

AFM STUDIES ON SURFACE MORPHOLOGY, TOPOGRAPHY AND TEXTURE OF NANOSTRUCTURED 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 Zn and Al targets. After the deposition, the films were annealed in vacuum from 200 to 500 °C. Surface roughness measurement of ZAO thin films in the nanometer scale can be accurately determined using the atomic force microscopy. Surface roughness and grain size for all the samples shows a direct relation between each other where the surface roughness increases as the grain size increases with increase of annealing temperature. 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.

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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 made possible to probe dynamic biological process [1], tribological properties [2, 3], mechanical manufacturing [4] and mainly thin film surfaces [5, 6]. 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. A repetitive or random deviation from the nominal surface forms the pattern of the surface. Surface texture includes roughness, waviness, lay and flaws. 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.

Transparent and conductive Zinc Aluminum Oxide (ZAO) films have been widely used in various optoelectronic applications [7-9]. It is well known that surface roughness of transparent conductive thin films have a significant influence on device performance [10]. In most papers, the film deposition was carried using ceramic oxide targets which is high cost and requires maximum power density. Therefore the deposition rates are limited due to the tolerable thermal load for the targets. Significant cost reductions are expected by using reactive sputtering of metallic targets. In this study we report for the first time the influence of annealing temperature on the morphological properties of ZAO films prepared by DC reactive magnetron sputtering technique using two individual high purity metallic targets of Zn and Al. 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 electrochromic device.

The aim of the present work is to study the effect of post-annealing temperature treatment on surface morphology, topography and texture of ZAO films by atomic force microscopy. A

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comprehensive analysis of ZAO film surface properties using parameters such as the average roughness, maximum peak to valley height, root mean square roughness, ten-point mean height roughness, surface skewness and surface kurtosis is made. These are parameters that allow insight into the surface properties and quality.

2. Experimental details

ZAO thin films were prepared by DC reactive magnetron sputtering technique. High purity metal targets of Zn (99.999%) and Al (99.99%) with 2 inch diameter and 4 mm thickness are used for deposition on glass substrates. The glass substrates were ultrasonically cleaned in acetone and ethanol, rinsed in an ultrasonic bath in deionised water for 15 min, with subsequent drying in an oven before deposition. The sputter chamber is initially evacuated to a base pressure of 6×10^{-6} Torr and then pure argon gas is introduced into the chamber at a flow rate of 25 sccm (standard cubic centimetres per minute) through mass flow controller (Model GFC 17, Aalborg, Germany). Pure oxygen is let into the chamber through another mass flow controller with oxygen flow rates of 2 sccm. Depositions were carried out at a working pressure of 3 mTorr after pre-sputtering with argon for 10 min. The distance between the target and substrate is kept at 60 mm and the deposition time is 30 min. The sputtering power of 110 W is applied for Zn target and 42 W for Al target. The deposited films were annealed in vacuum from 200 °C to 500 °C. Film thickness was measured by Talysurf profilometer. The resulting thicknesses of the films are ~ 350 nm. Surface morphology of the samples has 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.

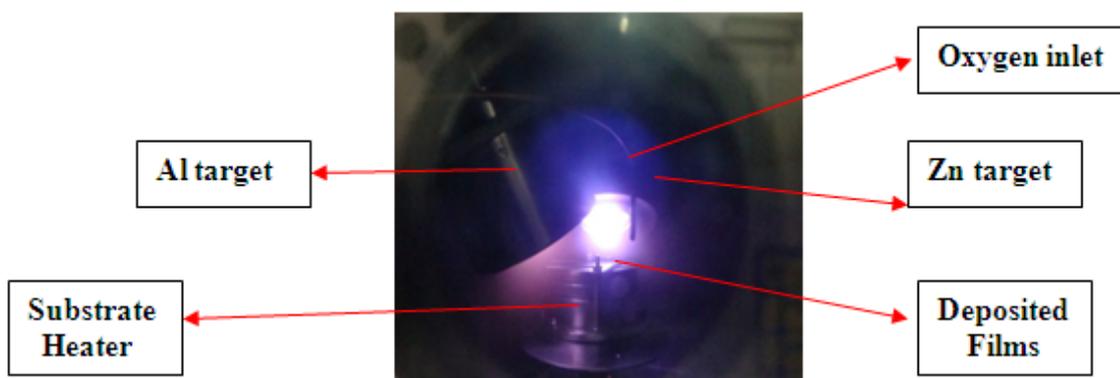


Fig. 1 DC reactive magnetron sputtering process of ZAO thin films

3. Results and discussion

Atomic force microscopic analysis is ideal for quantitatively measuring the nanometric dimensional surface roughness and for visualizing the surface nano-texture of the deposited film. Measuring the surface texture of ZAO thin films with horizontal length scale of 2 microns and a vertical length scale of 50 nm is critical for optoelectronic applications. Two-dimensional (2D) and three - dimensional (3D) AFM images of ZAO films annealed from 200 to 500 °C is shown in Fig. 2. The surface roughness of ZAO films increased from 14.52 to 25.07 nm as the annealing temperature increased from 200 to 500 °C. The surface of ZAO films has the shape of the hills and valleys. As the surface roughness increases, it is more likely that charged oxygen-related species are adsorbed on the crystalline film. Lin et al. [11] described that the high temperature can stimulate the migration of grain boundaries and cause the coalescence of more grains during the annealing processes. Fang et al. [12] further indicated at high temperature, more energy should be available for the atoms to acquire so that they may diffuse and occupy the correct site in the crystal lattice and grains with lower surface energy will grow larger at high temperature. The major grain

growth also yields an increase in the surface roughness as shown by previous studies [13, 14]. The results show that ZAO thin films exhibits waviness surface texture. The amplitude and phase images of ZAO thin films annealed at different temperatures are shown in Fig. 3. The “valley” region is relatively smooth, while the “hill” region consists of many crystal-like structures that exhibit certain orientations.

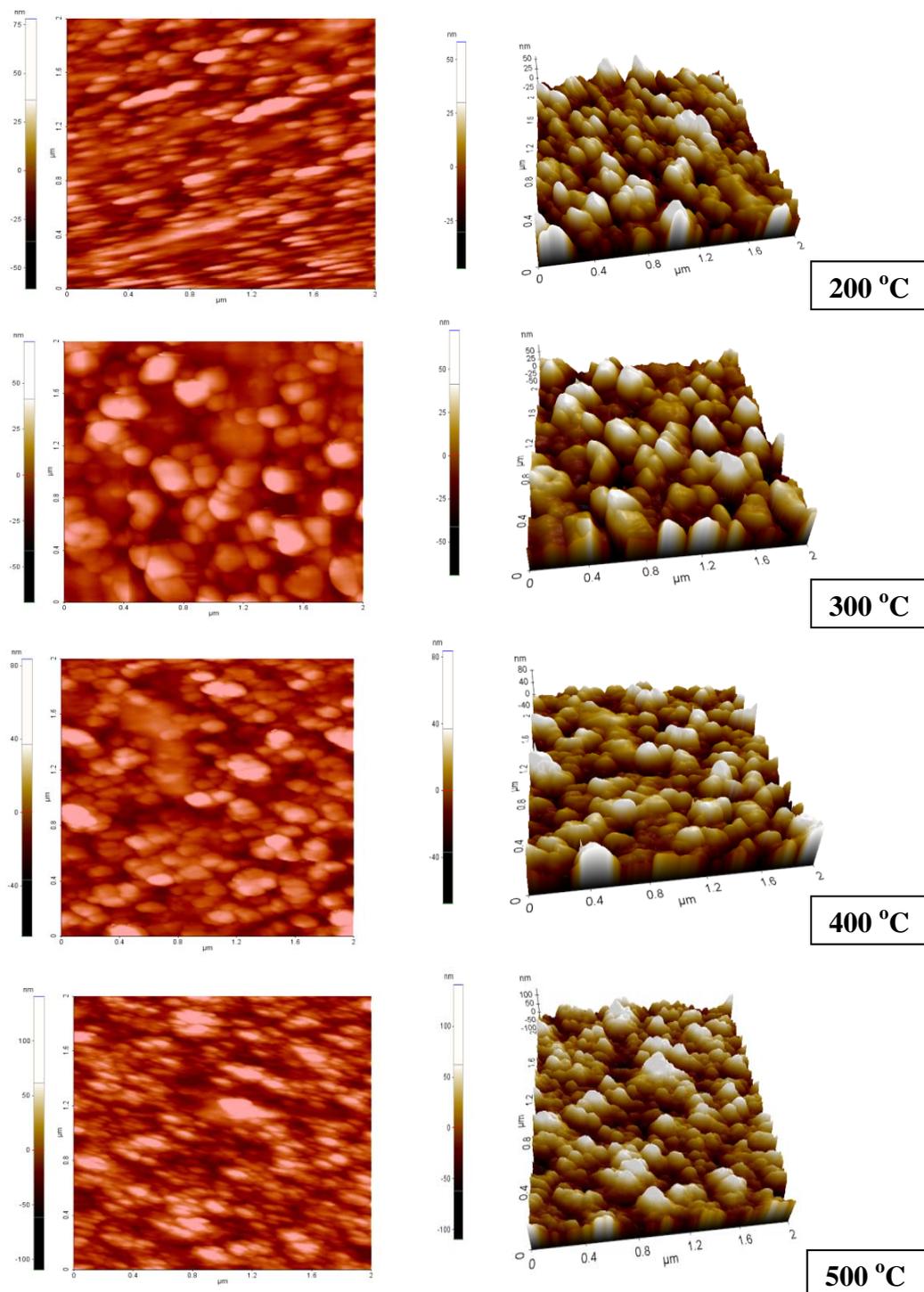


Fig. 2 AFM (2D and 3D) images of ZAO films annealed at different temperatures

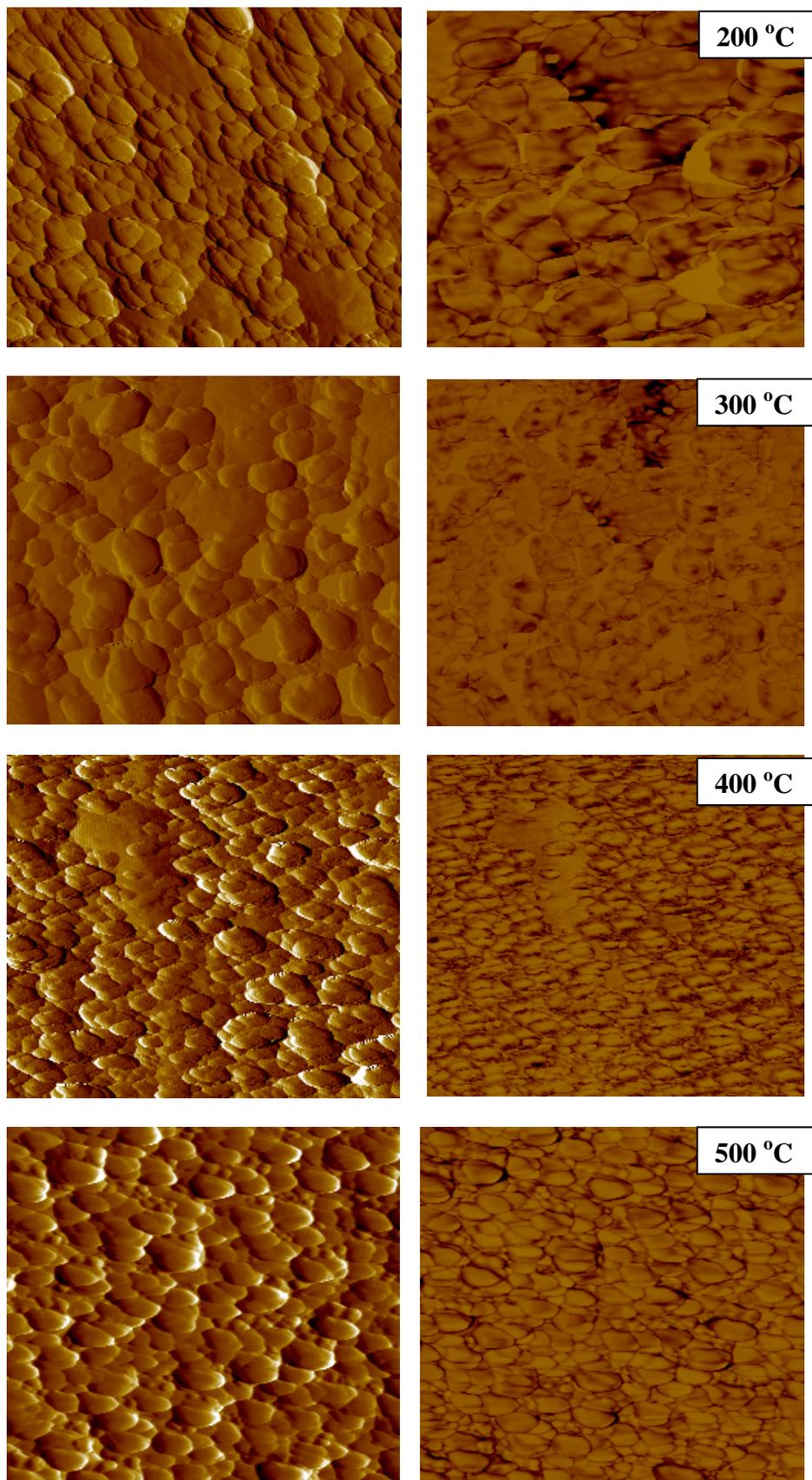


Fig. 3 AFM amplitude and phase images (Scan size $2\ \mu\text{m} \times 2\ \mu\text{m}$) obtained from the “hill” and the “valley” of ZAO thin films annealed at different temperatures.

Fig. 4(a) - 4(d) represent an analysis of the roughness 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. Surface roughness was characterized by calculating the roughness parameters R_q , R_a and maximum elevation. The surface roughness of transparent conductive thin films has a significant influence on device performance [15]. Thus measuring the surface roughness of ZAO films is important before the manufacturing of photoelectric devices. The roughness parameters are estimated by the analyzing the topography scans of the sample's surface. 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}).

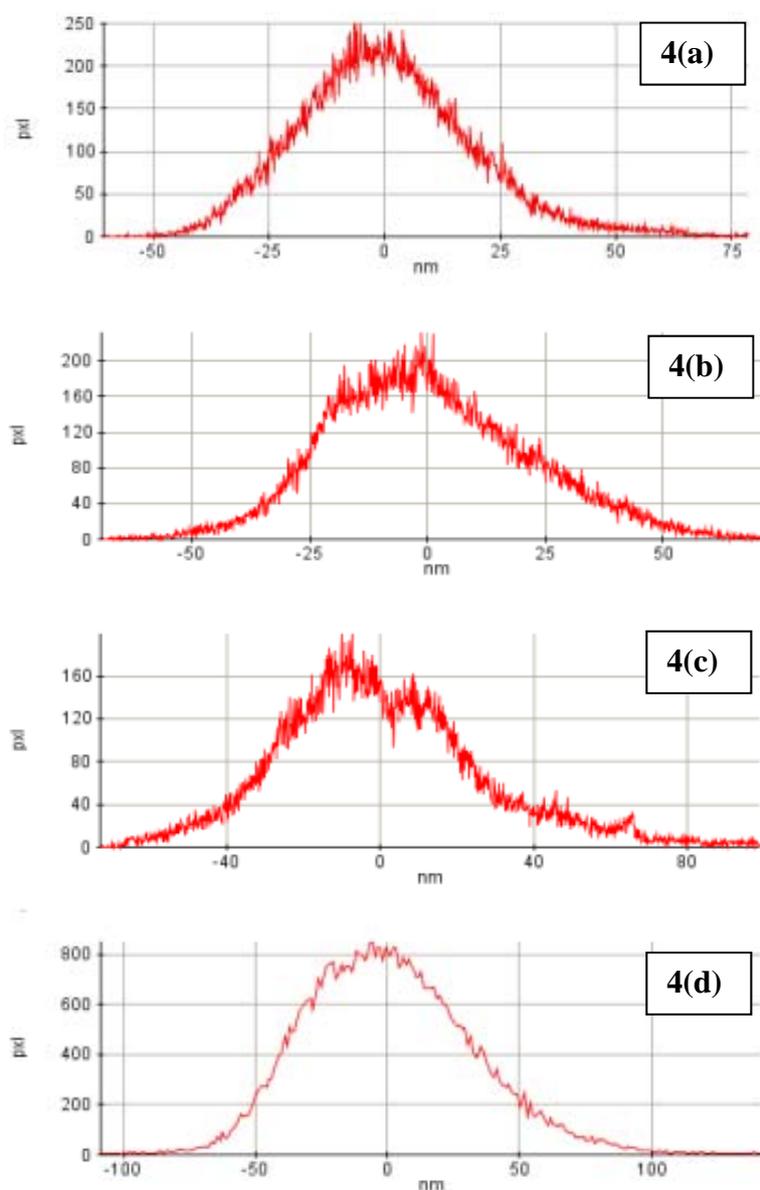


Fig. 4 AFM surface profile analysis of ZAO films annealed at a) 200 °C, b) 300 °C, c) 400 °C and d) 500 °C

The amplitude parameters of a sample are described by parameters which give information about statistical average values, shape of the histogram heights and other extreme properties. The average roughness (R_a) is the mean height as calculated over the entire measured length/area. R_a is typically used to describe the roughness of machined surfaces. It is useful for detecting general variations in overall profile height characteristics and for monitoring an established manufacturing process. Maximum peak to valley height roughness (R_t) is the vertical distance between the highest and lowest points in the evaluated length/area and describes the overall roughness of the surface. Root mean square (RMS) roughness (R_q) is the square root of the distribution of surface height and is considered to be more sensitive than the average roughness for large deviations from the mean line/plane and is also used in computing the skew and kurtosis parameters. RMS roughness (R_q) describes the finish of optical surfaces. It represents the standard deviation of the profile heights and is used in computations of skew and kurtosis. Ten-points mean height roughness (R_z) is the difference in height between the average of five highest peaks and five lowest valleys in the evaluation profile/surface and is more sensitive to occasional high peaks or deep valleys than R_z . Roughness skewness (R_{sk}) is used to measure the symmetry of the variations of a profile/surface about the mean line/plane and is more sensitive to occasional deep valleys or high peaks. R_{sk} illustrates load carrying capacity, porosity, and characteristics of nonconventional machining processes. Negative skew is a criterion for a good bearing surface. Usually, R_{sk} is used to distinguish two profiles of the same R_a or R_q values but of different shapes. Kurtosis is a measure of the distribution of spikes above and below the mean line. Kurtosis describes machined surfaces and is rarely used for optical surfaces. It is sometimes specified for the control of stress fracture. Roughness kurtosis (R_{ku}) is used to measure the distribution of the spikes above and below the mean line/plane. For spiky surfaces, $R_{ku} > 3$; for bumpy surfaces, $R_{ku} < 3$; perfectly random surfaces have kurtosis 3.

Table 1 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 annealed ZAO films. Maximum peak to valley height (R_t) is also considered a very important parameter because it gives a good description of the overall roughness of the surface. The Table 1 also 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. R_z is mathematically given by the following formula [16]:

$$R_z = \frac{1}{n} (\sum_{i=1}^n P_i - \sum_{i=1}^n V_i) \quad (1)$$

where n is the number of sampling points along the assessment length, which is 5 in this study, P_i is the height of the i^{th} peak and V_i is the depth of the i^{th} valley with respect to the line profile. In addition, R_q values for all the samples are higher than R_a values, which can be mathematically explained according to the following equation:

$$R_z = \frac{1}{L} \int_0^L |y(x)| \cdot dx \quad (2)$$

$$R_q = \sqrt{\frac{1}{L} \int_0^L |(y(x))^2| \cdot dx} \quad (3)$$

where L is the length of the profile on the x -axis used for measurement and $y(x)$ is the variation of the height from the profile line for each data point.

For a Gaussian distribution of asperity height, statistical theory shows that the ratio of R_q to R_a should be 1.25. Ward [17] notes that the asperity height distribution of most engineering surfaces (tribology) may be approximated by a Gaussian distribution with R_q/R_a values of up to 1.31. As shown in Table 1, the values of R_q/R_a using data collected from AFM imaging are reasonably close to the value of 1.25 predicted by theory. This result is significant since it indicates that, at the imaging scale, the asperity height distribution of these surfaces are approximately Gaussian and that the statistical relationships for surface roughness are applicable. In Table 1, negative values of the skewness indicate that the valleys are dominant over the scanned

area and positive values show that the peaks are dominant on the surface. Continued negative values would indicate cracks, representative of valleys. The distribution of positive and negative values indicates the existence of protruding grains. For ZAO film annealed at 300 °C the value of R_{ku} is below three, which shows that the distribution over the scanned area has relatively few high peaks and low valleys with a bumpy surface. The films annealed at 200 °C, 400 °C and 500 °C 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. Films with high R_{ku} values have high R_t and R_z values as well. This is due to the strong relation between these parameters, where R_{ku} is mathematically directly related to the peak heights and valley depths according to the following formula:

$$R_{ku} = \frac{1}{NR_q^2} (\sum_{i=1}^N Y_i^4) \quad (4)$$

Where R_{ku} is the RMS roughness parameter and Y is the height of the profile at point number i .

Table 1. Roughness parameters of ZAO thin films annealed at different temperatures

Annealing Temperature (°C)	R_a nm	R_q nm	R_t nm	R_z nm	R_q/R_a	R_{sk}	R_{ku}
200	14.52	18.51	57.05	133.06	1.275	-0.44	3.47
300	16.87	21.12	44.52	140.33	1.252	-0.30	2.96
400	20.65	26.57	44.19	170.06	1.286	-0.55	3.60
500	25.07	31.52	86.02	243.86	1.257	-0.40	3.31

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. 5(a) – 5(d) for ZAO films annealed at different temperatures. Table 2 gives the statistical information on surface area, volume, length and perimeter distributions of nanostructured ZAO thin films.

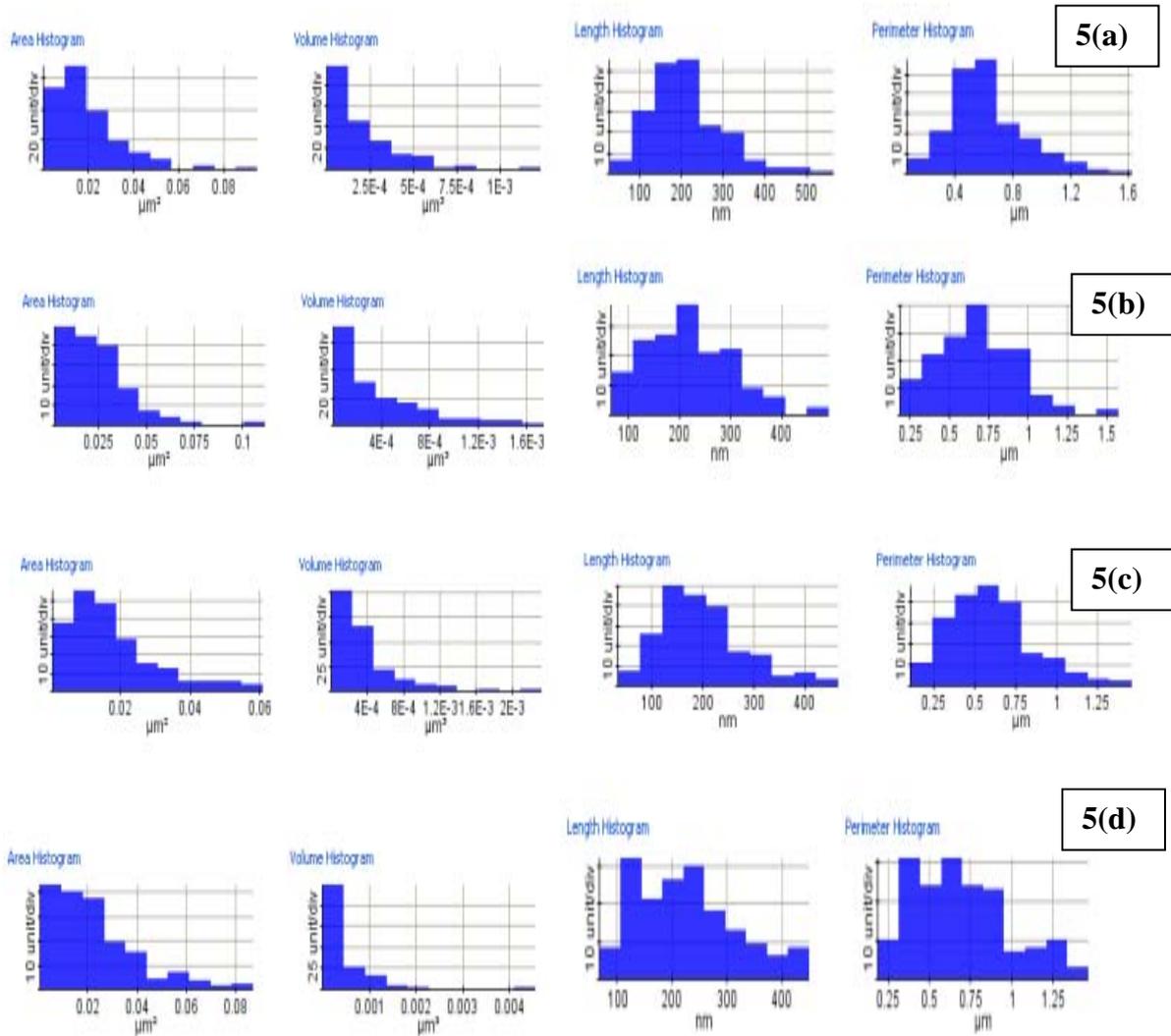


Fig. 5 Quantitative analysis of nanostructured ZAO thin films annealed at a) 200 °C, b) 300 °C, c) 400 °C and d) 500 °C

Table 2. Statistical information on area, volume, length and perimeter distributions of nanostructured ZAO thin films.

Annealing Temperature (°C)	Area (μm^2) $\times 10^{-2}$	Volume (μm^3) $\times 10^{-4}$	Length (nm)	Perimeter (μm)
200	1.88	1.90	208.27	0.621
300	2.30	3.57	219.05	0.678
400	2.18	4.23	223.60	0.686
500	1.73	3.37	196.82	0.597

4. Conclusions

Atomic force microscopy (AFM) was employed to monitor surface morphologies of annealed ZAO thin films prepared on glass substrates by DC reactive magnetron sputtering method. The AFM topography of annealed films from 200 to 500 °C showed that the film surface is having waviness surface texture. A significant difference occurs at annealing temperature of 500 °C, where the film surface exhibits a higher roughness and clear grains were observed. 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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