Study the Effect of Oblique Deposition on Surface Characterization and Microstructure of Evaporated Cadmium Thin Films

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ABSTRACT

The influence of oblique angle deposition on the surface morphological and microstructure of metallic cadmium films are studied. The films were deposited normally and obliquely at different angles (0˚, 50˚ and 70˚) by vacuum evaporation technique. XRD technique used to study the crystalline structure of these films and shows that the polycrystalline nature of these films and All layers irrespective of deposition parameters develop a preferred (002) plane. The grain size increased with the increase of deposition angle and clearly facetted morphology was observed. The surface morphology of the deposited materials has been studied using atomic force microscopes (AFM) and optical reflection microscope. The AFM results demonstrate that the film deposition at higher oblique angle (70°) has higher surface roughness. Reflection microscope results showed that the smoothness and homogeneity of the films are decreasing with increasing the deposition angle □□$_d$), and the surface roughness increased with increasing angle. The optical characteristics of the prepared thin films have been investigated by UV-VIS spectrophotometer in a wavelength ranging (350-900) nm and shows by increasing the inclination angle of deposition will lead to transmission decreases of the films.

Keywords: Glancing angle deposition, cadmium thin films, AFM, Morphology.

دراسة تأثير الترسيب المائل على تركيب وتشخيص أغشية الكادميوم المحضرة بالفراغ

الخلاصة

تم في هذا البحث دراسة تاثير زاوية الترسيب المائلة على التغير في الخصائص التركيبية لأغشية الكادميوم (Cd) المعدنية . الأغشية المحضرة رسبت بشكل منتظم (مستقيم) و بشكل مائل بزوايا مختلفة (0˚, 50˚, 70˚) بتقنية التبخير الحراري في الفراغ . تم استخدام تقنية حيود

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الاشعة السينية لتحديد الخصائص التركيبية , وأظهرت ان أغشية الكادميوم المحضرة ذات طبيعة متعددة التبلور وذات اتجاهية (002) . كما اوضحت الدراسة المجهرية بأن الحجم الحبيبي للأغشية المحضرة يزداد بزيادة زاوية الترسيب . اما طوبغرافية (طبيعة السطح) الأغشية المرسبة تم دراستها باستخدام مجهر القوة الذري (AFM) للتحليل السطحي بالاضافة الى استخدام المجهر البصري العاكس . وقد اظهرت نتائج مجهر القوة (AFM) ان التركيب السطحي للأغشية المرسبة يحصل فيه زيادة في خشونة السطح عند زيادة ميلان زاوية الترسيب الى (70°) . واتفقت هذه النتائج مع نتائج المجهر الضوئي بأن هناك نقصان في نعومة وإنتظامية سطح الغشاء مع زيادة زاوية الترسيب , وتزداد خشونة السطح مع زيادة زاوية الترسيب . كما شخصت الخصائص البصرية باستعمال مطياف UV-VIS عند مدى أطوال موجيه (350-900) نانومتر , بالاضافة إلى ذلك أظهرت الخصائص البصرية ان زيادة ميلان زاوية الترسيب سوف يؤدي الى نقصان نفاذية الأغشية المحضرة

الكلمات المرشدة : الترسيب المائل , أغشية الكادميوم ، مجهر القوة الذري ، طبيعة سطح الغشاء .

INTRODUCTION

Oblique angle deposition technique (also known as glancing angle deposition) has attracted the interest of many researchers, due to its ability to generate nanostructures relatively easily [1]. In addition to these complex morphologies, specific crystallographic phases can be selectively grown using oblique-angle deposition [1, 2]. Also oblique deposition produces columnar structures due to the shadowing effect and random fluctuations during film growth [3].

In a common oblique angle deposition system, a uniform deposition flux is obtained by evaporation techniques such as, thermal evaporation, sputtering, electron-beam evaporation, and laser beam ablation .The deposition flux approaches a stationary substrate at an angle (with respect to the substrate normal) referred to as the incident angle or the deposition angle (θ_d) [4]. By this technique it has been possible to produce zigzags, helices, vertical columns, capping layers and other film microstructures from various materials including metals, insulators and semiconductors [4]. The shadowing effect is the dominant mechanism controlling the formation, distribution and shape of 3D nanostructures in oblique angle deposition without substrate motion [5]. For a perfectly uniform deposition flux, which has no angular distribution, and if the deposition is on a smooth substrate, then the resulting nano-columns still can broaden along the direction perpendicular to the plane of deposition flux and substrate normal [6,7] . In this study, we employ atomic force microscopy (AFM) to investigate surface roughness parameters of Cd films prepared by vacuum glancing angle deposition technique. AFM images of prepared Cd films indicate changes in surface behavior of the film . Also, the results provide evidence that the oblique deposition greatly affect the final surface morphology of metallic Cadmium thin films .

EXPERIMENTAL DETAILS

Thin film deposition

Rectangular films of Cadmium material were grown from (Balzers BAE 080) evaporation system under vacuum of 2×10^{-5} torr on glass substrates . All metallic Cadmium pellets of 99.99% purity evaporation at room temperature using a molybdenum boat heater . Cd thin films were deposition at (θ_d = 0°, 50°, 70°) (the angle between the normal to the substrate and direction of incidence of the evaporated atoms). The source – substrate distance was 15cm, glass substrates which held at an angle to the direction of evaporation source. The thickness of the films, determined by using optical – interference method employing He-Ne laser (0.632μm) , this method depends on the interference of the laser beam reflected from thin film surface and substrate .

Characterization of the films

The film crystalline structure was investigated using standard X-ray diffraction system (LabX-XRD-6000/shimadzu) which has the following characteristics : source radiation of CuKα with (λ = 1.54060 Å) over the range of 2θ = 20° - 80° , scanning speed (5 degree/min) and incidence angle (20-70) degree . The crystallite size (grain size) (D) were calculated using the Scherrer formula from the full-width at half-maximum (FWHM) (Δ) [8]:

$$D=\left(\frac{0.94\lambda}{\Delta\cos\theta}\right) \qquad (1)$$

Where λ is wavelength of the X-rays and θ is Bragg angle.

The morphological information was obtained by atomic force microscope (AFM) and a reflection optical microscope .AFM studies were performed by (AA3000 scanning probe microscope)(Angstrom Advanced Inc.) in contact mode. All AFM images were acquired in ambient air, in constant force mode and digitized into (256 pixels × 256 pixels) also The image size of the surface morphology was (5×5 μm^2) . A variety of scans were acquired at random locations on the film surface. Reflection optical microscope study was performed by (Leitz-metallux3), provided with magnification of (40X) and contact with computer setting.

The optical measurements were done by measuring spectral transmittance of the samples in the wavelength range (λ = 350 – 900 nm) using Ultraviolet-Visible spectrophotometer (Metertech-SP 8001/UV-VIS spectrophotometer) .

RESULTS AND DISCUSSIONS

Thickness results

In thermal evaporation and by placing the substrate on spherical stand, the material deposition rate will be the same at all substrate regions in normal incident ($\theta_{deposition}$ = 0°), while in oblique incidence, when substrate normal makes an angle with the deposition direction, the amount of incoming particles which are captured by the substrate will be less than in the case of normal incidence, due to geometrical considerations [9, 10]. From the table (1). It is clear that the deposition film thickness decreases with increasing deposition angle. Therefore increasing the inclination angle of deposition will decreases the deposited materials per unit area.

X-Ray Diffraction Analysis

The XRD patterns for both normal (upright) and oblique depositions of metallic Cd thin films are shown in Figure (1). The representative XRD pattern recorded for Cd films deposition at ($\theta_d = 0^\circ$) normal deposition is presented in Figure (1*a*), whereas the XRD pattern for oblique angle deposition at ($\theta_d = 50^\circ$ and 70°) are shown in Figure (1b) and (c). One can observe that all analyzed samples shows polycrystalline nature. The narrow and strong peak localized at ($2\theta = 31.84^\circ$) corresponds to the reflection on (002) plane (preferred orientation) of hexagonal Cd structure and indicates that the grains in respective film are preponderantly oriented with their c-axis normally to the substrate surface as given in Figure(1*a*) [11], while in oblique deposition, the presence of these reflections indicates a high degree of crystallization with a preferred orientation perpendicular to the substrate normal. In addition to the low intensity peaks corresponding to (100) and (101) observed for normal and oblique grown.

The XRD spectrum of Cd thin film deposition by oblique angle deposition at ($\theta_d = 50^\circ$ and 70°) showing in a crystalline structure with strong peak position at ($2\theta = 31.72°$ and $31.78°$) corresponding to (002) plane . The peak intensity of normal incident is higher than that of oblique deposition due to the growth of the materials incorporated in the diffraction process , and this is also due to the film thickness in normal incident is larger than that in oblique deposition [8]. The coalescence of small grains into bigger ones is caused by the oblique deposition. From Figure (1b) and (c) it can be also observed that the peak intensity of the prominent diffraction peak decreases in oblique deposition at 50° and also shifts towards lower 2θ values. At 70° angle deposition, the crystalline of the film improves by increases the intensity of the prominent peak and shifts towards higher 2θ values as compared to corresponding 50° angle deposition [12]. However, in all cases the intensities of the (100) and (101) peaks were extremely low in comparison with the (002) one. This indicates a preferential orientation of the micro-crystallites with the (002) direction perpendicular to the substrate [8]. In general the oblique angle had great effect on the growth of polycrystalline cadmium, in particular for the higher oblique angles.

The FWHM was found to decrease markedly with increasing the inclination angle of deposition; such a decrease reflects the decrease in the concentration of lattice imperfections due to increase in the crystallite size [13]. Table (1) shows the variation of the grain size and film thickness with the deposition angle. It is clear from this Table that grain size for preferred orientation calculated from Scherrer equation increases with increasing deposition angle.

Reflection Optical Microscopy

Surface topography of Cd thin films is presented in Figure (1*a*). the surface of the normally deposited film is smooth while Pinholes and many voids are observed in oblique deposition see Figure (1b) and (c) respectively, which increases with raise the deposition angle. The smoothness and homogeneity of the films are decreasing with increasing the deposition angle $\theta_{d,}$ due to the columnar structure which contributes to a great extent in changing the film properties with respect to those of the corresponding bulk material [14]. These differences could be reasonably explained by an inclination of the columns and increasing of the void volume in samples prepared at high vapor incidence angles [4].

Atomic force microscope (AFM)

Atomic Force Microscopy (AFM) has been used to see the top surface morphology of the film from which we observed the formation of islands. It gives us information regarding the average size and size distribution of the islands and also provides information regarding the shape of the island [15]. AFM images indicate changes in surface behavior of the film. Also, the results provide evidence that the oblique deposition greatly affect the final surface morphology of metallic Cadmium thin films as can be shown in Figure(2) for scan area (5×5 μm^2). It can be seen from Figure (2*a***)**, a uniform and composed of agglomerates of very small grains for film as-deposition at ($\theta_d = 0^\circ$) (upright deposition), after increasing the deposition angle ($\theta_d = 50^\circ$ and 70°) as can be seen from Figure (2b) and (c) the agglomerates of nanograins were substituted by larger grains [13]. This agreement with XRD results shows previously.

In quantitative analyses on AFM images, it is known that the height roughness (R_a) and *rms* roughness have been used to describe the surface morphology (roughness). Roughness average (R_a) is defined as the mean value of the surface height relative to the center plane, and (R_{rms}) is the standard deviation of the surface height within the given area (has been calculated from the height distribution Figure(3) . From the results of AFM, found that the values of (R_a and Rrms) increased as the oblique angle deposition is increased. This behavior is due to the aggregation of the native grains into larger clusters [16]. The metallic Cd thin film deposition by oblique angle ($\theta_d = 70^\circ$), has larger clusters and becomes rougher. The above analyses specify that height roughness (R_a) and rms roughness (R_{rms}) are strongly affected by the degree of aggregation and cluster size of the films. The most popular parameter characterizing the morphology of surface is the (*rms*) roughness (R_{rms}) , which represents the root mean square height of a surface around its mean value [17].

A relatively uniformly distributed grain structure is observed from Figure (2). As can be see, two typical morphological features are recognized readily by visual inspection of Figure (2 and 3) the first feature is that the granules of various scales exist in all the films and are distributed evenly in some ranges. In addition, the granules possess different irregular shapes, sizes and separations. The second feature is that the evolution of the *rms* roughness as a function of oblique angle deposition up to 70°, where valleys, mountains and island clusters become bigger as the inclination angle of deposition increase [16].

the interesting results average grain size (D) , *rms* roughness (R_{rms}) and roughness average (R_a) have been listed in Table (1). It is clear that the film roughness increasing with increases deposition angle and this results it in good agreement with those in literature [4, 10]. This may be due to in columnar structure of films produce by oblique deposition technique accompanied by self shadowing effect [6, 10]. Also this may be due to the proportional of the grain size with film thickness [14]. In addition to the films produce by normal deposition reveals isotropic surface, while oblique deposition are more anisotropy. These results could be attributed to both deposition angle and film thickness. Normal deposition leads to a preference for mounds to coalesce along the direction perpendicular to the incoming beam [18].

Figure (3) show the height distribution (Granularity distribution) of AFM image of scan size 5×5 μm^2 .We observed the height distribution to be symmetric.

Table (1) . Characterizing parameter of the surface Cd thin film morphology.

angle deposition θ_d (degree)	Film thickness **t** (nm)	grain size ***D*** (nm)	FWHM Δ (deg)	rms roughness R_{rms} (nm)	height roughness R_a (nm)
0° normal	**300**	**20.87**	**0.413**	**21.9**	**12.8**
50° oblique	**240**	**23.17**	**o.372**	**25.4**	**20.5**
70° oblique	**170**	**26.43**	**0.326**	**32.4**	**25.9**

Optical results

Figure (4) Shows the transmission curves at room temperature of cadmium thin films deposited at different incident angles (θ_d = 0°, 50°, 70°). It is clear from figure that the transmission of the film deposition at normal incident (θ_d = 0°) in the region 400-900 nm was greater than after deposition at oblique incident (θ_d = 50°). Further higher oblique incident at (θ_d = 70°) showed the same trend indicated in Figure (4), the transmission was further decreased. It is clearly, that the increasing of the deposition angle has an obvious effect on the transmittance decreasing, and this is resulted from roughness increasing for film surface from obtained by increasing the columnar growth with needle and rod like shape [3, 4]. This structure increases the film trapping efficiency when rays falling on it and this reduce the transmittance.

CONCLUSIONS

In this work, we have performed a combined XRD – AFM techniques to investigate the different surface features of polycrystalline metallic cadmium thin films deposited on glass substrates by thermal evaporation process at different vapor incident angle. XRD analyzed shows that all layers irrespective of deposition parameters develop a preferred (002) plane and evaluated crystallite size increase from 20.87 to 26.43 nm with increasing deposition angle . The AFM images of the normal and oblique deposition films reveal the formation of a porous granular surface, while the surface roughness values are in the range between 21.9nm and 32.4nm which increase after inclination angle of deposition increase. Island formation of film was observed clearly for film deposition at (θ_d = 70°) from AFM images. These micro-structure properties are deeply affected after the films prepared by oblique angle deposition. Reflection microscope results showed that the smoothness and homogeneity of the films are decreasing with increasing the deposition angle. From transmission characteristics, it is clearly that the transmission is found to decrease with increasing the inclination angle of deposition.

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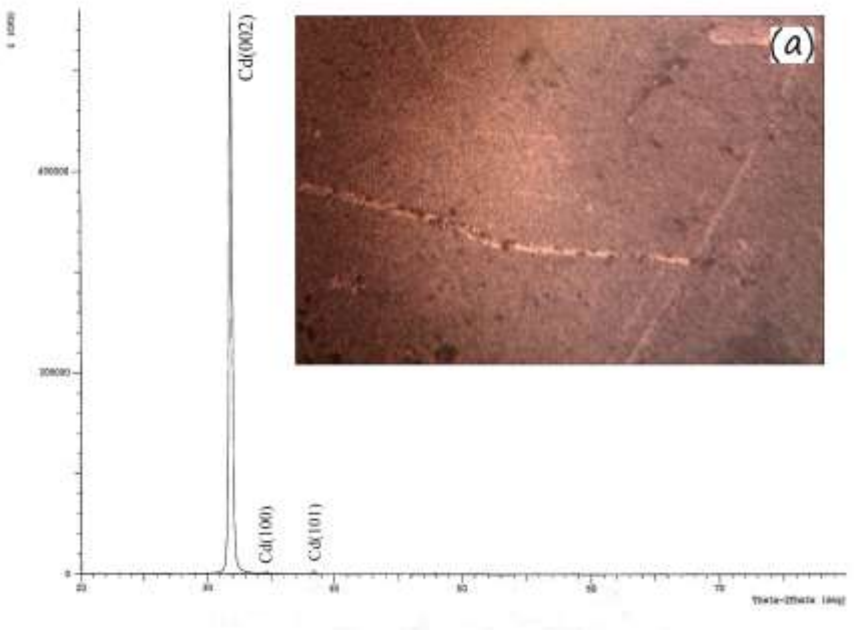
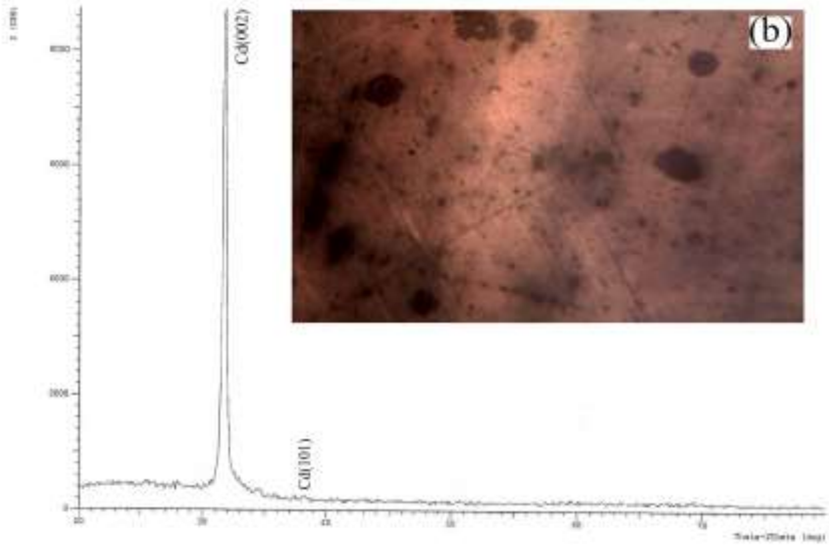
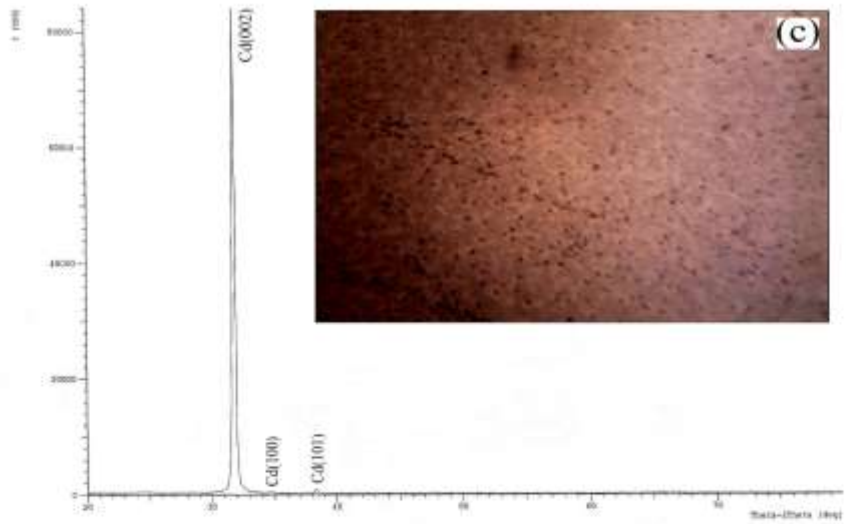



Cd(002)
Cd(101)
(b)


Cd(002)
Cd(100)
Cd(101)
(c)

Figure(1). XRD study and Optical reflection microscope images for Metallic Cadmium films deposition at different oblique angle deposition (a) normal incident at $\theta_d = 0^{\circ}$, (b) oblique deposited at $\theta_d = 50^{\circ}$, (c) oblique deposited at □$\theta_d = 70^{\circ}$.

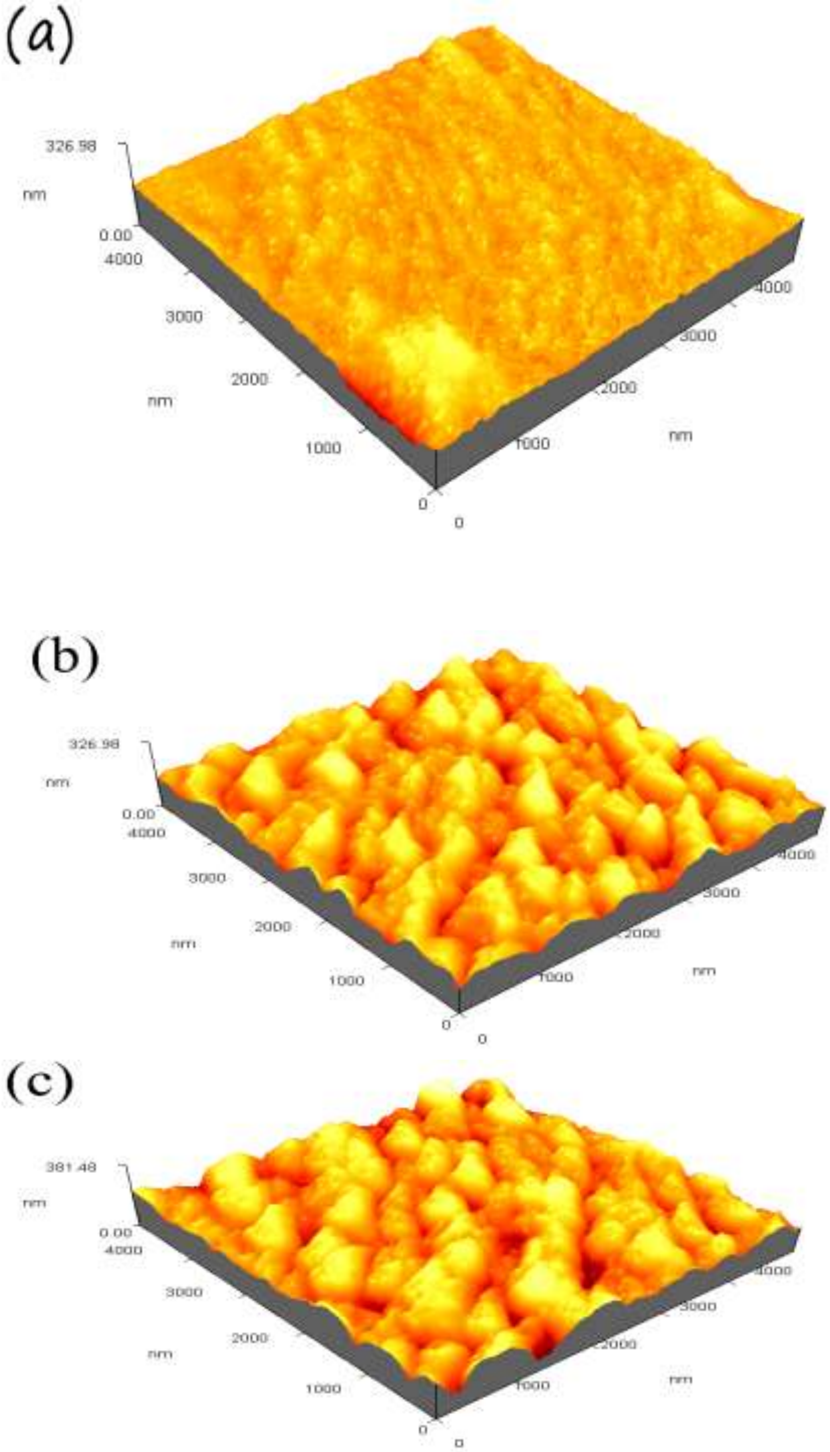
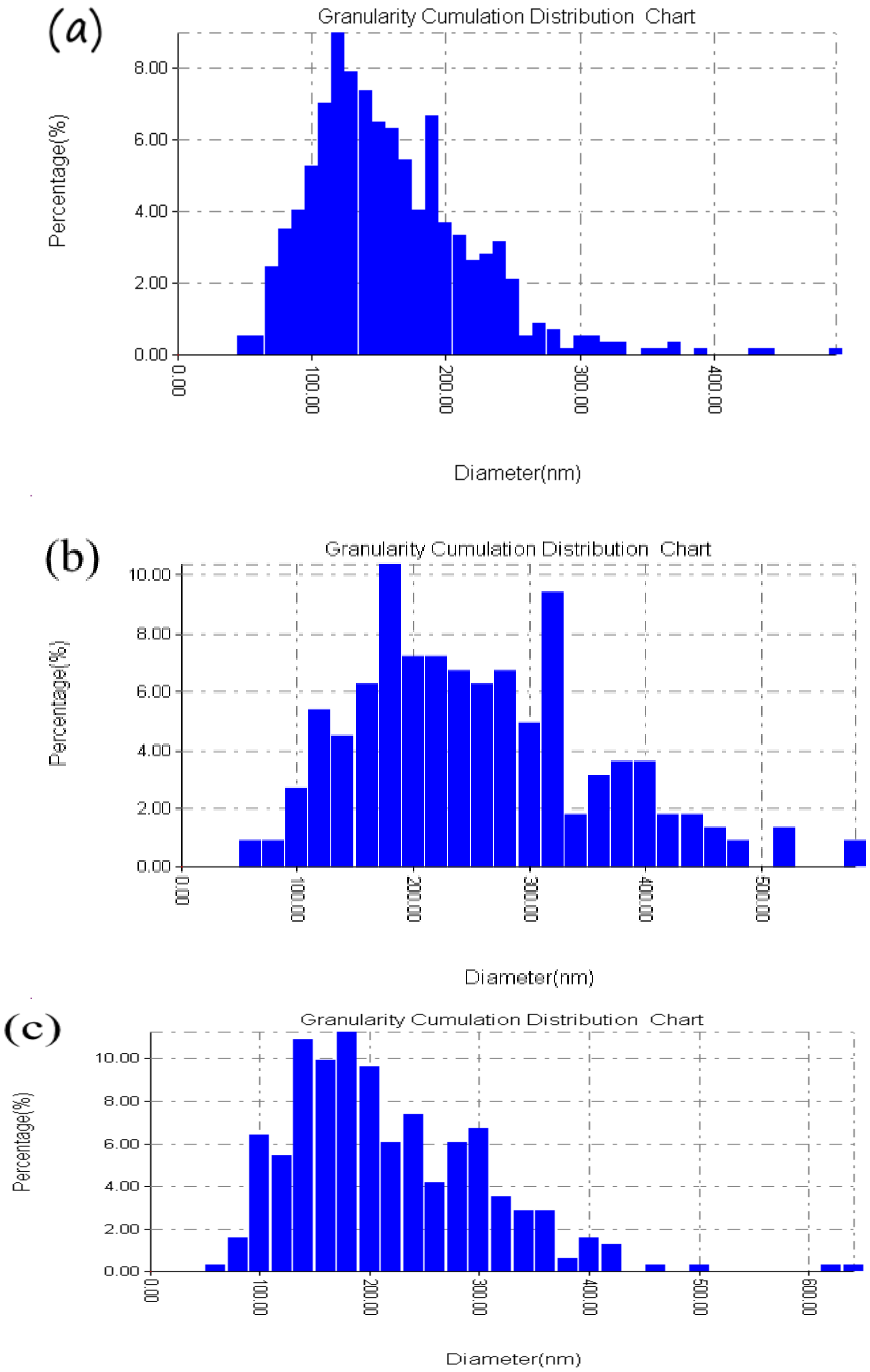


Figure (2): AFM surface topography of metallic Cd thin film , topography images shows the small- scale(5×5 □m^2) for
(a) normal incident at □$\theta_d = 0^\circ$) (upright deposition),
(b) (oblique angle deposition $\theta_d = 50^\circ$),
(c) (oblique angle deposition $\theta_d = 70^\circ$) .



Figure(3): The height distribution for the AFM images of Cadmium thin films . Scan area 5×5 μm^2 (a) as- deposited at $\theta_d = 0^\circ$,

(b) oblique deposited at $\theta_d = 50^\circ$,

(c) oblique deposited at $\theta_d = 70^{\circ}$.

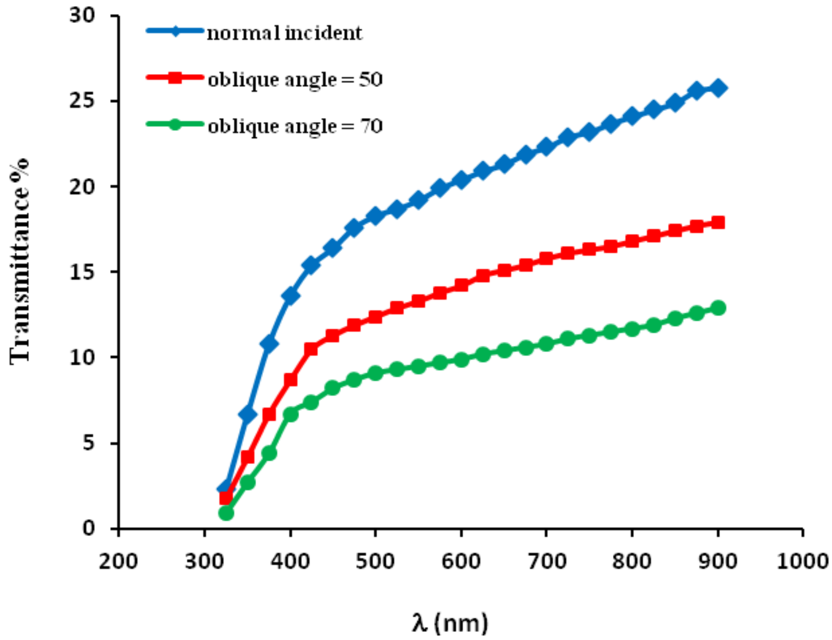


Figure (4). Transmittance spectrum of cadmium deposited at (0°, $50^{\circ}$and 70°) as a function of wavelength.