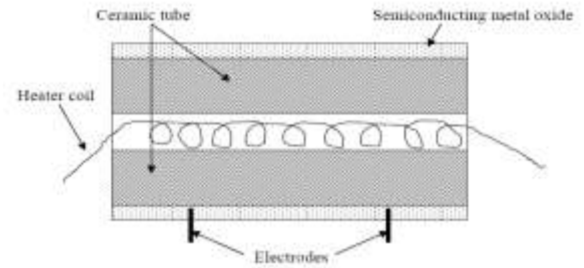

Figure 3: Schematic diagram Taguchi gas sensor

The mechanism of Taguchi sensor can be explained as, oxygen is adsorbed on its surface when semiconductor SnO_2 (n-type) is heated at a certain temperature. As the result, electrons which are majority carrier in the metal oxide semiconductor are transferred to the adsorbed oxygen molecules [13].

$$2\,e^- + O^2 \rightarrow 2O^- \quad (1.4)$$

In this reaction oxygen takes electrons out of semiconductor bulk, leave positive charges and form a space charge layer at the surface of SnO_2. At SnO_2 grain boundaries, a space charge layer (potential barrier) prevents charge carriers to flow inside the sensor, therefore surface conductivity decreases. Figure 4 describes the model of inter grain potential barrier and shows that the surface potential formed serves as a potential barrier against electron flow. Since the sensing material, SnO_2, is in the form of a compressed powder, i.e. sintered particles, the space charge induced by the oxygen adsorption can be a significant fraction of the whole grain; therefore the adsorption causes the resistance of the SnO_2 film to increase considerably. When reducing gas molecules react with the adsorbed oxygen species at high temperature they oxidized and acting as electron donor during the reduction-oxidation process.

$$R+O^- \rightarrow RO+e^- \quad (1.5)$$

As the result, the surface density of negatively charged oxygen decreases, so the height of potential barrier at

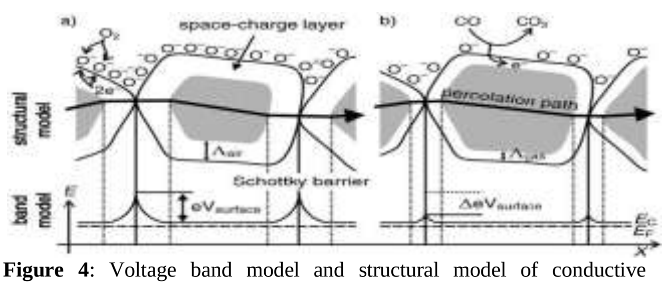

Figure 4: Voltage band model and structural model of conductive mechanism on exposure toreference gas. (a) Presence of CO (b) Absent of CO (adapted from [14, 15])

grain boundary is reduced along with the sensor resistance [13].

Metal oxide semiconductors shows high sensitivity to most combustible gases (NO, CO), fast response time, good reliability because of these advantages metal oxide materials are used in most of sensors. However, they are the relatively poor selectivity and high operation temperature. The selectivity can be altered by modifying the grain size or doping with catalytic additives, or by changing the operation temperature. The modifications have been made to improve the selectivity [16-20] shows inadequate results.

2. **Background and Related Work**

Capacitive gas sensors measure variations of the capacitance due to either a change of the dielectric constant or a change in thickness of the active layer induced by the gaseous species to be detected. As known that capacitance can be expressed as,

$$C= \varepsilon o\, \varepsilon_r A / d \quad (2.1)$$

Where εo = Permittivity of vacuum
ε_r = Relative permittivity
A= Area of an electrode
d= Distance between electrodes

Distance between electrodes can be view as thickness of dielectric layer. By change any of these parameters, there is a variation in capacitance value by which target gas concentration can be detected [5].

Changes in area of electrode are uncommon expect humidity gas sensor, because of most inorganic gases have same relative permittivity. Changes in dielectric constant due to interaction between target gas molecules and electrode is usually unexpected for a gas sensors. Therefore, the detection of target gases based on change in capacitance due to changes in distance between electrodes, in terns change in dielectric thickness becomes most promising.

3. Literature Survey

In this section survey of various detection mechanism based on capacitive type gas sensors are used to detect the target gases so for are presented. As known, the value of capacitance varies due to variation in any of parameters like area of an electrodes or distance between electrodes or relative permittivity. The following sections give brief details of various gas mechanisms.

3.1. Capacitive Gas Sensor Based on Changes in Relative Permittivity

Relative permittivity of a gas sensor changes when it is in contact with target gas. Humidity gas sensor is a very good capacitive type sensor because of high dielectric constant of water. It has a large dielectric constant of 78.5 at 298K. By adsorption of water with humidity sensor there is a change in relative permittivity, which provides a simplest detection mechanism.

Polymer and ceramics are the two types of sensitivity materials which are most used in capacitive type humidity sensor. Relative permittivity of polymer such as polyimides is of 3 to 6 where as water has 78.5 of relative permittivity. With the adsorption of water, capacitance of polyimide changes by which humidity can be measured.

An Alumina film is used as ceramic based capacitive type humidity sensors. Alumina film is fabricated by the process of vapor deposition or by reactive ion plating or by anodic oxidation [21-23]. For good sensing characteristics it requires a controlled micro structure of Alumina film [24-25]. Due to absorption of water the relative permittivity of porous Alumina film is increase

in turn increases in capacitance of the film.
In humidity sensors capacitive type sensors shows large sensitivity than other type sensors [26-28]. Capacitive type sensors it has small size, rapid response, low cost, simple structure [29-30]. Because of its simple structure, it can be easily fabricated on IC's.

3.1.1. CO & Hydrocarbon Sensor

An Aluminophosphate-5(AlPO) material with porous of uniform size (molecular sieves) is used as the dielectric sensitivity material for detection of carbon dioxide and carbon monoxide [31]. Heat stability is the attractive property of AlPO-n family [32]. Based on pore sizes and acidity enables the uniform adsorption property. Adsorption of CO or CO_2 molecules with AlPO-5, changes the dielectric property of molecular sieves.
Zeolite is used as sensitivity material for detection of hydrocarbon. When Zeolite is exposed to hydrocarbon there is increased in the value of capacitance. The response time depends on thickness of the Zeolite film and operating temperature. The bipolar interaction between molecular sieves and adsorbate plays an important role in capacitive type gas sensors even though the sensitivity of a sensor is low.

3.1.2. CO_2 Sensor

For detection of CO_2, 3-amino-propyl-trimethoxysilane and propyl-trimethoxysilane are used as sensitivity material. In this gas sensor, the sensitivity material is developed by organically modified silicates [33]. Even though the exact detection mechanism of CO_2 has not been discussed, under the exposure of CO_2 the dielectric property of sensitivity material changes due to adsorption with it and the capacitance of gas sensor decrease as shown in the figure 5.

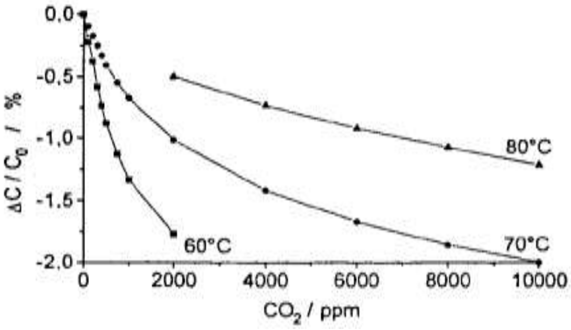

Figure 5: Capacitance variation of the sensor with increase in CO_2 concentration

Without any interference of humidity, this type of gas sensors can be operated at lower temperature becomes the attractive part of it.

3.2. Capacitive Gas Sensor Based on Changes in Thickness of Dielectric Layer

There are two types of detection mechanism on basis of changes in dielectric thickness or distance between electrodes. First approach is concentration of target gas molecules which interacts with the sensitivity material will change the dielectric thickness. On repeated exposure to the target gas molecules, sensitivity of a sensor gradually decreases or it disappears and also sensor shows more recovery time. The second approach of detection method uses formation of depletion layer due to interaction between p & n type semiconductors or metal-oxide-semiconductor or metal-semiconductor. In this type, thickness of dielectric changes, due to the electronic interaction between target gas molecules and semiconductors. This approach gives fast & reproducible response.

3.2.1. MOS Type Capacitive Sensor

Mechanism of metal oxide semiconductor type capacitive sensors is, on exposed MOS to target gas molecules the depletion layer of metal changes because of changes on work function. On observing the graph as shown in figure 6, characteristics curve shifts to the left on adsorption of target gases (eg. NH_3). This absorbed gas changes the work function of the metal gate electrode which changes the thickness of the depletion layer.

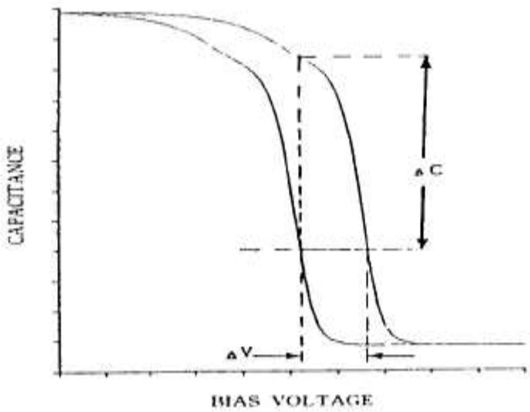

Figure 6: Capacitance-bias potential characteristics of MOS capacitor on exposure of NH_3

3.2.2. Capacitive Type CO_2 Gas Sensor based on PN Junction

In industrial and biological process, control and detection of CO_2 plays a vital role. Most CO_2 sensors which are explained so far are related on solid electrolyte gas concentration cell [34-38]. In view of reducing cost, miniaturization and simplification in design, various detection mechanisms are proposed [39-42].
While developing CO_2 sensors selectivity becomes an important target because reactivity of CO_2 is weak and produce small output signal. In this context, based on control of depletion layer near PN junction is formed between metal oxide semiconductors have been recently reported [43-45].
With the observation of graph, the exposure of CO_2 gas molecules in different concentration, the capacitance does not change when use of $BaTiO_3$ and PbO as alone. When equimolar mixture of $BaTiO_3$ is combined with metal PbO and used as dielectric material between two parallel Ag electrodes shows a significant change in capacitance value on exposure of 2% CO_2. As shown in figure 7, capacitance significantly decreases on exposing sensor to 2% CO_2 [46].

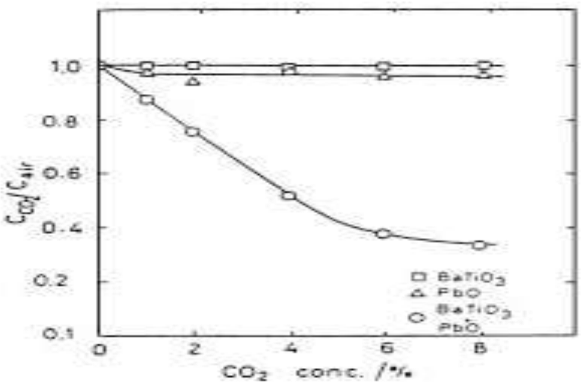

Figure 7: Sensitivity graph of different materials for different CO_2 concentration

Gas sensors sensitivity is strongly depends on what type of metal oxides are used and it can be improved by mixing other metal oxides with $BaTiO_3$ and is summarized as in table 2 [47]. In table operating temperature defines the detection CO_2 when it is mixed different oxides and sensitivity of oxides when it is exposed to 2% CO_2.

Table 2: CO_2 sensitivity characteristics of the mixed oxide capacitor

Sl. No.	Oxides	Operating Temperature/K	Sensitivity C_{co2}/C_{air}
	CaO	>1173	0.891
	MgO	1140	0.329
	La_2O_3	1039	0.451
	Nd_2O_3	823	0.641
	Y_2O_3	1032	0.794
	CeO_2	934	0.410
	PbO	774	0.711
	NiO	828	0.441
	CuO	729	2.892
	ZrO_2	915	0.740
	Co_3O_4	801	0.362
	Fe_2O_3	614	0.678
	Bi_2O_3	718	0.824

Some of oxides like CaO, MgO, La_2O_3 are sensitivity to CO_2 but generally requires high operating temperature. The stability of any sensor is related to operating temperature. CuO-$BaTiO_3$ equimolar mixture shows good sensitivity towards CO_2 among all oxides, on observation. As studied [48-49], the equimolar mixture CuO-$BaTiO_3$ gives the sensitivity of CO_2 and concentration is calculated as $(CuO)_x$ $(BaTiO_3)_{1-x}$. If concentration is below 0.3, capacitance of the device decreases and capacitance increases if concentration is above 0.3 on exposure to 2% CO_2. It is hard to find the capacitance if concentration is 0.7. The optimum equimolar concentration of CuO-$BaTiO_3$ is $(CuO)_{0.5}(BaTiO_3)_{0.5}$. The equimolar mixture of $(CuO)_x$ $(BaTiO_3)_{1-x}$ is exposed to different gases like H_2, CO, H_2O, CH_4, CO_2. On all, the mixture $(CuO)_x$ $(BaTiO_3)_{1-x}$ shows high selectivity for CO_2 detection as shown in the figure 7.

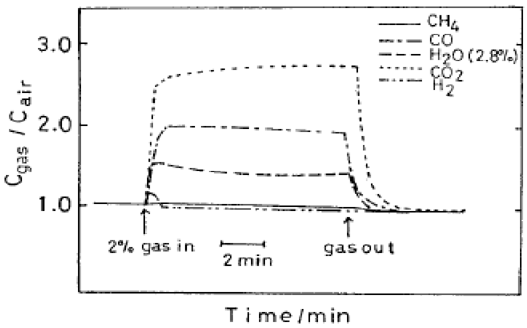

Figure 7: Comparison of sensitivity of equimolar mixture CuO-$BaTiO_3$ with other gases on exposure to 2% CO_2.

By comparing with other gas molecules, the response of Ag on selectivity was observed. $(CuO)_x$ $(BaTiO_3)_{1-x}$ mixture with Ag shows relatively high selectivity for CO_2 as shown in the figure 8.

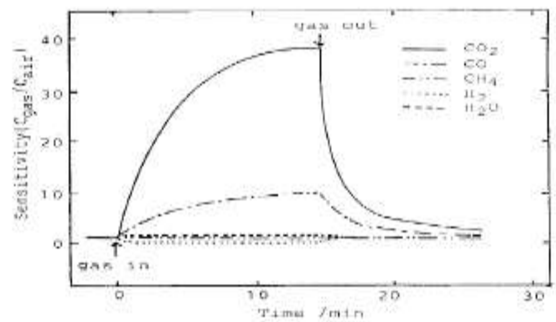

Figure 8: Sensitivity comparison graph for different materials with Ag after exposure to air containing 2% CO_2

On examination, $(CuO)_x$ $(BaTiO_3)_{1-x}$ added with Ag gives more attracting results in monitoring CO_2 gas present in atmosphere and also shows better stability and sensitivity. On mixing P type CuO and N type $BaTiO_3$ semiconductors there is a formation of potential barrier at the grain junctions between CuO and $BaTiO_3$. This mixture shows non linear I-V curves in air with potential barriers existing at the grain junction and I-V curves shows ohmic characteristics in air containing 2% of CO_2. Narrow depletion layer leads to increase in capacitance when height of the potential barrier decreased.

Number of sensing points, sensitivity and selectivity of a sensor can be increased by forming hetero junction of P & N type semiconductors. Therefore mixed oxides are useful and effective materials for the development of capacitive type gas sensors.

6. Conclusion

The main objective of the paper is to, provide a gas sensor which shows good selectivity and sensitivity. Since there is a need for low cost and more reliable sensors. By knowing about basics of capacitive type gas sensors which is based on change in capacitance, capacitive type gas sensors have good prospects given that the capacitor structure is so simple enabling miniaturization and achieving high reliability and low cost. A capacitive sensor enables sensitivity detection by amplification of capacitance done by simple oscillator structure. The oscillator circuit consist only standard resistor and sensor capacitor therefore circuit is simple and low cost. And key performance of capacitive gas sensor is its selective detection of target gas in a gas mixture.

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