INVESTIGATION OF ELECTRICAL AND GAS SENSING PROPERTIES OF SCREEN PRINTED GRAPHENE OXIDE THICK FILMS

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Abstract: Nowadays, gas sensors are useful in a number of fields including industrial, residential, agricultural, cold storage, and food storage. The present study describes the preparation of Graphene oxide (GO) thick films using a conventional screen printing technique and investigates their electrical and gas sensing properties. On a glass substrate, the films were prepared. After preparation, the films were annealed for one hour at 180°C in a muffle furnace. Investigation of the electrical parameters of prepared GO thick films were accomplished using resistivity, temperature coefficient ratio (TCR), and activation energy. Form electrical characterization prepared GO thick films shows semiconducting behaviour. Gas sensing studies were carried out in the presence of H₂S, NH₃, LPG, NO₂ and ethanol gases in the surroundings of GO thick films at various surrounding temperatures. Investigations were conducted into the properties of sensitivity, selectivity, PPM variation, response, and recovery time. The H₂S gas was found to have the highest sensitivity. The sensitivity to H₂S gas with a concentration of 150 PPM was 79.3% at 90 °C temperature. It was also found that the response and recovery times were about ~ 35 sec and ~ 60 sec seconds respectively.

Keywords: Graphene oxide, resistivity, glass substrate, hydrogen sulphide, sensitivity.

1. INTRODUCTION:

In recent years, metal oxide semiconductors have dominated surface-controlled resistance sensors, which constitute the majority of gas sensors. These sensors use sensitive materials from semiconductor resistance gas sensors [1]. To determine the composition and concentration of a gas, gas sensors use chemical and physical processes to turn the gas into an electrical signal output. The domains of flammable detection, explosive detection, the detection of poisonous and dangerous gases, and environmental control all make extensive use of gas sensors [1, 2]. Metal oxide materials have exceptional physical and chemical characteristics, are inexpensive to create, and may be prepared easily. As a result, MOS based gas sensors has used them more and more [2, 3].

Environmental monitoring is one application of gas sensors, which are devices that transform a gas volume fraction into electrical signals. A brand-new kind of two-dimensional crystal substance called graphene has a lot of great qualities, such a big specific surface area, a lot of conductivity, and a lot of Young's modulus. Due to these characteristics, it is perfectly suited for use with gas sensors [4, 5].

Graphene oxide (GO) is the derivative of graphene [5]. Chemically oxidising graphene or graphite yields GO, a derivative of graphene with functional groups including oxygen. GO is anticipated to be a crucial material in the actual application of graphene because it is easily synthesised through a facile solution process [6, 7].

The objectives of this study were to prepare GO thick films using screen printing methods and investigate the electrical and gas sensing properties of prepared GO films.

2. EXPERIMENTAL WORK:

2.1 Preparation of GO thick films by using screen printing technique:

Commercial available graphene oxide nano powder (99.9% purity) was utilized as the functional material for preparation of GO thick films in this study. The Figure 1 revels the flowchart of experimental work.



Figure 1: Flowchart of experimental work

2.3 Characterizations of GO thick films:

2.3.1 Electrical characterizations of GO thick films:

The resistivity, TCR (thermal coefficient of resistance) and activation energy at high and low temperatures parameters were used in the electrical study of prepared films. Equations 1, 2 and 3 were used to estimate resistivity, TCR and activation energy respectively [8].

$$\rho = \left(\frac{R \times b \times t}{l}\right) \Omega - m \tag{1}$$

Where,

 ρ = Resistivity, R = resistance of films,

b = breadth of film, t = thickness of the film, L = length of the film.

$$TCR = \frac{1}{R_o} \left(\frac{\Delta R}{\Delta T} \right) / {}^o C$$
⁽²⁾

Where,

 ΔR = change in resistance between temperature T_1 and T_2 , ΔT = temperature difference between T_1 and T_2 and R_o = Initial resistance of the film sample

$$\Delta E = \frac{\log R}{\log Ro} \times KT \tag{3}$$

Where,

 ΔE = Activation energy, R = Resistance at elevated temperature, R₀ = Resistance at 0°C.

2.3.2 Gas sensing study of GO thick films:

The properties of gas sensing were examined using a static gas sensing apparatus. Prepared GO thick films were used to as a sensing element.

21



Figure 2: Schematic diagram of electrical and gas sensing system.

The resistance of the thick film was measured at various operating temperatures in an environment of air as well as at different gas concentrations (ppm). Ra stands for film resistance in an atmosphere of air, while Rg stands for film resistance in an atmosphere of gas. The film resistance was calculated using the half-bridge method. The electrical and gas sensing static system used in this work is shown schematically in Figure 2. Gas sensing investigation were carried out in the presence of H_2S , NH_3 , LPG, NO_2 and ethanol gases in the surroundings of GO thick films at different operating temperatures. Under typical controlled environment circumstances, gas sensing studies were conducted out in a static gas system. Equation 4 was used to analyze the electrical resistance of thick films in the presence of air (Ra) and H_2S gas (Rg) in order to calculate the gas sensitivity [9].

$$S\% = \left|\frac{R_{air} - R_{gas}}{R_{air}}\right| \times 100 \tag{4}$$

3. RESULT AND DISCUSSION: 3.1 Electrical Characterization:

3.1.1 Resistivity:

The resistivity of the prepared GO thick films was calculated using Equation 1. Resistivity Play a very vital role in the gas sensing mechanism. The resistivity was found to be 65214.6Ω -m for the prepared GO thick films.



Figure 3: Resistance v/s temperature of prepared GO thick films

23

Figure 3 shows the plot of resistance versus temperature of prepared GO thick films. As seen in Figure 3, the resistance of the film decreases with increasing temperature, and the decrease in resistance with increasing temperature implies semiconductor behaviour of prepared films [8, 10].

3.1.2 Temperature coefficient of resistance (TCR):

The TCR of GO thick films was calculated using Equation 2. It is found that the temperature coefficient of resistance for GO thick films is -0.00613153 /⁰ C. TCR for GO thick films was revealed to be negative. The TCR of films is negative, demonstrating the semiconducting nature of the GO thick films [10].

3.1.3 Activation energy:

The activation energy of the prepared GO thick films was calculated using Equation 3. Figure 4 illustrates how the Arrhenius plot shows that the pattern is reversible in both heating and cooling cycles. At lower temperatures region prepared GO thick films have an activation energy value was found to be 0.065644 eV, whereas at higher temperatures region, it was found to be 0.147332 eV. Figure 4 depicts the Arrhenius plot of prepared GO thick films [11].



Figure 4: Arrhenius plot of prepared GO thick films

Table-1: Electrical	parameters of prepared	GO thick films
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Resistivity (Ω-m)	TCR (/°C)	Activation energy (eV)	
		LTR	HTR
65214.6	-0.00613153	0.065644	0.1473327

3.2 Gas sensing study of prepared GO thick films:

3.2.1 Sensitivity:

The sensitivity of prepared GO thick films was determined using the equation 4. Figure 5 depicts the gas sensitivity of GO thick films at different operation temperatures and selected gases. Figure 5 illustrates that the maximum response found to be to H_2S gas as compare to other selected gases. The maximum sensitivity was found at a temperature at about 90 °C and the gas concentration was 150 ppm. It is also observed that the prepared films shows less sensitivity to the LPG gas compared to other selected gases.

Equation 4 was used to calculate the prepared GO sensitivity of thick films. The gas sensitivity of GO thick films at various operating temperatures and with various gases is shown in Figure 5. Figure 5 shows that, when compared to other selected gases, H_2S gas elicited the greatest amount of response. At a temperature of about 90 °C and a gas concentration of 150 ppm, the highest

sensitivity was reported. Additionally, it has been noted that the prepared films are less sensitive to the LPG gas than they are to other selected gases.



Figure 5: sensitivity vs. operating temperature of GO thick films

3.2.2 Response and recovery time:

Response time and recovery time, which are measured in terms of the time needed to attain 90 percent of the maximum response when gas is in and 10 percent when gas is out, respectively, are the other two crucial characteristics for a gas sensor [12]. Figure 6 shows the response-recovery time behavior of the GO films at its ideal operating temperature. Figure 6 shows that the H_2S response and recovery times are around ~ 35 sec and ~ 60 sec seconds respectively.



3.2.3 Selectivity:

Selectivity is the ability of a sensor to react selectively to a collection of gases [12]. The highest selectivity to H_2S gas is shown in Figure 7. The temperature at which a sensor operates has a direct impact on the selectivity of the sensor toward a particular gas.

24



3.2.4 H₂S gas PPM variation:

Figure 8 illustrates the variation in GO thick film sensitivity with H_2S concentration of gas at 90° C. This film was exposed to various H_2S gas concentrations as shown in figure 8. The highest sensitivity values for H_2S gas were observed to be 150 ppm.



Figure 8: Variation of sensitivity with H₂S gas concentrations

3.2.5 H₂S gas sensing mechanism:

 H_2S is a reducing gas [13]. A reducing gas deteriorates the charge-carrying sensing layer of GO by transferring electrons, which causes a drop in hole carrier density and, in turn, a drop in conductivity. Due to the adsorption of gas molecules, the electrical resistance of the GO sensors specifically rises with exposure to H_2S . As a result of the reduction in holes caused by the adsorption of electron-donating gas, the Fermi level shifts to the valence band and an increase in electrical resistance is observed. The change in resistance gives response in the form of sensitivity [13-15].

CONCLUSIONS:

The sensing performance of the prepared GO thick films sensors can be described in the following ways based on the obtained results.

- 1. GO thick film could be prepared on glass substrate by using low cost screen printing technique.
- 2. Prepared GO thick films showed maximum sensitivity to H_2S gas.
- 3. The prepared thick film sensor showed very rapid response and recovery to H_2S gas.

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