

## The Structural and Optical Properties of Cobalt dioxide (CoO<sub>2</sub>) Thin Films deposited via (SCSP) Technique for photovoltaic applications

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### ABSTRACT

The research included the preparation and then studying the structural and optical properties of the cobalt dioxide (CoO<sub>2</sub>) films. The latter films were prepared using a semi-computerized spray pyrolysis technique (SCSPT). The X-ray diffraction gave polycrystalline nature with crystal system trigonal (hexagonal axes), and the Energy Dispersive X-ray spectroscopy (EDX) showed that all films contain the elements (Co and O) indicating formation of (CoO<sub>2</sub>) films with high purity. FTIR measurements showed of chemical bonds of CoO<sub>2</sub> clearly. Scanning Electron Microscopy (SEM ) Showed clearly that the formed thin films under the optimum conditions were homogeneous, dense and compact, and Atomic Force Microscopy (AFM) results showed that the topography of the film surface where surface roughness was found to be 7.91 nm, root mean square was 9.69 nm., and the average granularity diameter was 78.00 nm. The optical properties (absorbance, absorption coefficient, extinction coefficient, refractive index, optical Conductivity, the real  $\epsilon_1$  and imaginary  $\epsilon_2$  part of the dielectric constant ) were decreased with increasing the wavelength, while the transmittance increases with increasing wavelength. The optical energy gap was (1.98eV) and this is a good optical energy gap values for photovoltaic applications.

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### الخصائص التركيبية والبصرية للأغشية الرقيقة لثاني أكسيد الكوبالت المرسبة خلال تقنية

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### الكلمات المفتاحية:

الخصائص التركيبية والبصرية لـ  
CoO<sub>2</sub>

### الخلاصة

تضمن البحث تحضير ثم دراسة الخصائص التركيبية والبصرية لأغشية ثاني أكسيد

تقنية الرش الحراري  
شبة المحوسبة  
ثاني اوكسيد الكوبلت

الكوبالت ( $CoO_2$ ) تم تحضير الأغشية باستخدام تقنية الانحلال الحراري بالرش شبه المحوسبة (SCSPT) أعطى حيود الأشعة السينية طبيعة متعددة البلورات مع نظام بلوري ثلاثي الزوايا (محاور سداسية) ، وأظهر التحليل الطيفي للأشعة السينية المشتتة للطاقة (EDX) أن جميع الأفلام تحتوي على العناصر الكوبلت و الاوكسجين التي تشير إلى تكوين أغشية ( $CoO_2$ ) بنقاوة عالية . أظهرت قياسات FTIR الروابط الكيميائية لثاني أكسيد الكوبلت بشكل واضح. كما أظهر الفحص المجهر الإلكتروني (SEM) بوضوح أن الأغشية الرقيقة المتكونة في ظل الظروف المثلى كانت متجانسة وكثيفة ومضغوطة . وأظهرت نتائج المجهر القوة الذرية (AFM) تضاريس سطح الفيلم حيث تم العثور على خشونة السطح لتكون ٧.٩١ نانومتر ، الجذر التربيعي ٩.٦٩ نانومتر ، وكان متوسط قطر الحبيبات ٧٨.٠٠ نانومتر. الخواص الضوئية (الامتصاصية ، معامل الامتصاص ، معامل الانقراض ، معامل الانكسار ، الموصلية الضوئية ، الجزء الحقيقي والخيالي من ثابت العزل) انخفضت مع زيادة الطول الموجي ، بينما تزداد النفاذية مع زيادة الطول الموجي. كانت فجوة الطاقة الضوئية ١.٩٨ إلكترون – فولت وهذه قيمة جيدة لفجوة طاقة بصرية للتطبيقات الكهروضوئية .

## 1. INTRODUCTION

Spray pyrolysis technique (SPT) is one of the most promising techniques which have been employed to prepare metal oxide thin films like  $CoO_2$ ,  $ZnO$ ,  $CuO$ ,  $CeO_2$ , etc [1][2]. The SPT technique achieves the required compatibility for the development of solar cells, p-n junction diode, hetero junction diodes, and electrochemical electrodes [3][4]. Spraying technique is the most common in today time because of its applicability to producing a variety of conducting and semiconducting materials. Spraying deposition is a notable method for synthesizing thin layers that utilized for different applications. Spray pyrolysis has several features such as inexpensiveness, no vacuum chamber required, and the ability to synthesize sub-micron thin films [5]. In the case of films, it can be sprayed over an area larger than a lab-scale that can be employed at industrial production processes [6]. Five types of cobalt oxide ( $CoO_2$ ,  $Co_2O_3$ ,  $CoO(OH)$ ,  $Co_3O_4$ , and  $CoO$ ) Cobalt oxide thin films have attracted substantial research effort in recent years because of their potential application in various technological areas. They can be used as high temperature solar selective absorbers [7]. The conductivity of cobalt oxide is usually p-type at

low temperature and intrinsic at high temperature[8], measured values of the band gap are around 1.6 eV[9][10].

## 2. EXPERIMENTAL PART

$CoO_2$  thin films were prepared using the SPT by mixing raw material as shown in the table(1) in (100) ml of deionized water.

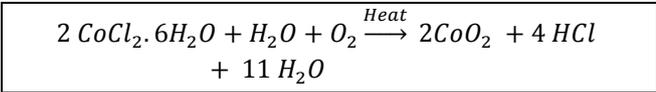


Figure1: shows a solution of  $CoO_2$

Table 1: Properties of selective raw materials

| Compound                    | Chemical formula                     | Molecular weight (gm/mol) | Concentration | Purity | Supplier      |
|-----------------------------|--------------------------------------|---------------------------|---------------|--------|---------------|
| Cobalt chloride hexahydrate | CoCl <sub>2</sub> .6H <sub>2</sub> O | 237.93088                 | (0.02) M      | 98%    | Sigma-Aldrich |

The CoO<sub>2</sub> films were deposited on the glass substrate, the substrate temperature was (350±5C<sup>o</sup>), by using the air as a carrier for precursor solution. The separator between sprayer nozzle and substrate was (35cm). Evaporation and decomposition were occurred during the deposition precess.The formation reaction of (CoO<sub>2</sub>) thin film can be described by the chemical equation below:



It was done using a semi-computerized spray pyrolysis technique(SCSPT) which was handmade specifically for this work to be prepared and irradiated by laser to thin films. Where in this system the spray nozzle and the irradiation laser move in the X-Y plane according to coordinates such as speed, distance and area of deposition were controlled by the researcher as well as many other parameters figure(2):

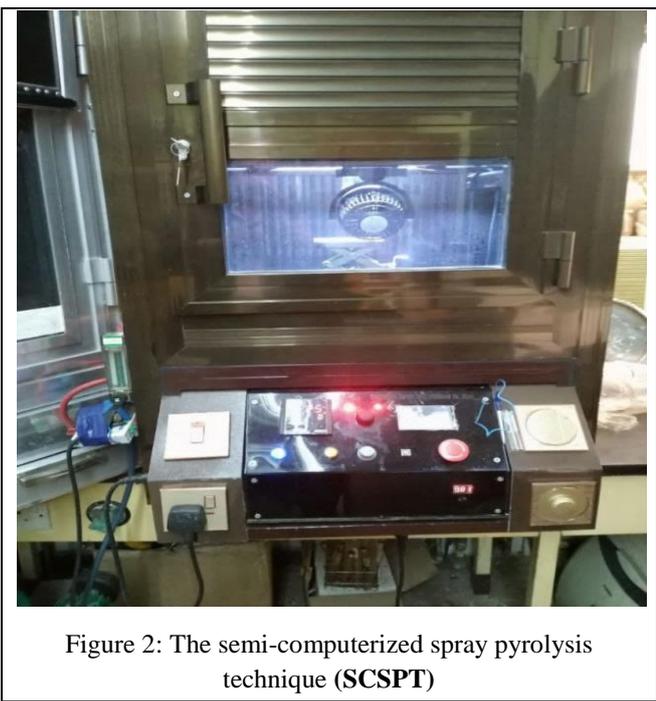


Figure 2: The semi-computerized spray pyrolysis technique (SCSPT)

the Structural and optical properties of CoO<sub>2</sub> films studies using (XRD,EDX,AFM,FTIR and UV-VIS ).The samples were of thickness (1µm) measured using interference fringes method

**3. RESULTS AND DISCUSSION:**

**3.1: Structural Properties:**

**3.1.1 X-ray diffraction:**

The XRD diagram of the cobalt dioxide (CoO<sub>2</sub>) films can be seen in figure(3) which is representing a polycrystalline structure with trigonal crystal system. It shows diffraction peaks at 2θ = (31.8306°, 24.2525°, and 23.454°). Cell parameters used were as follows a= 2.82080 Å c= 4.24030 Å. , the grain size D=108.029 nm and stress values=5.3368.These values were matched JCPDS 42-1467 Data Card [11]. The latter is in agreement with[12].

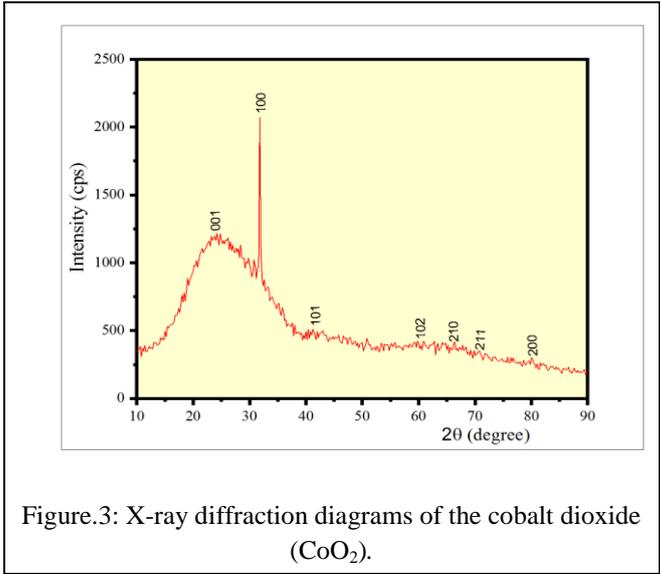


Figure.3: X-ray diffraction diagrams of the cobalt dioxide (CoO<sub>2</sub>).

**3.1.2 Elemental Analysis of cobalt oxide (CoO<sub>2</sub>) film :**

The energy dispersive X-ray spectroscopy (EDX) of cobalt oxide (CoO<sub>2</sub>) given in figure(4 ). It can be clearly observed that all films contain the elements (Co,O) as expected, indicating formation of the (CoO<sub>2</sub>) films with high purity , this result agreed with [13].

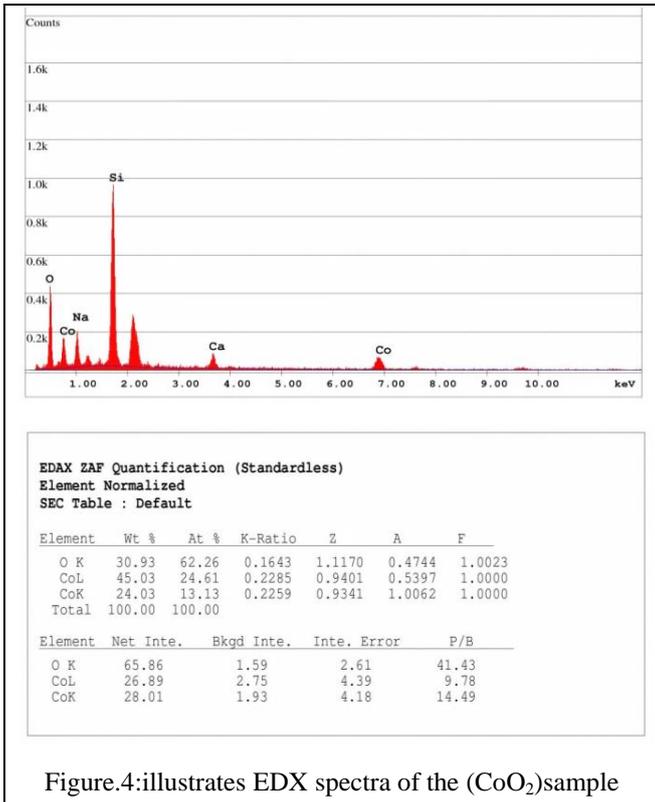


Figure.4:illustrates EDX spectra of the (CoO<sub>2</sub>)sample

### 3.1.3 SEM of cobalt oxide (CoO<sub>2</sub>) film :

The surface topography of cobalt oxide (CoO<sub>2</sub>) films was studied using a scanning electron microscope (SEM) It was observed that the formed thin films under the optimum conditions namely, homogeneous, dense and compact. The SEM micrographs of cobalt oxide (CoO<sub>2</sub>) films displayed in Figure(5).

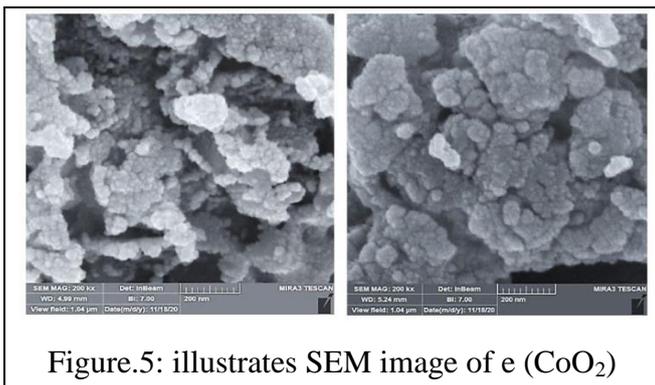


Figure.5: illustrates SEM image of e (CoO<sub>2</sub>)

### 3.1.4 Atomic Force Microscopy(AFM)of CoO<sub>2</sub> film:

Surface Topography of the prepared thin films is displayed in figure(6), where the AFM was examined the CoO<sub>2</sub> films. Surface roughness was found to be 7.91 nm, root mean square is 9.69nm,and the average granularity diameter was 78.00 nm.

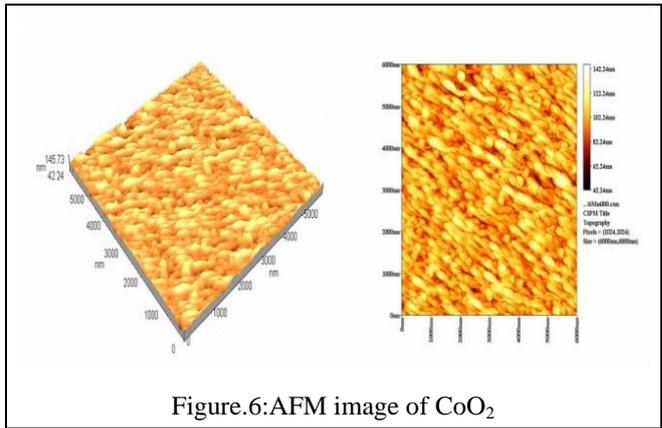


Figure.6:AFM image of CoO<sub>2</sub>

### 3.1.5 FTIR of cobalt oxide(CoO<sub>2</sub>) thin film :

The FT-IR spectra of the (CoO<sub>2</sub>) samples, ranging from 4000 to 400 cm<sup>-1</sup>, in figure(7) the strong peak was at about 3500 cm<sup>-1</sup> which is attributed to the stretching vibration, of the OH group of water molecular[14][15]and hydrogen-bound OH groups [16] . The weak band at 1635 cm<sup>-1</sup> is due to the bending mode of water molecules. [17].Two distinctive bands at 664 and 570 cm<sup>-1</sup> are the characteristics of the Co-O [18-24] .The band at 1066 cm<sup>-1</sup> was characteristic of the C-O[25,26].

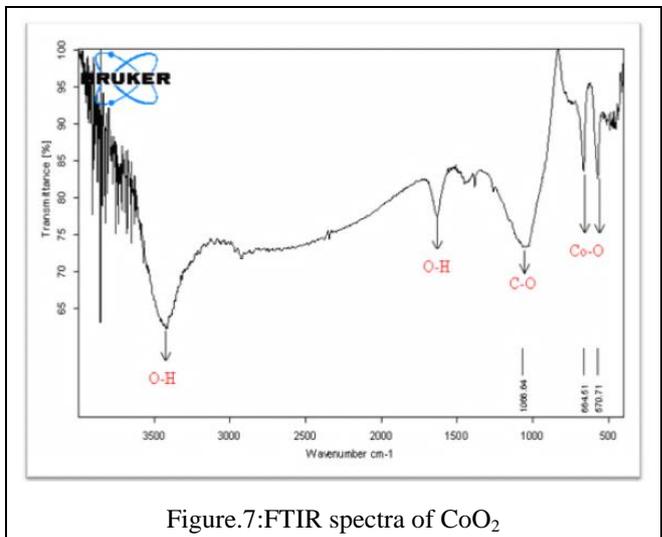


Figure.7:FTIR spectra of CoO<sub>2</sub>

## 3.2 The Optical Spectrum of CoO<sub>2</sub> Films:

### 3.2.1 Absorbance:

Figure.8 illustrates the optical absorbance (A) of cobalt oxide (CoO<sub>2</sub>) films with wavelength. Through the figure it can be noticed that the absorbance decreases with increasing the wavelength ,this physically means that incident photon was not able to excite the electron and transfer it from valence

band to the conduction band because the energy of incident photon was less than the value of the energy gap value of the semiconductor. This therefore leads to the absorbance decrease with increasing of wavelength[27].

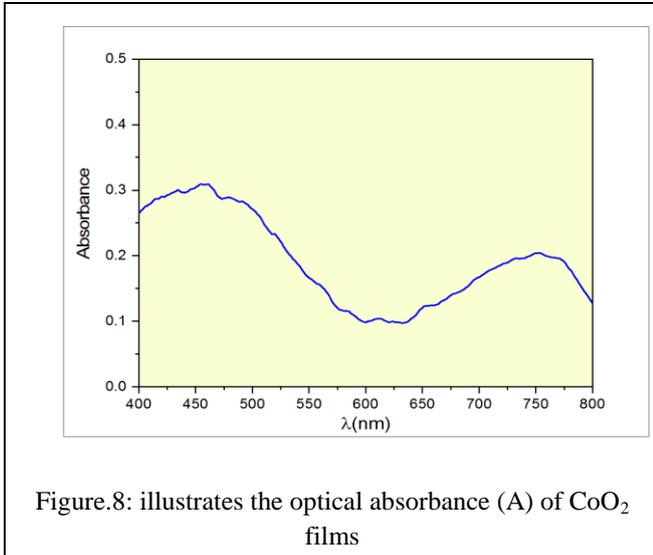


Figure.8: illustrates the optical absorbance (A) of CoO<sub>2</sub> films

**2.2 Absorption coefficient:**

The absorption coefficient( $\alpha \text{ cm}^{-1}$ ) was calculated in the fundamental absorption region using Lambert law [28].

$$I = I_0 \exp(-\alpha t) \dots \dots \dots (1)$$

Where t: is film thickness, I: is the intensity of transmitted light. If  $(I / I_0) = T$  then

$$\alpha = 2.303 \left( \frac{A}{t} \right) \dots \dots \dots (2)$$

Figure.9 illustrates the absorption coefficient ( $\alpha$ ) with wavelength for cobalt oxide (CoO<sub>2</sub>)films. The measured values of the absorption coefficient are in order of  $10^4 \text{ cm}^{-1}$ . Also, in this figure observed the absorbance coefficient decreases with increasing wavelength the reason for this is that the absorption coefficient depends on the absorbance.

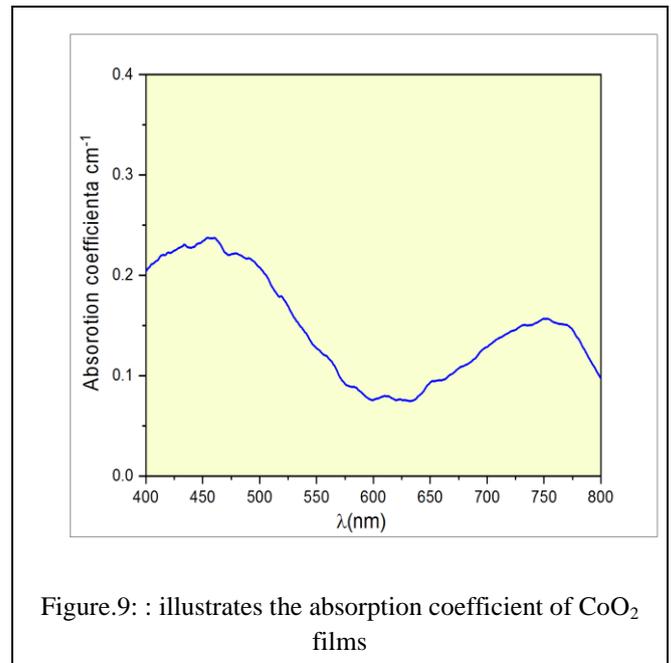


Figure.9: : illustrates the absorption coefficient of CoO<sub>2</sub> films

**3.2.3 Transmittance :**

Figure.10 shows the transmittance spectrum in wavelength range (400- 800) nm for the cobalt oxide (CoO<sub>2</sub>)films , Transmittance demonstrated behavior opposite of absorbance as shown transmittance of prepared film increases with increasing of wavelength[27].

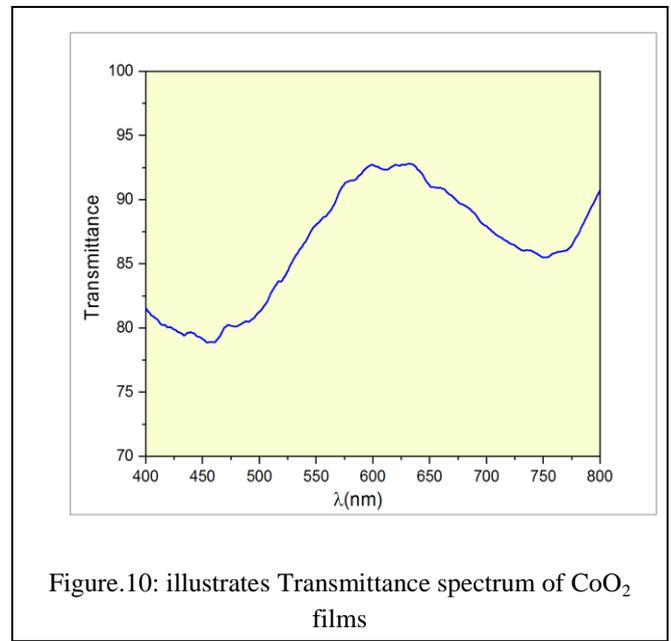


Figure.10: illustrates Transmittance spectrum of CoO<sub>2</sub> films

**3.2.4 Extinction coefficient:**

The extinction coefficient ( $K_0$ ) can be evaluated by the following relation [29]:

$$K_0 = \alpha \lambda / 4\pi \dots \dots \dots (3)$$

Where  $\lambda$ : is wavelength and  $\alpha$  : The absorption coefficient

Figure.11 shows the variation of extinction coefficient of the cobalt oxide (CoO<sub>2</sub>) films with the wavelength, in this figure it can be observed that the extinction coefficient decreases with increasing wavelength being dependent on absorption coefficient( $\alpha$ ).

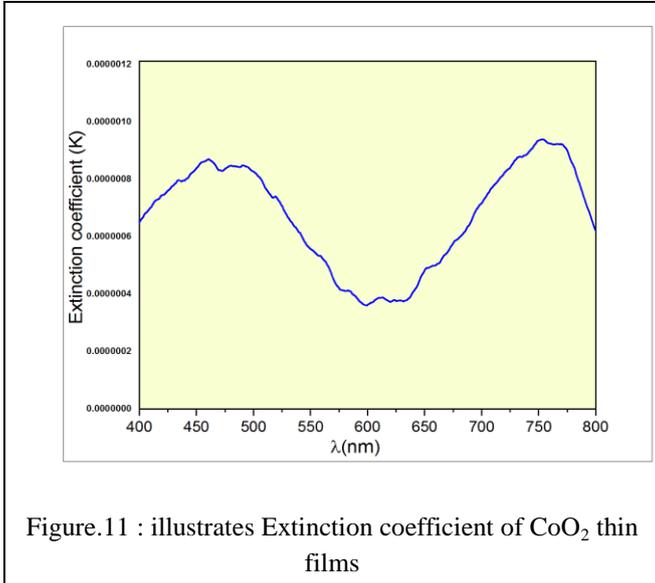


Figure.11 : illustrates Extinction coefficient of CoO<sub>2</sub> thin films

**3.2.5 Refractive index:**

Refractive index(n) one of the fundamental properties of an optical that can be measured from the following relation [30]:

$$n = \left[ \frac{(1 + R)^2}{(1 - R)^2} - (k_o^2 + 1) \right]^{1/2} + \frac{(1 + R)}{(1 - R)} \dots (4)$$

Figure.12 shows the refractive index of the cobalt oxide (CoO<sub>2</sub>) sample It can be noticed from this figure that the refractive index (n) decreases when the wavelength increases.

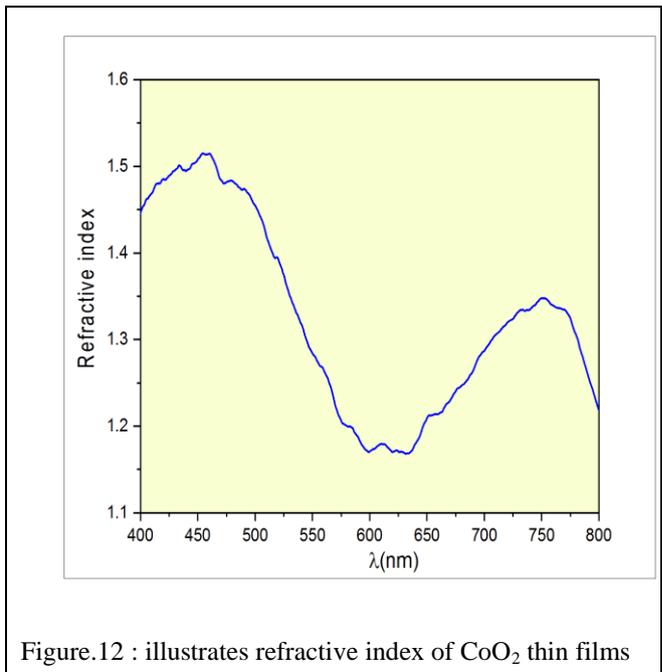


Figure.12 : illustrates refractive index of CoO<sub>2</sub> thin films

**3.2.6 Dielectric constant:**

The real ( $\epsilon_1$ ) and imaginary ( $\epsilon_2$ ) dielectric constant is determined using the relation[31]:

$$\epsilon_1 = (n^2 - K_o^2) \dots \dots \dots (5)$$

$$\epsilon_2 = (2 n K_o) \dots \dots \dots (6)$$

Figure.13.A.B shows the variation with a wavelength in (nm) of the real  $\epsilon_1$  and imaginary  $\epsilon_2$  part of dielectric constant values cobalt oxide (CoO<sub>2</sub>) sample . It can be noticed from this figure that the real part  $\epsilon_1$  and imaginary  $\epsilon_2$  part of dielectric constant decreases when the wavelength increases. This is because the real  $\epsilon_1$  and imaginary  $\epsilon_2$  part of dielectric constant depend on the extinction coefficient and the refractive index.

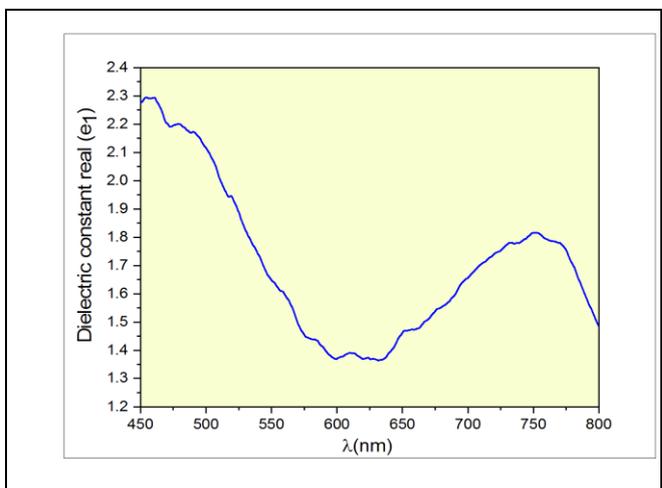


Figure.13.A : Real part  $\epsilon_1$  of the dielectric constant of  $\text{CoO}_2$  thin films

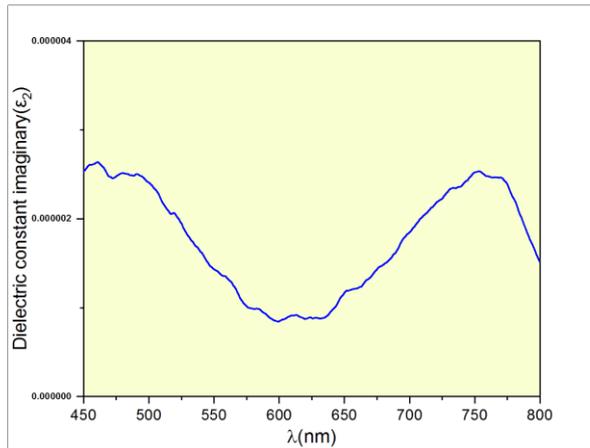


Figure.13.B : imaginary part  $\epsilon_2$  of the dielectric constant of  $\text{CoO}_2$  thin films

**3.2.7 Optical Conductivity:**

Optical conductivity ( $\sigma$ ) is expressed in the following relationship[31]:

$$\sigma = \frac{\alpha n c}{4\pi} \dots\dots(7)$$

where c: speed of light.

Figure.14 shows the optical conductivity ( $\sigma$ ) of cobalt oxide ( $\text{CoO}_2$ ) sample variation of with the wavelength It can be noticed from this figure that the optical conductivity decreases when the wavelength increases. This is because it depends on the absorption coefficient and the refractive index.

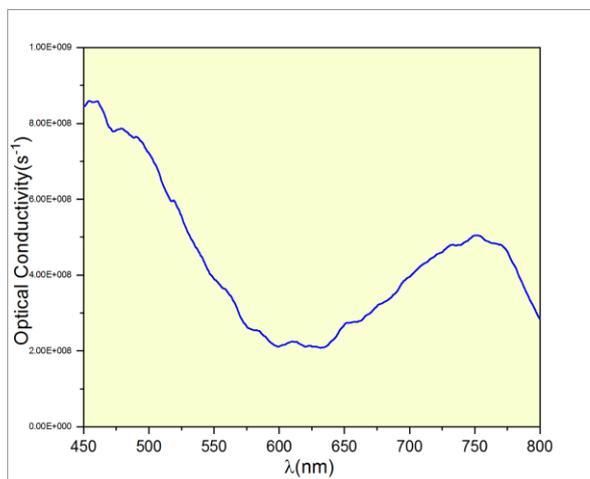


Figure.14 : illustrates optical conductivity ( $\sigma$ ) of ( $\text{CoO}_2$ )

**3.2.8: Optical energy gap:**

To measure the optical band gap for the thin films, we use Tauc's relation as follows [31]:

$$\alpha h v = A(hv - E_g)^n \dots\dots\dots(8)$$

Where A: is constant, hv: photon energy,

$E_g$ : the optical energy gap, and an index (n) could take different values according to the type of electronic transition.

The optical energy gap of cobalt dioxide ( $\text{CoO}_2$ ) sample in Figure.15. The optical energy gap (1.98eV) and that good variation of optical energy gap values were observed for photovoltaic applications[9].

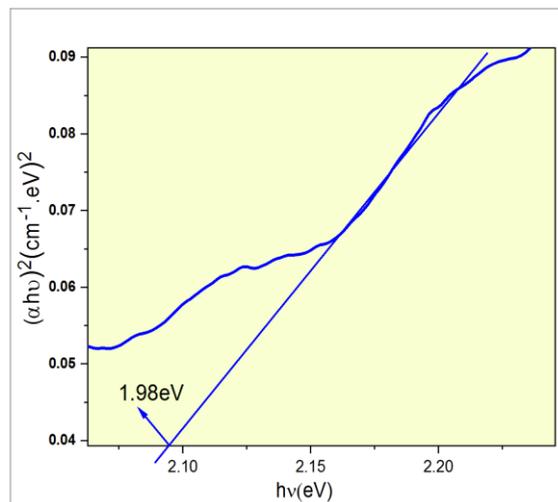


Figure.15 :optical energy gap of  $\text{CoO}_2$  thin films

**4. CONCLUSION :**

The structural and optical analysis of ( $\text{CoO}_2$ ) films shows that using (SCSP) technique a useful method for the deposition of cobalt dioxide ( $\text{CoO}_2$ ) films. The XRD diffraction gave polycrystalline nature with Crystal system trigonal (hexagonal axes), and the EDX showed that all films contain the elements (Co and O) indicating formation of the ( $\text{CoO}_2$ ) films with high purity, also showed FTIR measurements of chemical bonds of  $\text{CoO}_2$  clearly. (SEM) Show clearly was that the

formed thin films under the optimum conditions were homogeneous, dense and compact ,and (AFM) showed the topography of the film surface where surface roughness was found to be 7.91 nm, root mean square is 9.69 nm., and the average granularity diameter was 78.00 nm.. The optical properties(absorbance, absorption coefficient, extinction coefficient, refractive index, optical Conductivity, the real  $\epsilon_1$  and imaginary  $\epsilon_2$  part of the dielectric constant )were decreased with increasing the wavelength, while the transmittance increases with increasing wavelength. The optical energy gap was (1.98eV) and this is a good of optical energy gap values for photovoltaic applications.

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