Using Spray Pyrolysis Technique to Prepare PBS Lead Sulfide Thin Films and Study Their Structural and Electrical Properties as Function of Thickness

Rana Kadhim Abdulnabi

***Abstract:** Lead sulfide (PbS) thin films are prepared by "solution "from, lead acetate" pb (CH3COO)2 and Thiourea CS(NH2) using Spray Pyrolysis Technique (SPT) on glassy substrates at temperature of 200C° with a variable thicknesses of 100, 200, 300 nm. The "structural properties are approved by X-Ray Refraction (XRR), which helped in displaying the films having a cubic structure such as NaCl The size of the crystal grains of PbS films are shown to be increased when increasing the thickness of PbS films. Roughness of the films was measured according to the Root Mean Square (RMS). This was achieved using Scanning Microscope (SM). With increasing film thickness to the mentioned values, results showed an increase with RMS, electrical conductivity of the PbS film to $0.0186*10^{-3}$, $0.4166*10^{-3}$ and $0.9090*10^{-3}$ $(\Omega.cm)^{-1}$ and with charge-carrier concentration as well to $0.298*10^{11}$, $1.1*10^{11}$, $12*10^{11}$ cm^{-3}.:*

الخلاصة:

في هذا البحث تم تحضير اغشية كبريتيد الرصاص(*PbS*) بأستخدام محلول من خلات الرصاص $(CH_3COO)_2$ *PbS* والثايوريا $CS(NH_2)$ على قاعدة زجاجية بدرجة حرارة $200\ \overset{o}{C}$ بطريقة الرش الكيميائي الحراري .

تم دراسة الخصائص التركيبية عند اسماك متغيرة *(100,200,300)* بأستخدام اشعة *X- Ray* التي اوضحت الشكل البلوري للاغشية وقد لاحظنا زيادة الحجم الحبيبي للاغشية بزيادة سمك الغشاء , كما تم قياس خشونة السطح للاغشية بأستخدام فحص *STM*.

كما تم دراسة الخصائص الكهربائية ومنها التوصيلية الكهربائية والتي قيمها $(0.0186,0.4166,0.9090)*10^{-3}(\Omega.cm)^{-1}$ وحاملات الشحن للاغشية والتي قيمها $(0.298,1.1,12)*10^{11}\ cm^{-3}$

***Keywords:** lead sulfide, chemical pyrolysis, thin film, semiconductor*

I. INTRODUCTION

Lead sulfide PbS (a narrow gap semiconductor material) is considered as an attractive semiconductor material that is used in widely spread applications, such as electronic and optoelectronic devices [1, 2, 3]. PbS importance is due to number of factors such as its cubic lattice in unit cell of cube face center [4, 5, 6], its relatively small band gap (0.41 eV at 300 K^0) and the relative large excitation according to Bohr Radius "18 nm" [7]. Furthermore, PbS exhibits large size effects of quantization because of the small effective mass of electron and holes and the large dielectric constant [8, 9, 10]. These properties have been correlate with the growth conditions and the nature of substrates. A great deal of research efforts has been devoted to the synthesis of PbS nanoparticles of various size in a controlled manner, for its vigorous utilization in many fields, such as infrared photography and infrared detection application [11,12], diode laser, humidity, temperature sensors, decorative, Solar control coatings [13,14], Pb2+ ion selective sensors [15] and solar absorption [16]. Furthermore, many other research groups have shown a great interest in the development and study of this material by various deposition processes. A lot of methods have been used for the deposition of "PbS" thin films, for example thermal evaporation method [17], chemical bath method (CBM) [18], microwave heating [19, 20] , electro-deposition (ED) [21], and chemical spray method (CSM) [22]. Among different methods, spray pyrolysis is an attractive one due to its capability of growing large-area films with respectable uniformity, ease of preparation and low cost [17, 23].

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II. EXPERIMENTAL PART

Experimental part is divided to four major steps:

A. Reaction Solution Preparation

Specific weights of lead acetate [pb$(CH_3COO)_2$] (0.325) gm. and Thiourea $(CS(NH_2)_2$ (0.076) gm. were taken separately each in 100 ml of distilled water to create the melted reaction solution to achieve a molarity equals (0.01) for each of the two compounds (solutions). To prepare the final solution, 20 ml from both lead acetate and Thiourea solutions are mixed together. This compound solution then are placed in a glass tub of 100ml capacity and placed in the magnetic mixing device (magnetic stirrer) for 15 min.
Lead sulfide, is then deposited on heated glass substrate (200^oK), by spraying a suitable amount of solution from a sprayer with a 20cm distance between the heated substrate and the nozzle. Filtered Argon gas was used in the spraying system as the carrier gas during the deposition process.

B. Preparing Substrates

One of the important factors that affect the quality of the film is the cleanliness of the glass substrates to be used for deposition. Therefore glass substrates are treated as follows prior to deposition;

1. washed with cleansing agent and water,
2. Placed in Chromic Acid solution for (20-15 minutes),
3. Removed from the organic solvent, washed in distilled water, and placed in an ultrasonic device for (15) minutes.
4. The dried air dryer.

C. Deposition Process

After the preparation of the final reaction solution and placing it inside the glass tube contained within the spraying system, the cleaned glass substrates to be used in

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deposition are placed on the heating base within 20 cm distance. Deposition process is in the form of spraying pulses of 2 seconds period and 28 seconds stop period between pulse-running. The Argon (carrier gas) flow rate of the spraying system was fixed on the value of 20 L / min.

III. RESULTS AND DISCUSSION

A. Structural Properties

Fig. (1): For films deposited at varies thickness are shown is "sharp" peaks in [1 1 1] and [2 0 0], but small peaks of the [2 2 0], [3 1 1], [3 2 2] and [4 0 0]" planes, as confirmed by "standard ASTM" card. When increase the thickness film the intensity of the peaks increased and the crystallization of PbS thin films were improved that. However, this result was not achieved when the thickness applied was 300nm. For all films the better orientation value of [200] plane has the highest value comparing with other planes. This result indicate a strong orientation growth the length of that plane. The size of the crystal grains (D) of the PbS films was determined by the Scherer formula [24].

$$D=0.9\lambda/\beta \cos\theta \qquad \ldots\ldots (1)$$

Where, λ is the wavelength of the X-ray, 1.5406 Å, θ is the peak position and β is the full width at half maximum (FWHM) of the peaks.

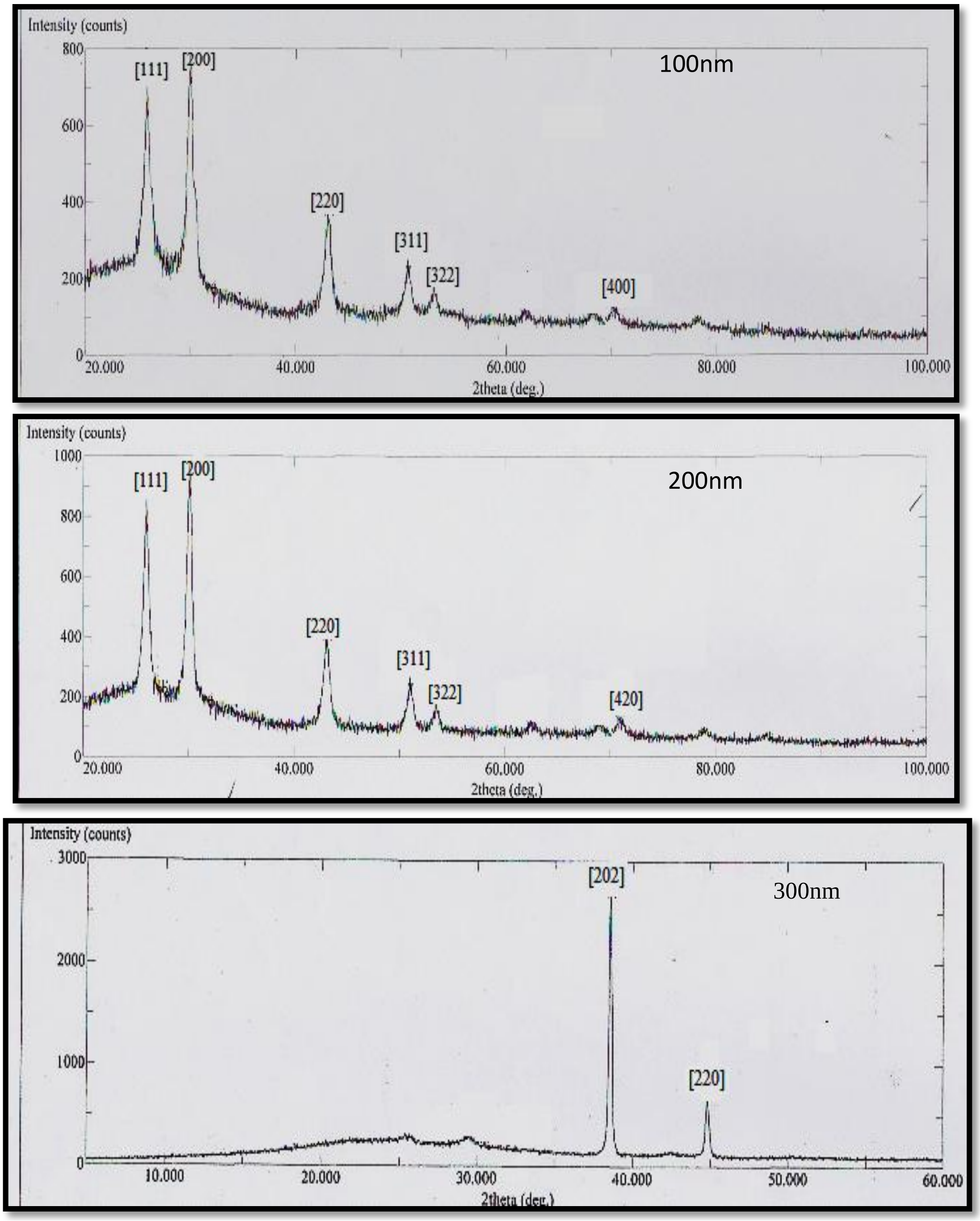

Figure (1) XRD of the PbS thin films with varies thickness

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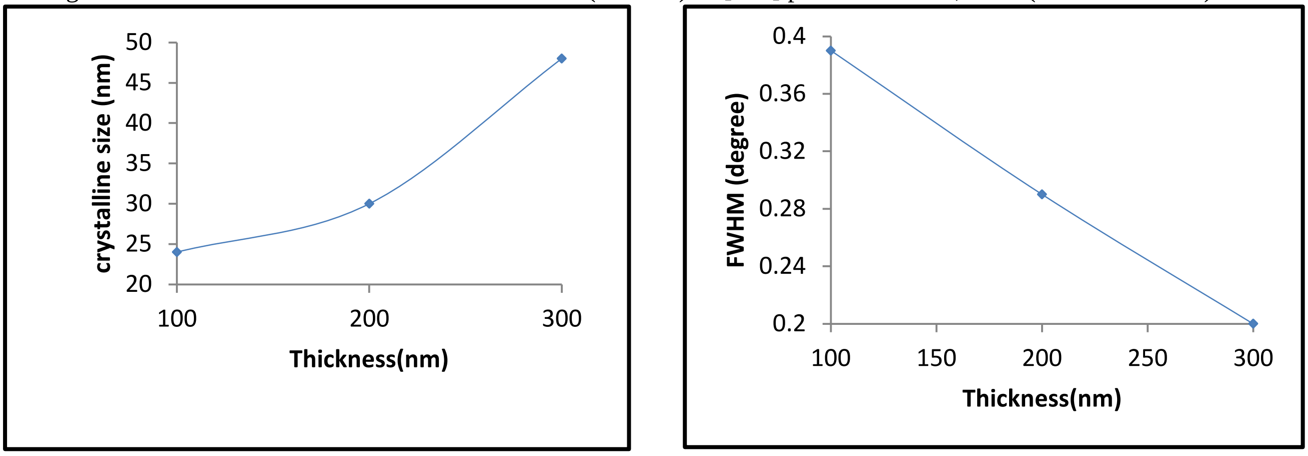
Fig 2a- The size of the crystal grains of PbS films with varies thickness in the plan [200] diffraction peak were 24.49-47.66 nm for films at thickness 100, 200,300 nm. Films increased when increase the thickness film But in **fig.2b** shows, When increasing in the thickness. The full width half maximum (FWHM) of [200] plane decreases; from (0.380 to 0.2040).



(a) **(b)**

Figure (2) a) grain size for the PbS films with varies thickness b) Full width at half maximum for the PbS films with varies thickness

In Fig (3), STM images for surface is gives the PbS thin films were clearly smooth. The calculate root mean square (RMS) of surface roughness of the PbS films in varies thickness 100,200,300 nm. Were 11.084, 11.448 and 12.551 nm for PbS films.

The fig (4) shows the larger grains size was caused by when increase of the RMS roughness with the increase of film thickness

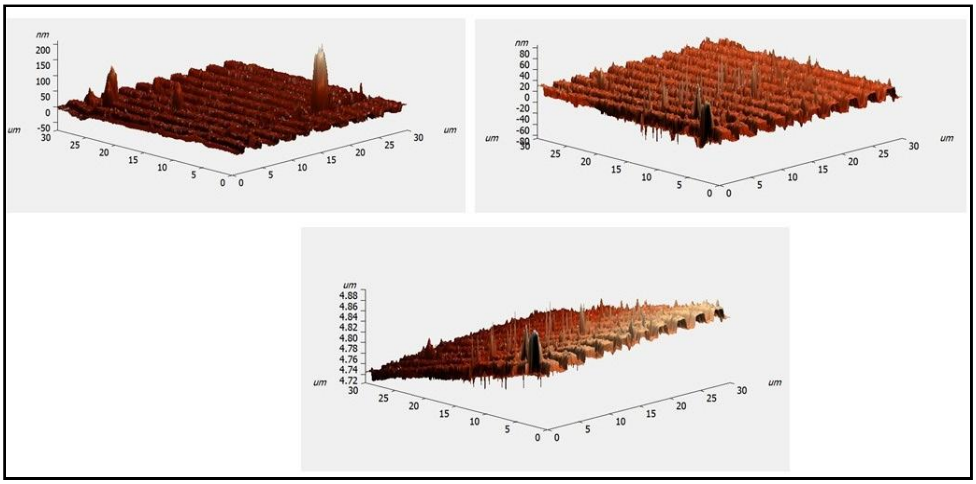


Figure (3) STM analysis of PbS films with varies thicknesses

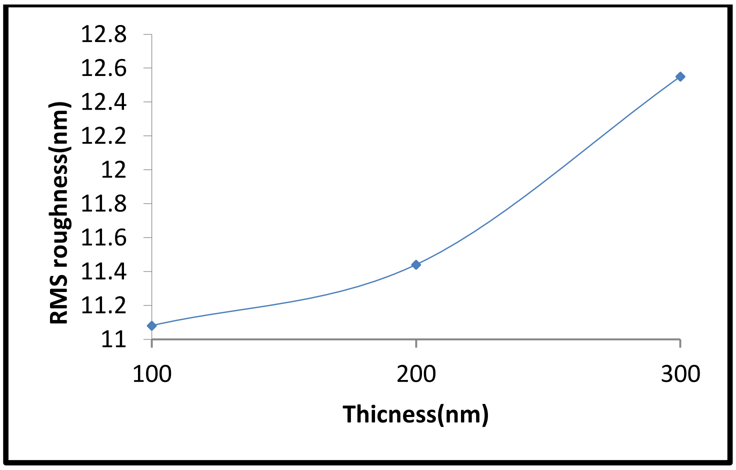


Figure (4) RMS roughness with varies thicknesses

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IV. ELECTRICAL PROPERTIES

Table (1) The Electrical Conductivity and Carrier Concentration with Thickness Films

Thickness nm	Conductivity $(\Omega.cm)^{-1}$	Carrier concentration (cm^{-3})	Conduction type
100	$0.0186*10^{-3}$	0.298×10^{11}	p
200	$0.4166*10^{-3}$	1.1×10^{11}	p
300	$0.9090*10^{-3}$	12×10^{11}	p

In electrical properties,When the increase in the thickness leads to increasing of the conductivity and the carrier concentration of the PbS film too. These can be described conductivity is extrusive proportional with carrier concentration as it is known. This results show the PbS films exhibit p-type conductivity

V. CONCLUSIONS

In this research spray pyrolysis method was used to deposing Lead Sulfide thin films (PbS) on glass substrates. A study of changing thickness effects on the structural and electrical properties of the films was done. X-ray patterns acquired proved the proper phase structure of the PbS films according to ASTM. The better orientation value of (200) plane has the highest value measure among other planes. Increasing the thickness film, the grain size of the PbS films increased, and the crystallite size in the range 24.49 - 47.66 nm. The root mean square roughness of film surface increased with the film thickness. About the electrical properties, the PbS films with diverse thickness always show p-type conductivity.

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