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ATOMIC FORCE MICROSCOPY STUDIES ON DC REACTIVE MAGNETRON SPUTTERED ZINC ALUMINUM OXIDE THIN FILMS

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Zinc Aluminum Oxide (ZAO) thin films were deposited on glass substrates by DC reactive magnetron sputtering technique with high purity individual Zn and Al targets. Surface roughness and grain size for all the samples increases with the increase of film thickness from 250 to 500 nm. The average roughness, maximum peak to valley height, root mean square (RMS) roughness, ten-point mean height roughness, surface skewness and surface kurtosis parameters are used to analyze the surface morphology of ZAO films. In the present work software-based image processing of AFM data provides quantitative information on ZAO nanostructured thin films. Statistics on groups of particles measured through image analysis and data processing includes particle counts, particle size distribution, surface area distribution and volume distribution.

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1. Introduction

Atomic force microscopy (AFM) is an excellent tool to study morphology and texture of diverse surfaces. The knowledge of the surface topography at nanometric resolution has made possible to probe dynamic biological process, tribological properties, mechanical manufacturing and mainly thin film surfaces [1]. The versatility of this technique allows meticulous observations and evaluations of the textural and morphological characteristics of the films, showing better facilities than other microscopic methods. The continued development of advanced optoelectronic devices requires the careful characterization of surface and microstructural features. However thin film structural details (i.e., grain size, crystallographic texture, and roughness) result from the atomistic processes such as surface diffusion, grain boundary motion, and depositing flux shadowing which operate during film growth. In addition it is possible that different evolutionary regimes of structure development may result from the variation of these atomistic mechanisms with processing conditions. Using adequate software, it is possible to evaluate characteristics such as roughness, porosity, average size, and particle size distribution, which influence directly the optical, mechanical, surface, magnetic and electrical properties of thin films. The properties of thin films depend sensitively on the surface morphology which in turn is determined by the growth process. From the perspective of applications, surface roughness is of much interest as it is directly related to optical scattering and resistivity due to grain boundary and thickness. Interfacial properties are also controlled by the surface structure. It is known that the roughness depends on the thickness of the film. As the thickness of a film increases, the roughness increases irrespective of the sample preparation procedure [2-4].

The study of surface morphology of the films with the variation in thickness and thermal processes gives an idea about the growth mechanism of these films[5–7]. Study of morphology and understanding of growth mechanism are essential to fabricate nanostructured materials in

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controlled way for desired properties. In fact, such systems are fully functional materials since their chemical and physical properties (catalytic, electronic, optical, mechanical, etc.) are strongly correlated with the structural ones (size, shape, crystallinity, etc.). As a consequence, the necessity to develop innovative bottom-up procedures (with respect to the traditional top-down scaling scheme) that allow the manipulation of the structural properties. Such studies find a renewed interest today for the potential nanotechnology applications[8]. Transparent and conductive Zinc Aluminum Oxide (ZAO) thin films have been widely used in various optoelectronic applications [9-11]. In our previous work we reported on microstructural, electrical and optical properties of ZAO thin films with different thickness [12]. The aim of the present work is to study the effect of film thickness on surface morphology, topography and texture of ZAO films by atomic force microscopy. A comprehensive analysis of ZAO film surface properties using parameters such as the average roughness (R_a), maximum peak to valley height (R_t), root mean square roughness (R_q), ten-point mean height roughness (R_z), surface skewness (R_{sk}) and surface kurtosis (R_{ku}) is made. These are parameters that allow insight into the surface properties and quality of the film.

2. Experimental details

ZAO thin films were prepared by DC reactive magnetron sputtering technique by using two individual high purity metallic targets of Zn and Al. The films were deposited on glass substrates with different thicknesses by varying the deposition rates. Table 1 shows the sputtering deposition conditions for ZAO films. The deposition conditions were optimized for ZAO films to exhibit a good surface roughness for light scattering and low resistivities for the development of high performance transparent electrodes for optoelectronic devices. Surface morphology, topography and texture of the samples have been studied using AFM (Park XE-100: Atomic Force Microscopy). The DC reactive magnetron sputtering process of ZAO thin films is shown in Fig. 1.

Table 1. Sputtering deposition parameters for ZAO thin films with different thicknesses

Sputtering target	:	99.999% pure Zn and 99.99 % pure Al (2 inch diameter and 4 mm thick)
Substrate	:	glass
Target–substrate distance	:	60 mm
Base pressure, p_u	:	4 x 10^{-4} Pa
Substrate temperature, T_s	:	RT
Sputtering pressure, p_w	:	0.5 Pa
Argon flow rate	:	30 sccm
Oxygen flow rate	:	2 sccm
Deposition time	:	25 – 30 min
Deposition rate	:	0.15 – 0.27 nm/s
Film thickness, t	:	250 - 500 nm
Sputtering power of Zn target	:	105 W
Sputtering power of Al target	:	40 W

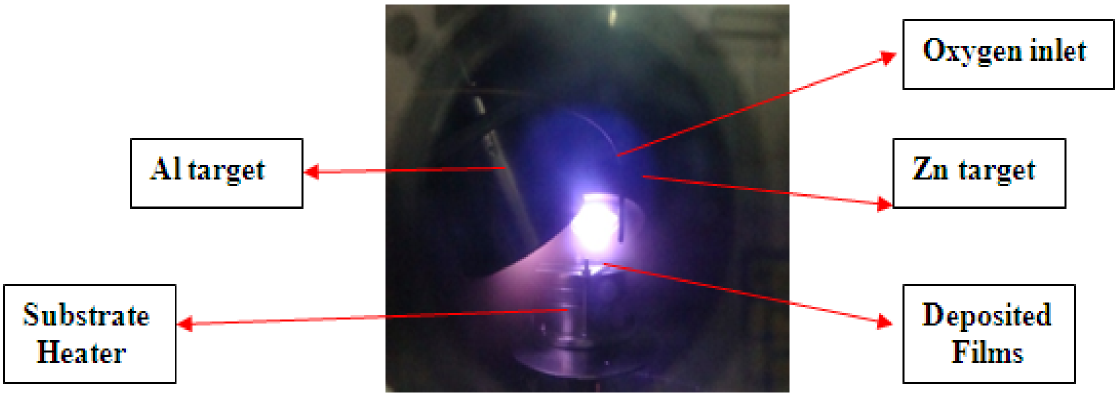

Fig. 1 DC reactive magnetron sputtering process of ZAO thin films

3. Results and discussion

Fig. 2 and 3 shows the two-dimensional (2D) and three-dimensional (3D) AFM images (Scan size 2 µm x 2 µm) of ZAO films with various thicknesses. The surface roughness of transparent conductive thin films has a significant influence on device performance [13]. Thus measuring the surface roughness of the films is important before the manufacturing of photoelectric devices. The surface roughness of the film increases from 14.58 nm to 19.86 nm with the increase of film thickness from 250 to 500 nm, it is more likely that charged oxygen-related species are adsorbed on the crystalline film. The surface morphology of ZAO films has the shape of hills and valleys. The valley region is relatively smooth, while the hill region consists of many crystal-like structures that exhibit certain orientations. As seen from the image of film thickness of 500 nm, the surface has been completely covered with several high regions as coniform structure and low regions as crater structure. The grain size distribution images for ZAO films of various thicknesses are shown in Fig. 4. The effects of grain size evolution during film deposition are evident on the variation of crystallographic texture and the RMS roughness with thickness. Although grain growth is still occurring, this roughening is due to shadowing of the arriving atomic flux leading to hillock growth and the increase in ridge height. The phase image can give better resolution as well as some information about material properties. In phase imaging, the phase lag of the tip relative to the excitation signal is monitored and recorded while the feedback keeps the amplitude at a fixed value. The change in phase is related not only to change of topography but also to the properties of surface. The phase images of ZAO films with different thickness are shown in Fig. 5. The different features can be identified as hillocks scattered randomly at average separations larger than the grain size, obvious grains and grain boundary grooves, and periodic ridge features with spacing smaller than the grain size but which extend uniformly over the surface of individual grains. The low frequency ''hillock'' roughness increases with thickness due to shadowing, resulting in hillock growth at a rate exceeding the average deposition rate. We assume that surface diffusion cannot effectively reduce the surface curvature at this length scale under these deposition conditions. The mean grain size places a lower limit on the size and spacing of hillocks, since hillocks are simply individual grains of height significantly greater than the film thickness.

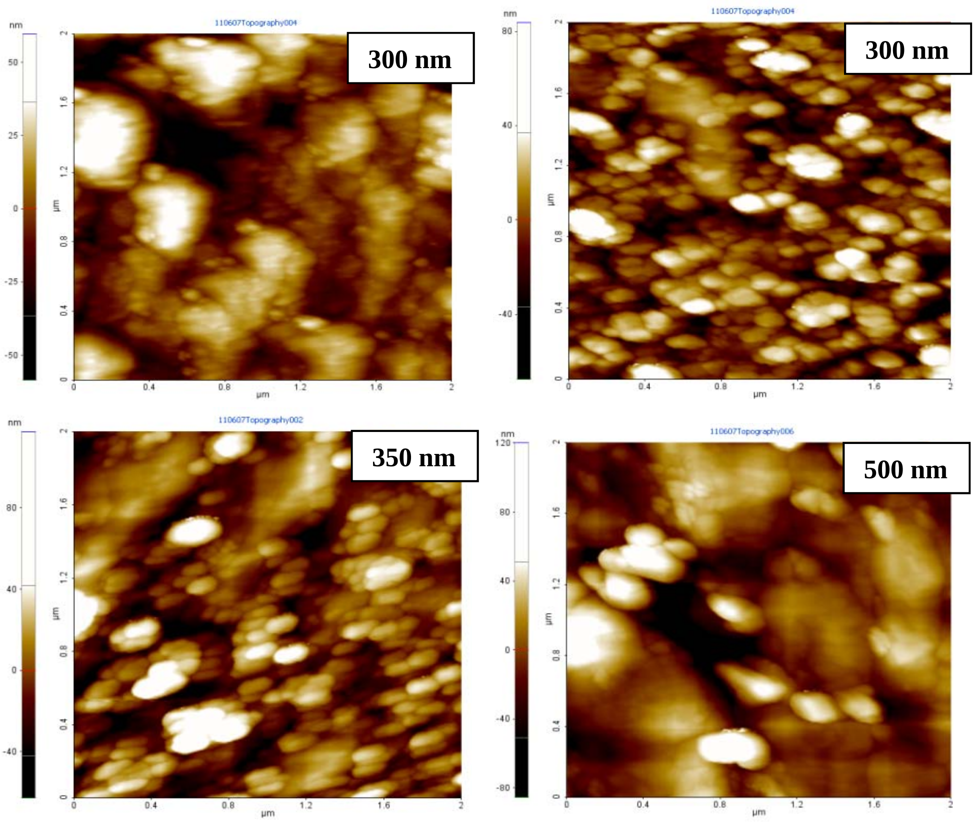

Fig.2 Topographical images (non-contact mode) of ZAO film of various thicknesses

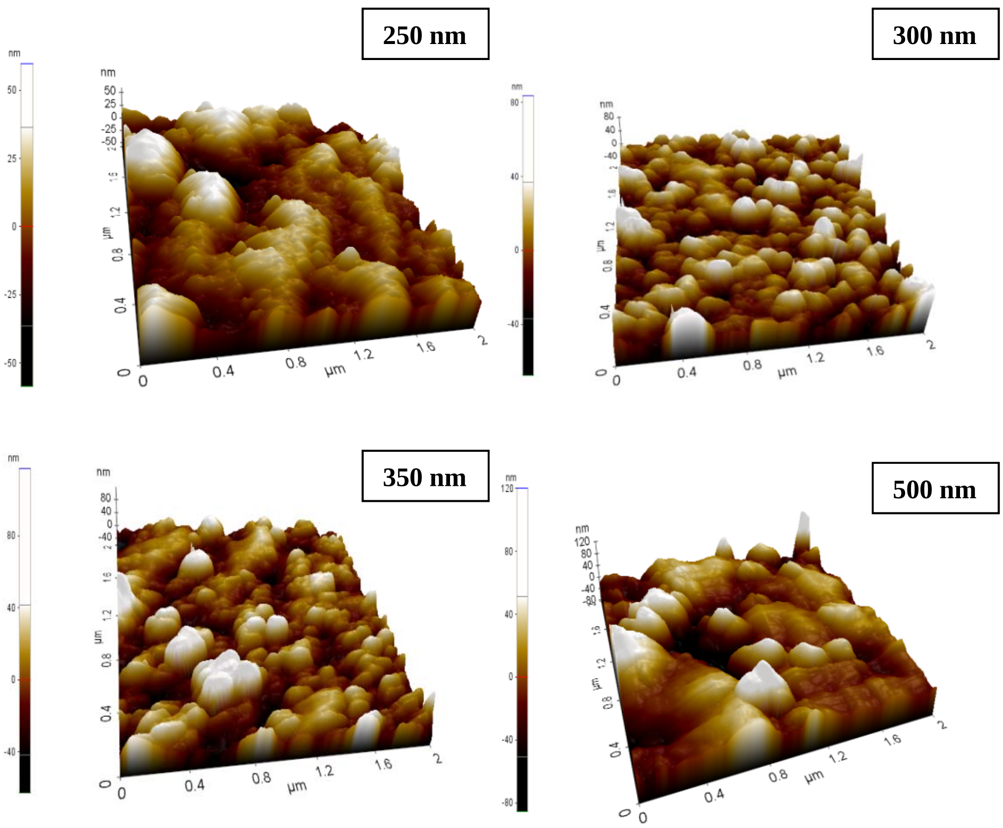

Fig. 3 AFM (3D) images of ZAO films with various thicknesses

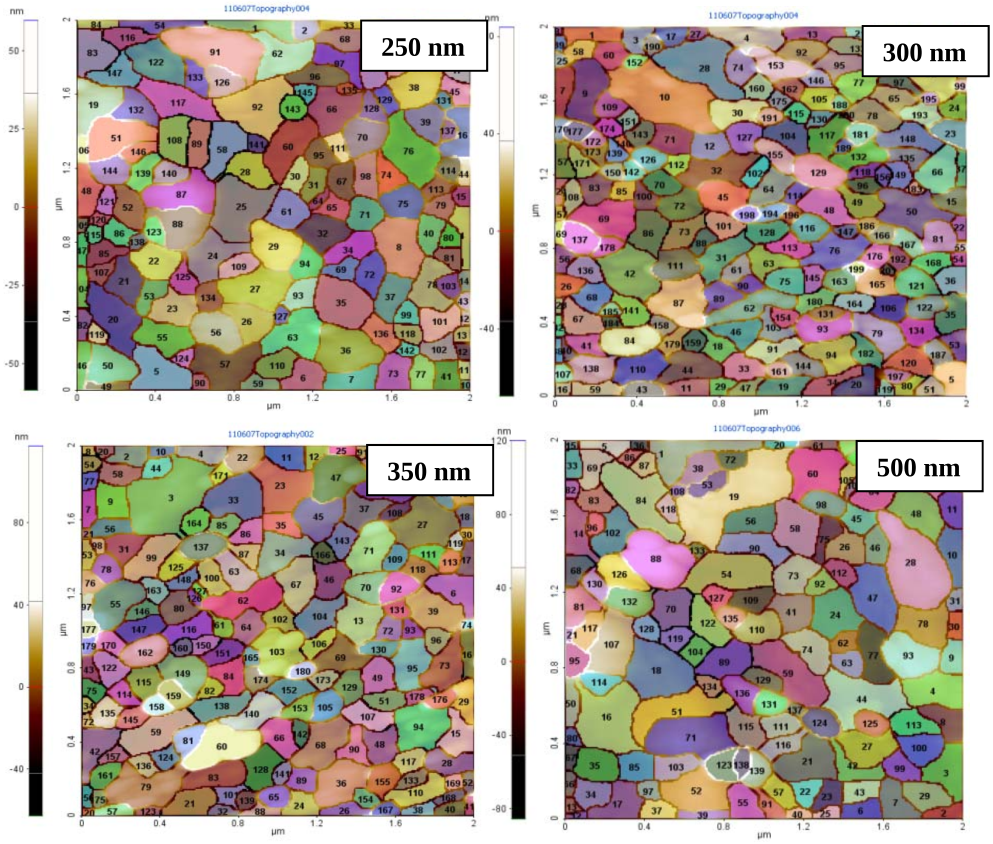

Fig.4 Grain size distribution images for ZAO thin films of various thickness

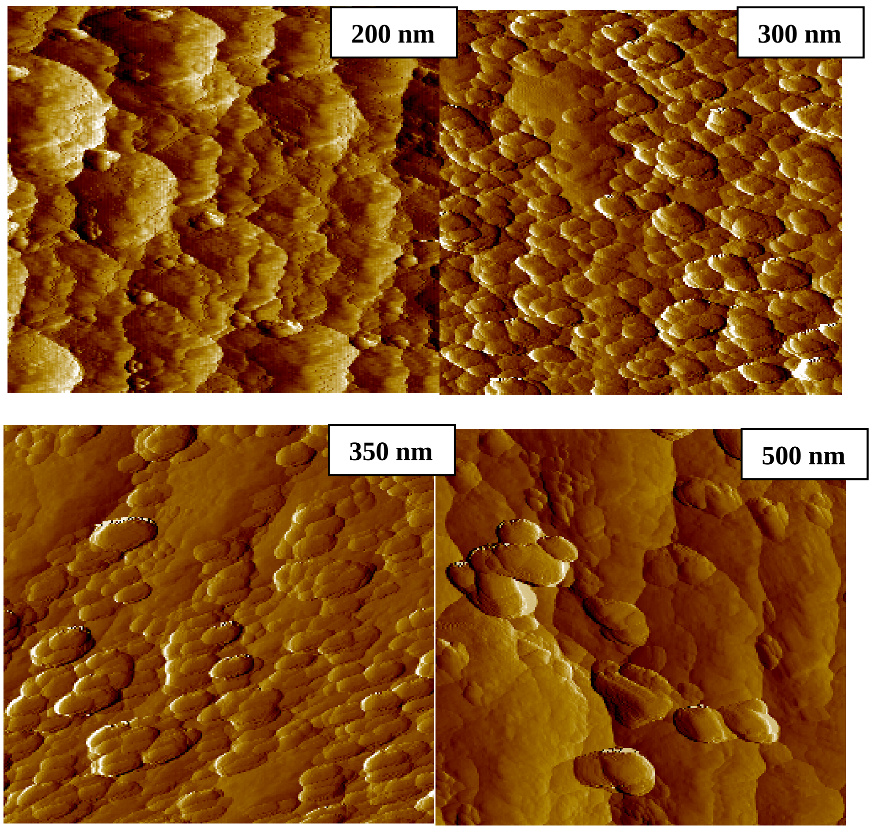

Fig.5 AFM phase images of ZAO thin films with different thicknesses

Fig. 6 represents the surface profile analysis of each thin film in the program carried out XEI's Park System. XEI program is used to convert acquired data into an image and to perform various analyses that meet the user's requirements. P[x] represents the horizontal momentum position. The surface profile parameters includes average roughness (R_a), root mean square roughness(R_q), maximum peak to valley height (R_t), ten point average roughness (R_z), skewness of the line (R_{sk}), kurtosis of the line (R_{ku}). Table 2 illustrates that the variations of the average roughness values (R_a) and ten-point mean height (R_z) values have the same trend as the variations of RMS roughness (R_q) values for all ZAO films with different thickness. The R_q values suggest relatively smooth surfaces and indicate two regimes of R_q variation with thickness. R_q is nearly constant for the film with thickness of 250 and 300 nm but it slightly increases monotonically for the films greater than 350 nm thick. Maximum peak to valley height (R_t) is also considered as a very important parameter because it gives a good description of the overall roughness of the surface. Table 2 shows that for high values of R_t, R_z is also high due to the strong dependence of R_z on the peak heights / valley depths. The RMS roughness values increases with the increase of film thickness from 250 nm to 500 nm due to the large grains as observed in the AFM micrographs. The larger values of the grain size also influenced the increasing RMS values of the samples. For the film of thickness 500 nm the surface was very flat and no very sharp peak appears in the domain. The increase of surface roughness for ZAO films under investigation agrees well with the increase in average grain size estimated from the SEM images [12]. It is important to note here that surface smoothness is a highly desired parameter for coatings that are used for optical applications because it reduces the reflection loss due to roughness-induced surface scattering.

The skewness values (R_{sk}) obtained from the ADF (Amplitude Distribution Function) analysis indicates the values of the average roughness R_a for both substrate and samples. The negative sign of R_{sk} for the substrate indicates that the surface morphology is a rough surface with flat regions and a low density of valleys or dips. For the film of thickness 350 nm the value of R_{sk} is consistent with a more symmetrical topography, both valleys and peaks have almost the same weight and the ADF is close to a gaussian distribution. This type of thin films synthesized for technological applications. Films with high values of R_{ku} have high R_t and R_z values as well. The film with thickness of 500 nm had high values of R_{ku} among the other films. ZAO films with different thickness had the value of R_{ku} more than three, the distribution will have relatively higher numbers of high peaks and low valleys with a spiky surface. In table 2 the negative values of skewness indicates cracks which is the representation of valleys.

Table 2. Roughness parameters of ZAO thin films with different thickness

Thickness (nm)	R_a nm	R_q nm	R_t nm	R_z nm	R_q/R_a	R_{sk}	R_{ku}
250	14.58	18.61	118.29	116.34	1.27	-0.09	3.01
300	14.94	18.81	151.33	136.36	1.26	-0.21	3.09
350	16.66	21.32	180.12	177.38	1.28	-0.38	3.08
500	19.86	21.04	205.75	194.39	1.06	-0.11	3.89

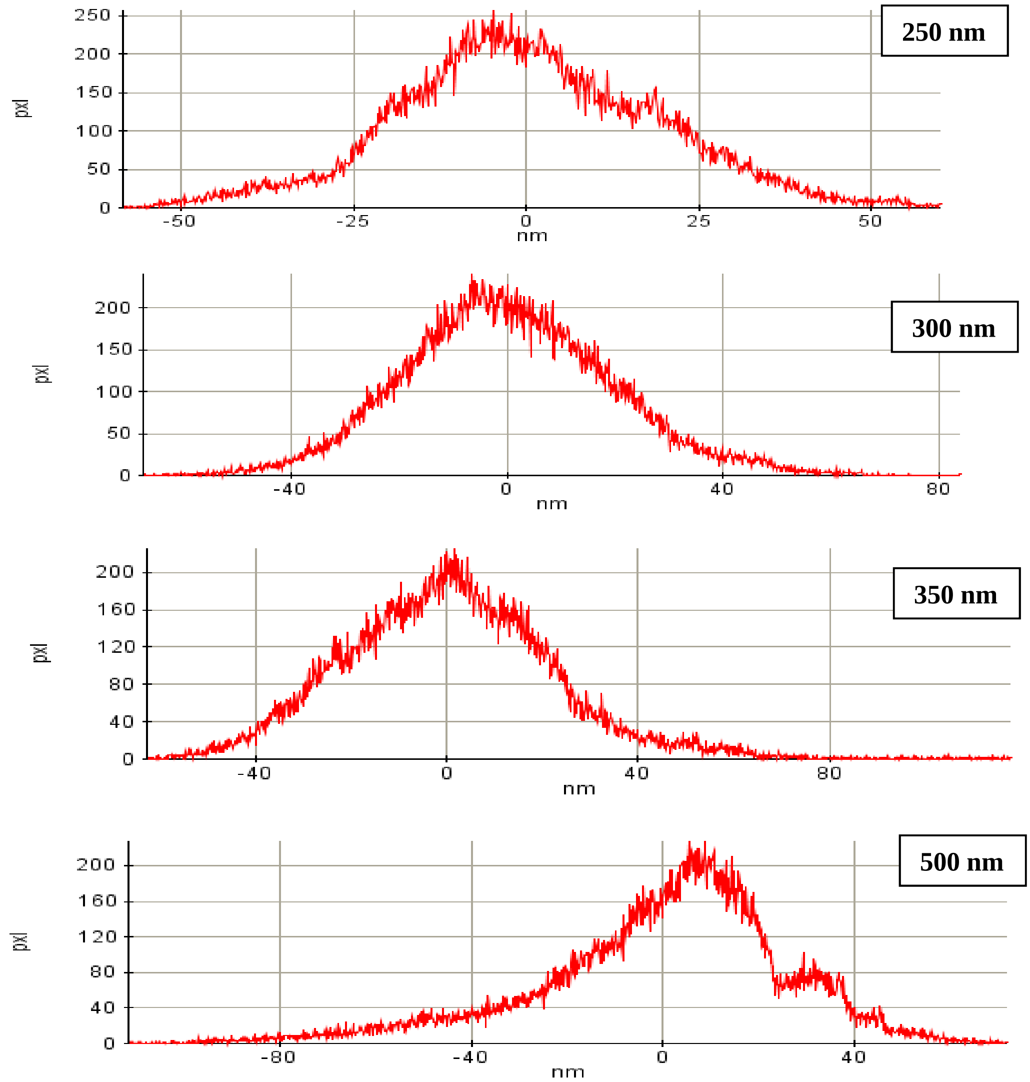

Fig. 6 AFM surface profile analysis of ZAO films with different thickness

Surfaces are binarized into grains and particles against a background. Binarization is carried out with respect to surface height/depth and bearing ratio. Software-based image processing of AFM data can generate quantitative information from individual grains or group of grains. Statistics on groups of particles can also be measured through image analysis and data processing. Commonly desired ensemble statistics include particle counts, particle size distribution, surface area distribution and volume distribution. For individual particles, size information (length, width, and height) and other physical properties (such as morphology and surface texture) can be measured. Quantitative analysis through histogram plots are shown in Fig. 7 for ZAO films with different thicknesses. Table 3 gives the statistical information on surface area, volume, length and perimeter distributions of nanostructured ZAO thin films.

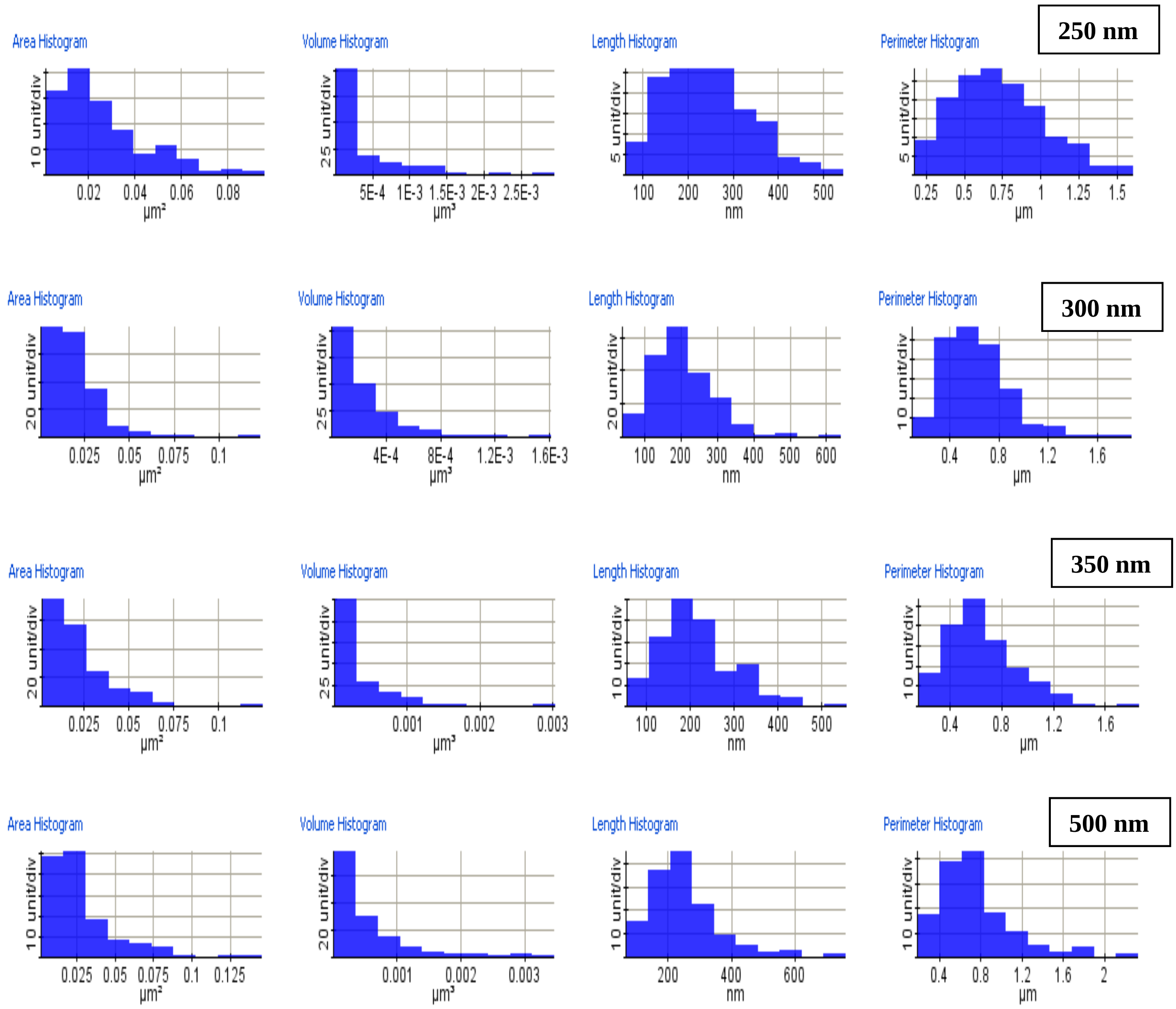

Fig. 7 Quantitative analysis of nanostructured ZAO thin films of various thicknesses

Table 3. Statistical information on area, volume, length and perimeter distributions of nanostructured ZAO thin films

Thickness (nm)	Grain	Area (μm^2) X 10^{-2}	Volume (μm^3) X 10^{-4}	Length (nm)	Perimeter (μm)
250	Mean	2.56	3.56	241.7	0.72
	Std.	2.84	4.53	94.6	0.30
300	Mean	1.84	2.36	202.2	0.61
	Std.	1.31	2.57	84.3	0.26
350	Mean	2.07	3.01	212	0.64
	Std.	1.64	3.6	83.2	0.27
500	Mean	2.71	5.02	247.8	0.24
	Std.	2.33	6.17	112.4	0.35

4. Conclusions

Atomic force microscopy (AFM) was employed to monitor surface morphologies of ZAO thin films prepared on glass substrates by DC reactive magnetron sputtering method. The topography of the films with thickness from 250 to 500 nm had waviness surface texture. This type of study provides a more comprehensive understanding of the influence of the obtaining conditions on morphological features of the films and could help in tailoring the deposition parameters according to surface morphology requirements for an optoelectronic device application.

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