

تحضير افلام ثلاثي أكسيد الأنتيمون الشوبية بثلاثي أكسيد انديوم

بطريقة التقطير لتطبيقات المستشعرات الغازية

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Sb₂O₃ Doped Indium Oxide Thin File Prepared dy drop casing Technique for gas sensor Application

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ABSTRACT

Thin films from Sb₂O₃:In₂O₃ were prepared with different weights and then the membranes were prepared using a drop casting method with a annealing temperature (250⁰C). Electrical properties have been studied and found when the activation energy decreases when the temperature rises and the conductivity increases. The films were examined as gas sensor against NH₃ gas at different operating temperatures (25, 100, 200 and 300) °C, and it was found that the samples a good sensitivity to NH₃ gas ~ 13.190 % at operating temperature 200°C, with relatively fast response time of 13.5 s and recovery time of 54 s.

Keyword: Drop casting , Thin film , Sb₂O₃ , In₂O₃ , Electrical properties , gas sensor

المخلص

حضرت أغشية رقيقة متعددة التبلور من ثلاثي أكسيد الأنتيمون النقي ، وافلام ثلاثي أكسيد الانتيوم المطعمة بثلاثي أكسيد انديوم بأوزان مختلفة بطريقة التقطير على قواعد من الزجاج وجفت بدرجة حرارة الغرفة ، وقد تم تلدينها بدرجة حرارة 250 درجة سيليزيه . وقد تم دراسة الخواص الكهربائية وجد ان طاقة زيادة عند درجة حرارة (25 و100 و200 و300) ووجد ان الافلام لها تحسبه تقريبا 190.13% عند سرعة استجابة 13.5

الكلمات المفتاحية : طريقة التقطير ، أغشية رقيقة ، ثلاثي أكسيد الأنتيمون ، ثلاثي أكسيد انديوم ، خواص كهربائية، مستشعر الغاز .

1- INTRODUCTION

Nanocomposites ,Composite Materials means the group of engineering materials produced by adding certain weight or volume ratios of one or more materials. (Reinforcement Materials) for Base Material (Matrix Material) So that the supporting materials are well combined and mixed with the mold material which ensures a homogeneous overlap in which the particles of the supporting materials are ideally distributed. The purpose of producing overlapping materials is to add certain properties to the mold material or to add traits that were not inherent to it [1, 2]. The nanocomposites applications are quite promising in the fields of microelectronic packaging, medicine, optical integrated circuits, drug delivery, sensors, packaging materials, coatings and adhesives....etc [3]. These advanced nanocomposites have many advantages such as low cost production and the possibility of device fabrication on large scale and flexible substrates [4].

2- Experimental

The pure thin film was prepared by using drop casting technique (0.1 gm of Sb_2O_3 , solve in 5 ml of ionic water with a 0.5 g PVA addition (PVA was used as a plaster only). The samples were doped with In_2O_3 addition were prepared with weights that were using the drop casting method, and then applied on a glass slide at room temperature. and Table (1) shows the ratios of the materials used in preparing the thin film $Sb_2O_3: In_2O_3$ in (5ml) ion watert.

Table (1) Ratios of materials used to prepare a thin film $Sb_2O_3:In_2O_3$

Sb_2O_3 gm	In_2O_3 gm	PVA mal	ion watert mal
Pure/0.1	0	0.5	5
0.09	0.01	0.5	5
0.08	0.02	0.5	5
0.07	0.03	0.5	5
0.06	0.04	0.5	5

study the electrical properties of $Sb_2O_3:In_2O_3$ films and interdigitated electrodes (IDEs) substrate to the electrical and sensor measurements. All substrates were washed thoroughly using Distilled water, and then left to Dry out.

3- Results and discussion

3-1-The electrical Properties of ($Sb_2O_3:In_2O_3$) thin film

The bulk electrical conductivity σ_{dc} is calculated for the prepared films by using equation (1)

$$\sigma = \sigma_0 \exp(-E_a / (k_B T)) \quad \dots\dots\dots (1)$$

The detailed results of these properties are:

3.1.1 The D.C Conductivity and Activation Energy

the relation between $\ln(\sigma/T)$ and the inverse absolute value of temperature for ($Sb_2O_3:In_2O_3$) thin films. The activation energy was calculated, and from these calculation, it can be seen that the high value existence for the activation energy in state of pure(Sb_2O_3) , and by adding Weight ratios (In_2O_3) the value of activation energy are decreasing for all ($Sb_2O_3:In_2O_3$) thin films as show in Figure.(2), the activation energy decreases with increasing the proportion of doped , this is consistent with the findings of the researcher [5]. This can be explained that increasing the proportion of impurities led to the approach of the Fermi level more towards the conduction band. in Figure (3) this conductivity increases with increasing temperature, as for the films vestiges we note there is also an increase in the conductivity values with the increase in the proportion of doped, this can be explained by the increase that led to the formation of impurity donor levels near the conduction band, with the findings of the researcher [6].

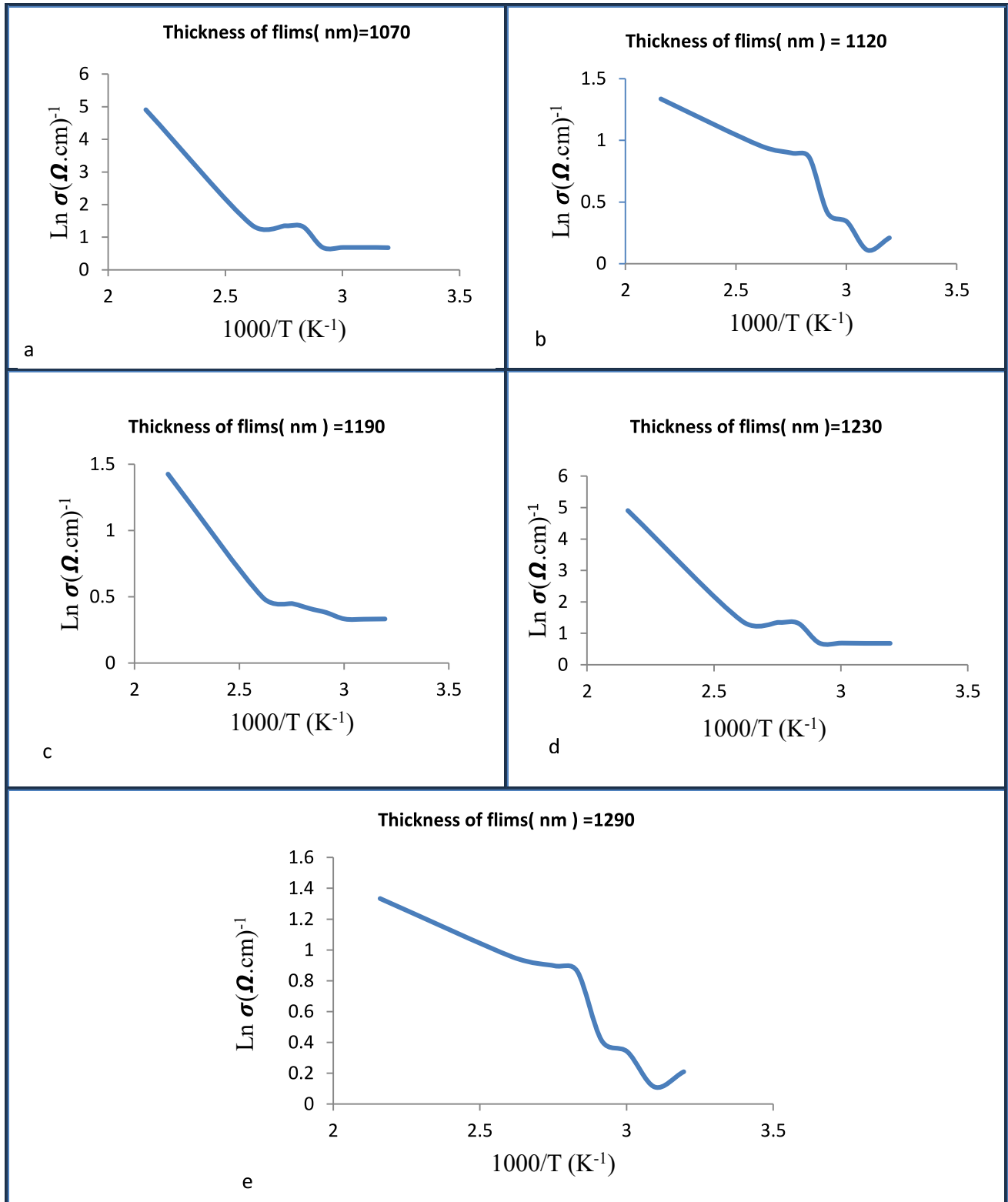


Figure (1): The Activation Energy of Sb_2O_3 - Sb_2O_3 : In_2O_3) Nanocomposites (a) Pure , (b) Sb_2O_3 9% wt In_2O_3 1% wt , (c) Sb_2O_3 8% wt In_2O_3 2% wt , (d) Sb_2O_3 7% wt 3 In_2O_3 3% wt , (e) Sb_2O_3 6% wt , In_2O_3 , 4% wt

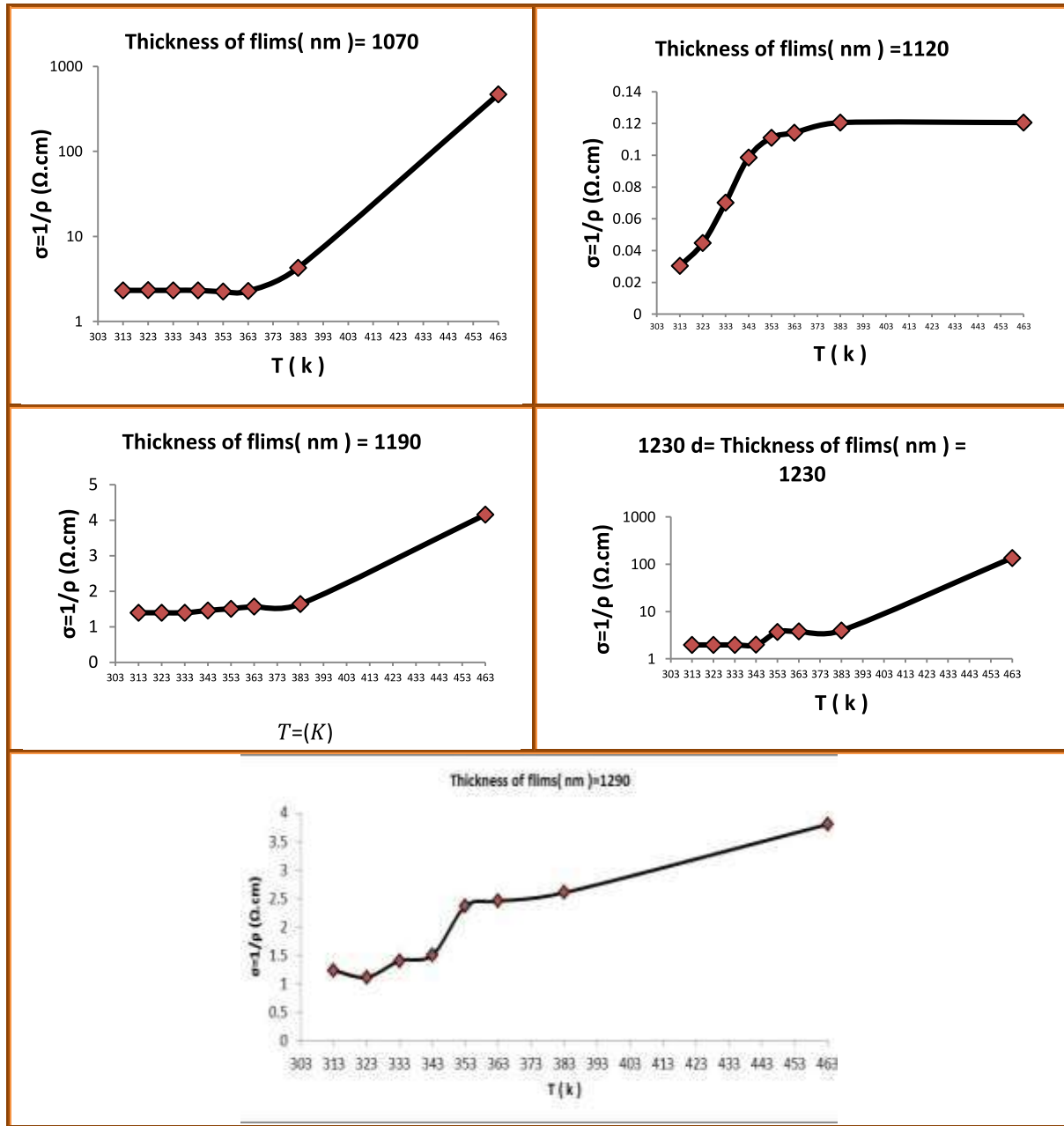


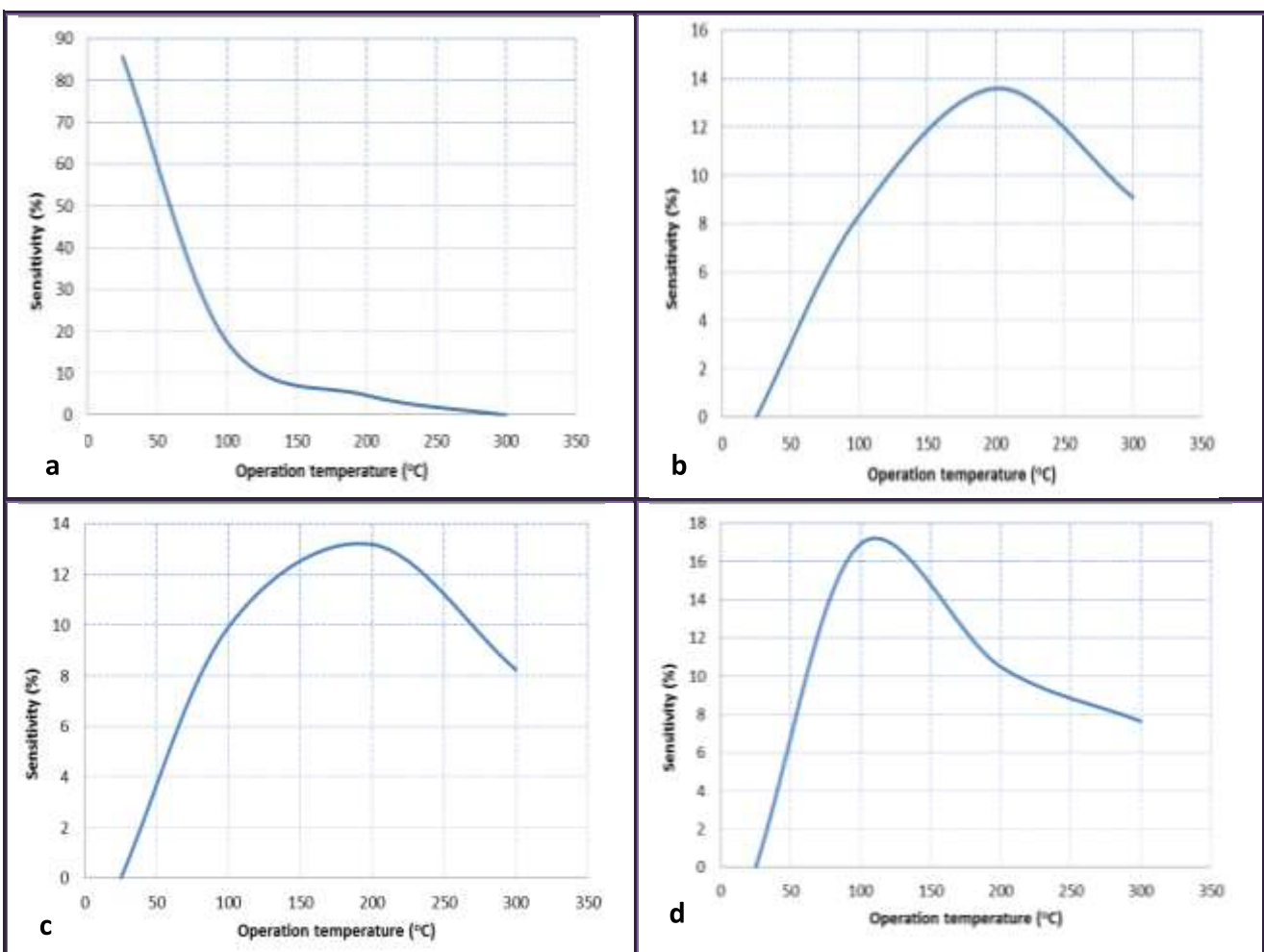
Fig (2) Variation conductivity with inverse absolute temperature (Sb_2O_3 - Sb_2O_3 : In_2O_3) nanocomposites (a) Pure (b) Sb_2O_3 9% wt In_2O_3 1% wt , (c) Sb_2O_3 8% wt In_2O_3 2% wt , (d) Sb_2O_3 7% wt In_2O_3 3% wt (e) Sb_2O_3 6% wt , In_2O_3 , 4% wt

4. Gas Sensor Measurement

4.1 Determination of Operation Temperature of the Sensor

The temperature at which the resistance of the sensor reaches a constant value is called operating temperature. The changing of resistance is just only influenced by the presence of amount of some gases of interest. Fig.(3 a) shows the variation of sensitivity as a function of operation temperature in the range (25,100 and 200) oC of pure (Sb_2O_3) sensor. The sensing test was done by using NH_3 : air mixed ratio and bias voltage 5V were applied on the electrodes for all samples. The variation of the temperature reveals resistance of the film decreases as the temperature increases from room

temperature to (100-200) °C showing a typical negative temperature coefficient of resistance due to thermal excitation of the charge carriers in semiconductor [7]. Figure (3.b,c,d,f) the sensitivity of all films increases with increasing of the operating temperature until (300) °C. This is attributed to increase in the rate of surface reaction of the target gas. The optimal temperature that has maximum values of temperature is (300) oC for all films. At this temperature the activation energy may be enough to complete the chemical reaction. Also we observed that increases and decrease in the sensitivity indicate the absorption and desorption phenomenon of the gases.the sensitivity is observed to increase at operating temperature (200) °C. After (200)°C temperature, the surface would be unable to oxidize the gas so intensively and the NH₃gas may burn before reaching the surface of the film at higher temperature. Thus, the gas sensitivity decreases with increasing temperature, also the increase of the of Sb₂O₃:In₂O₃causes decreasing in the sensitivity of thin film .The sensitivity of all films is shown in Table (2). These results are in agreement with [8].



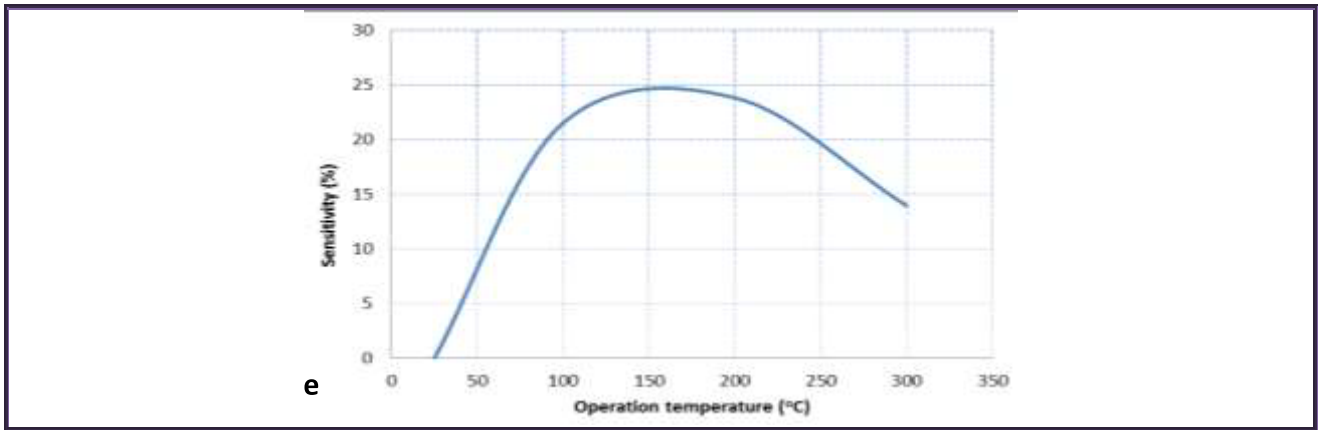


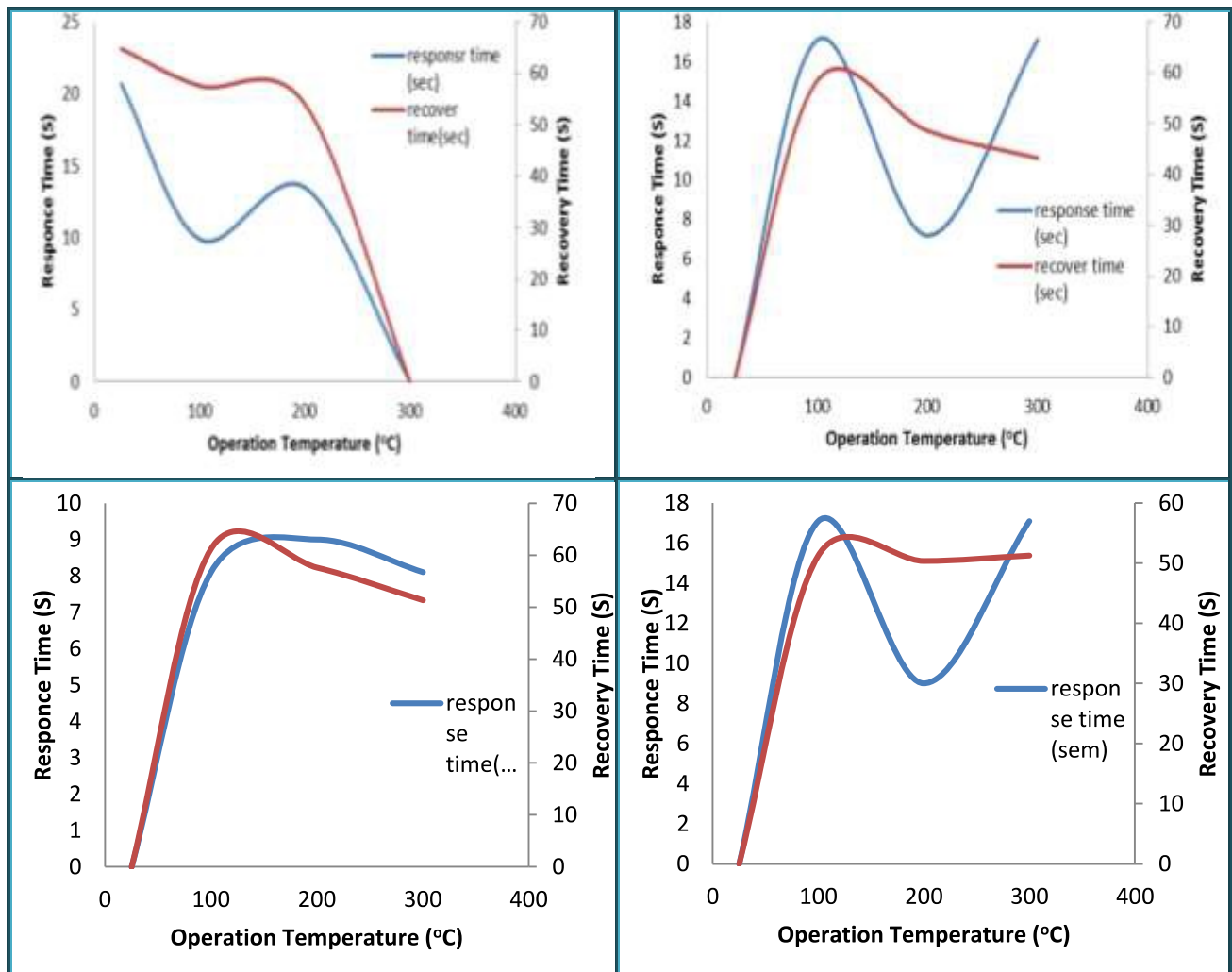
Fig (3): The variation of sensitivity with the operating temperature nanocomposites (a) Pure (b) Sb_2O_3 9% wt In_2O_3 1% wt , (c) Sb_2O_3 8% wt In_2O_3 2% wt , (d) Sb_2O_3 7% wt 3 In_2O_3 3% wt (e) Sb_2O_3 6% wt , In_2O_3 , 4% wt

Table (2): Sensitivity of the $\text{Sb}_2\text{O}_3:\text{In}_2\text{O}_3$ thin films with NH_3 gas

Sample	Operating Temp.($^{\circ}\text{C}$)	Sensitivity (%)
Sb_2O_3 purar	25	85.71428571
	100	17.49727174
	200	4.749568221
9% wt Sb_2O_3 , 1% wt In_2O_3	100	8.354559348
	200	13.60294118
	300	9.090909091
8% wt Sb_2O_3 , 2% wt In_2O_3	100	9.934640523
	200	13.19082377
	300	8.230769231
7% wt Sb_2O_3 , 3% wt In_2O_3	100	16.93160813
	200	10.49149338
	300	7.641921397
6% wt Sb_2O_3 , 2% wt In_2O_3	100	21.52880355
	200	23.83444339
	300	13.95572666

4.7.2 Response Time and Recovery Time

Figures (4a) to (4b) (4 c)(4d) (4e) show the relation between the response time and the recovery time with the operating temperature of the Sb_2O_3 , $Sb_2O_3:In_2O_3$ thin films for NH_3 gas and bias voltage 5V. From Table (4.5) note that the increasing Vol.% of In_2O_3 is due to increasing in response time and decreasing in recovery time at optimal temperature (200) oC The large recovery time would be due to lower operating temperature. At lower temperature O_2 species is more prominently adsorbed on the surface and thus it is less reactive as compared to other species of oxygen, O^- and O^{2-} .



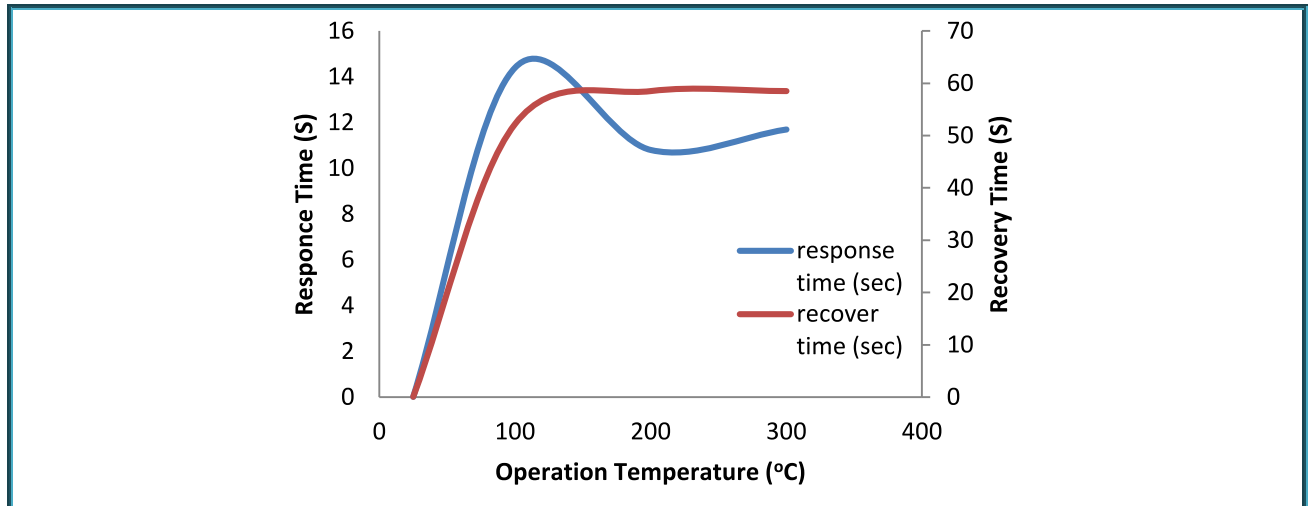


Figure (4.20e): The variation of response time and recovery time with operating (a) Pure (b) Sb₂O₃ 9% wt In₂O₃ 1% wt , (c) Sb₂O₃ 8% wt In₂O₃ 2% wt , (d) Sb₂O₃ 7% wt 3 In₂O₃ 3% wt (e) Sb₂O₃ 6% wt , In₂O₃, 4% wt

Table (3): The response time and the recovery time with the Temp.(°C) temperature of Sb₂O₃, Sb₂O₃:In₂O₃ thin films at different Wight rich

Sample	Temp.(°C)	Response Time (s)	Recovery Time (s)
Sb ₂ O ₃ pear	25	20.7	64.8
	100	9.9	57.6
	200	13.5	54
	300	0	0
9% wt Sb ₂ O ₃ , 1% wt In ₂ O ₃	25	0	0
	100	17.1	58.5
	200	7.2	48.6
	300	17.1	43.2
8% wt Sb ₂ O ₃ , 2% wt In ₂ O ₃	25	20.7	64.8
	100	9.9	57.6
	200	13.5	54
	300	0	0
7% wt Sb ₂ O ₃ , 3% wt In ₂ O ₃	25	0	0
	100	17.1	51.3
	200	9	50.4
	300	17.1	51.3
6% wt Sb ₂ O ₃ , 2% wt In ₂ O ₃	25	0	0
	100	14.4	52.2
	200	10.8	58.5
	300	11.7	58.5

4.8 Conclusions

Electrical Properties

- 1- The D.C electrical conductivity for the (Sb₂O₃ Sb₂O₃:In₂O₃) nanocomposites increases with increasing of temperature and with increasing of the Weight ratios of In₂O₃ wt%.
- 2- The activation energy for D.C electrical conductivity for (Sb₂O₃, Sb₂O₃:In₂O₃) nanocomposites decreases with increasing of the Weight ratios of In₂O₃ wt% .

4.8.4 Gas Sensor Measurement

- 1- The sensing measurements show that the films have high sensitivity to these gases and vapors. The best result recorded for sensitivity at 4% In₂O₃. Sensitivity reactions of films decrease with increasing of In₂O₃ %.
- 2- The gas sensor at the operating temperature of an in the increasing response time and an Decreases in the recovery time.

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