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Nanotribology and fractal analysis of ZnO thin films using scanning probe microscopy

Te-Hua Fang[1,3], Sheng-Rui Jian[2] and Der-San Chuu[2]

[1] Department of Mechanical Engineering, Southern Taiwan University of Technology, Tainan 710, Taiwan
[2] Institute and Department of Electrophysics, National Chiao Tung University, Hsinchu 300, Taiwan

E-mail: fang@mail.stut.edu.tw

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Abstract
Characteristics of crystalline structure, roughness and nanotribology of ZnO thin films deposited under various power conditions were achieved by means of x-ray diffraction and scanning probe microscopy (SPM). The ZnO thin films were deposited on the silicon (100) substrates by a radio frequency magnetron sputtering system. Fractal analysis was derived from SPM images, to calculate the fractal dimension complexity of the surface geometry, via a substituting structure function. The results show that the roughness decreases and nanowear rate increases as the sputtering power increases. In addition, the fractal dimensions of the ZnO thin films are also presented.

1. Introduction

Zinc oxide (ZnO) thin films have been attracting lots of attention for their technologically important application in the use of manufacturing transparent electrodes, piezoelectric devices, nonlinear electrical devices and surface acoustic wave devices (SAW) [1–5] due to their direct wide-band-gap of 3.4 eV, high transparency and piezoelectric properties, which play an important role in the field of advanced nanoscience.

The nanomechanical characterizations of a surface is significant today because the characterizations may be different for those bulk material surfaces that have had different treatments. The nanomechanical properties of films can be adjusted using accurate techniques to design the films. It then becomes possible to establish a specialized processing window on which the film characteristics can be optimized. On the whole, application success is also partly determined by the accuracy of our understanding of these characterizations. Measurement of the nanomechanical properties of the materials and correlation with the microstructures are very essential in recognizing their tribological behaviour [6].

By means of atomic force microscopy (AFM), it is possible to find out the surface topography with a spatial resolution that is measured on the atomic scale. Nevertheless, it is difficult to quantify the surface roughness and surface morphology in view of the fact that it is hard to demonstrate the surface mathematically. The mathematical description of a surface should accurately reflect its features, and should be in agreement with the various theoretical models that relate to surface structure. The ZnO film surface topography may be described in terms of its fractal geometry, which is a part of modern mathematics that uses fractional dimensions to describe disordered objects. Fractal analysis reveals a variety of objects, including self-affine or self-similar, i.e. objects that look similar at all levels of magnification. The concept of fractal dimension was used to explore the surface morphologies of ZnO thin films generated under various sputtering powers. The fractal dimensions were estimated by applying the structure function method [7, 8].

Consequently, the aim of this paper is to provide insight into the nanotribological wear and friction characteristics

[3] Author to whom correspondence should be addressed.

and fractal analysis of ZnO thin films deposited at various sputtering powers. The films are deposited on silicon (100) substrate by a radio frequency (RF) magnetron sputtering system. The microstructure properties of ZnO thin films are characterized by x-ray diffraction (XRD) and scanning probe microscopy (SPM) which is used to investigate the friction and wear characteristics of the deposited films on the nanoscale. Furthermore, the fractal dimension analysis at various sputtering powers is also presented.

2. Experimental details

ZnO thin films are produced on silicon substrate by RF magnetron sputtering deposition [9–11], which are deposited at different sputtering powers. The chamber is first evacuated to 10^{-6} Pa and then the oxygen/argon gas mixture is introduced for film deposition. The deposition rates range from 20.2 to 20.7 (nm min^{-1}) during the sputtering process and the thickness of deposited films 2.4–2.5 μm. The separation between the substrate and target is 14 cm. The ZnO thin films growth conditions are listed in table 1. A rectangular $20 \times 20\,\mu m^2$ area of the specimen surface is scanned at low resolution to verify the presence of acceptable surface conditions. If the surface is acceptable then the selected area is contracted to a $2 \times 2\,\mu m^2$ area for friction and wear experiments. In addition, the average surface roughness, R_a, of the current sample is recorded for comparison with the test areas of subsequent samples in the experiment. This is to ensure that samples with comparable initial surface conditions are used in all of the experiments. The pre-scan procedures for nanofriction, nanowear and fractal analysis are similar. For the fractal analysis part, the Shimadzu fractal software was adopted to describe the surface roughness in our experiments.

The crystal structure of ZnO thin films were analysed by XRD (D/MAX-RA Rigaku using CuKα, $\lambda = 1.5418$ Å) patterns. A SPM (Shimadzu SPM-9500J2) apparatus is used to measure the topographic properties of the specimens. AFM scanning uses only the two vertical quadrants to measure vertical deflection of the cantilever thus showing the surface profile of the specimen. Friction force microscopy (FFM) scanning however, uses the other two horizontal quadrants. It is noted that both topographic and friction measurements can be performed simultaneously. A constant scan speed of $2\,\mu m\,s^{-1}$ is used and a constant load of 100 nN is applied to the cantilever. Two different cantilevers of distinct stiffness are used in the experiments. The stiffer one is used in the wear experiments in order to obtain larger applied loads. The less stiff one is used for surface topographic measurements, in order to gain higher sensitivity. The measurement tip is made of Si_3N_4 with a cantilever of lower stiffness and thus yielding measurements of greater sensitivity.

Table 1. Growth conditions of the ZnO thin films.

Substrate	Si
Target	99.99% Zn
Gas	$O_2/Ar = 0.3$
Deposition pressure	0.3 Pa
Deposition time	120 min
Substrate temperature	200˚C
Power range	150 W, 175 W, 200 W, 225 W
Frequency	13.56 MHz

3. Results and discussion

3.1. Crystallographic characterizations

XRD has become the main tool to analyse the crystalline structures for piezoelectric films. In other words, the piezoelectric characterizations are dependant on the degree of the crystalline structure orientation [12]. Figure 1 shows the XRD patterns of ZnO films deposited on Si(100) substrate at various sputtering powers with strong *c*-axis (002) orientations. In terms of XRD data, it is possible to observe that an increase in the sputtering power manifests a clear orientation of the structural properties of ZnO thin films, in addition to that, the angular peak position of deposited films with a (002) orientation is located at $2\theta = 34.4^\circ$. As the sputtering power decreases, ZnO(002)-diffraction peak of the samples weakens and another ZnO diffraction peak becomes intensified. Similar results can be seen in previous studies [9, 12].

The Scherrer formula [13] was adopted to estimate the mean grain size of deposited films from the measured width of their diffraction curves. The mean grain sizes of the films were 33.64 nm, 37.38 nm, 40.05 nm and 42.06 nm which were deposited at the sputtering powers of 150 W, 175 W, 200 W and 225 W, respectively. The results showed that at the highest sputtering power at 225 W the grain size was the largest and had a more pronounced preferential orientation.

3.2. Morphology and nanofriction tests

In this paper a SPM apparatus is used to analyse the surface asperity of ZnO thin films at different sputtering powers. Island-like growths in the films are clearly observed in all the AFM and FFM images. The lighter area in figure 2 represents those areas with a higher height. Figures 2(*a*)–(*d*)

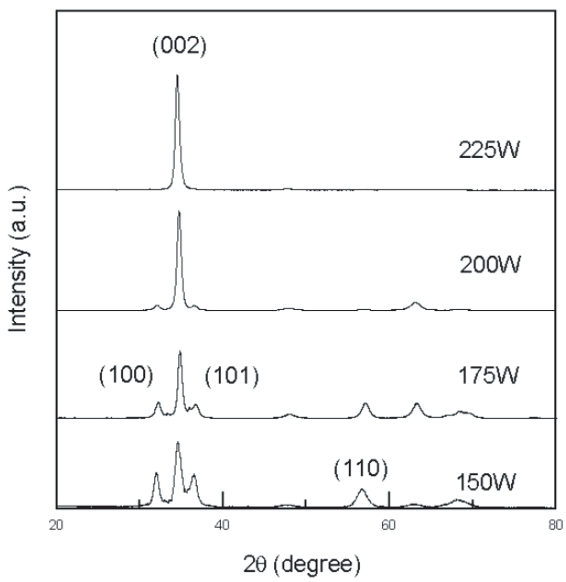

Figure 1. XRD patterns of ZnO thin films at various sputtering powers.

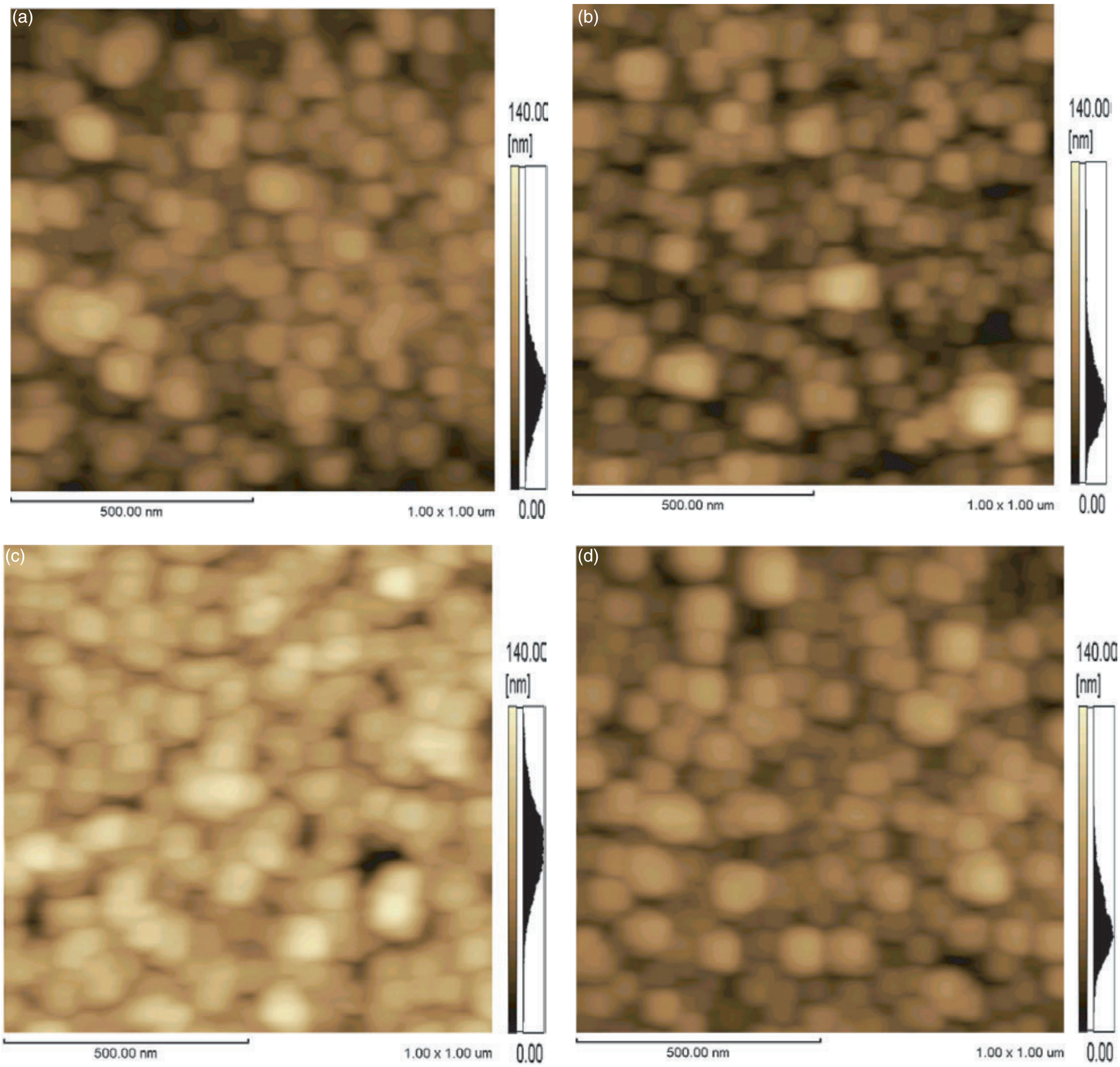

Figure 2. AFM images of ZnO thin films deposited on Si(100) substrate at the various sputtering powers of: (*a*) 150 W, (*b*) 175 W, (*c*) 200 W, (*d*) 225 W.

and 3(*a*)–(*d*) show the surface topographies and friction contour plots for four different sputtering powers. Comparison of figures 2(*d*) and 3(*d*) shows similar patterns of discrete areas with, however, inverse colouring. Where a light area is found in figure 2(*d*), indicating greater height, a dark area is found in figure 3(*d*), indicating greater friction. These indicate that on nanometer-scale there is a strong dependence between the slope of the surface asperity and the friction coefficient. Further discussion of this subject can be found in the thesis of Ruan and Bhushan [14–16], which investigated the influence of different surface asperity morphologies of the friction coefficient. It is well known that the height roughness parameters R_a and RMS have been used to describe the surface morphology as part of the quantitative analysis of AFM images. R_a is defined as the mean value of the surface height relative to the centre plane and RMS is the root-mean-square average roughness profile of the surface height within the given area [17]. These results are also shown in figure 4. It can be seen in figure 4 that the surface becomes smoother at higher sputtering powers.

The friction coefficient is the average ratio of the friction to the normal load, or when the slope is not zero it is the slope of the curve generated by the plotting friction force and normal load. In this study four experiments are carried out to obtain the friction coefficient. Figure 5 illustrates the relationship of the friction force and friction coefficient at the four different sputtering powers. Based upon the above-mentioned results, it would appear that the height roughness and friction coefficient decrease under the influence of a higher sputtering power. In other words, the above analysis indicates that height roughness R_a and RMS are strongly affected by their structures.

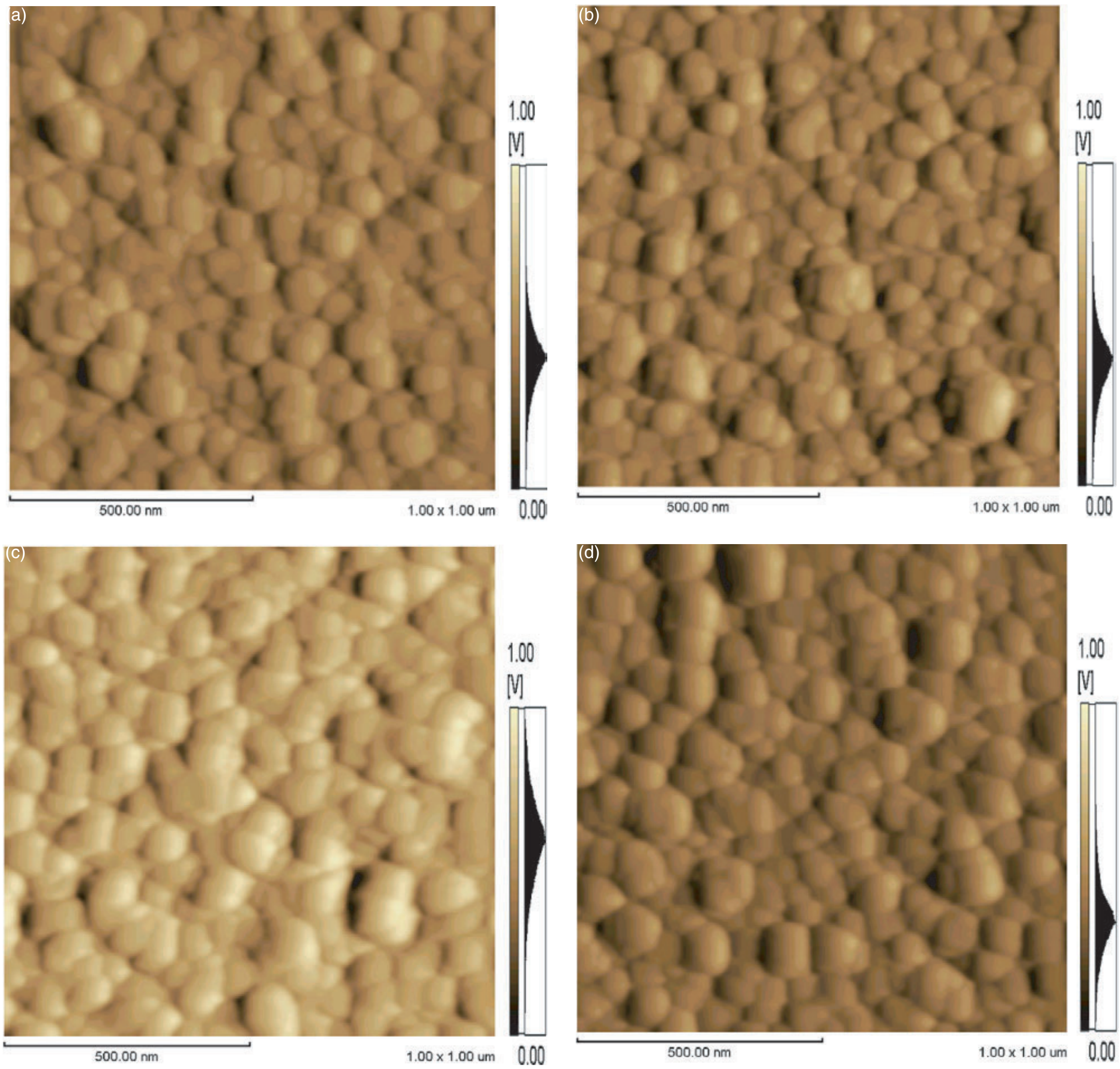

Figure 3. FFM images of ZnO film deposited on Si substrate at the various sputtering powers of: (*a*) 150 W, (*b*) 175 W, (*c*) 200 W, (*d*) 225 W.

3.3. Fractal analysis

Fractal analysis is used to investigate the surface science that significantly simplifies the analysis of the morphology surface properties on ZnO thin films. It has been established in previous research that roughness parameters based on conventional theories depend on the sampling interval of the particular measuring instrument used [6]. In addition, the fractal surface maintains the characteristics of continuity, non-differentiability, self-similar and self-affine. By definition, a fractal is a set for which the so-called fractal dimension, D_s, always exceeds the topological dimension. The fractal dimension lies within the range $2 \leqslant D_s \leqslant 3$ [17, 18], where the value of $D_s = 2$ denotes a flat surface, and an increasing value of D_s represents an increasing surface roughness. The structure function and variation methods are preformed successfully with images containing a few hundred pixels and optimumly describe the surface topography of ZnO thin films. Here, we have adopted the structure function method to analyse the fractal dimension of ZnO thin films [7].

In this methodology, the fractal dimension is calculated from the least-square degeneration line using a log–log plot of structure function $S(\tau)$ vs a large vector τ [7]. The fractal dimension D_s determines the relative amounts of the surface irregularities for different distance scales. The fractal dimensions were derived from the Shimadzu fractal software used for this experiment. The results of fractal dimension analysis are shown in figure 6 and indicate that the fractal dimension is ≈ 2 and has a lower complicated geometry. That is to say, the higher the fractal dimension the rougher the surface tends to be. In figure 6, the result of the fractal

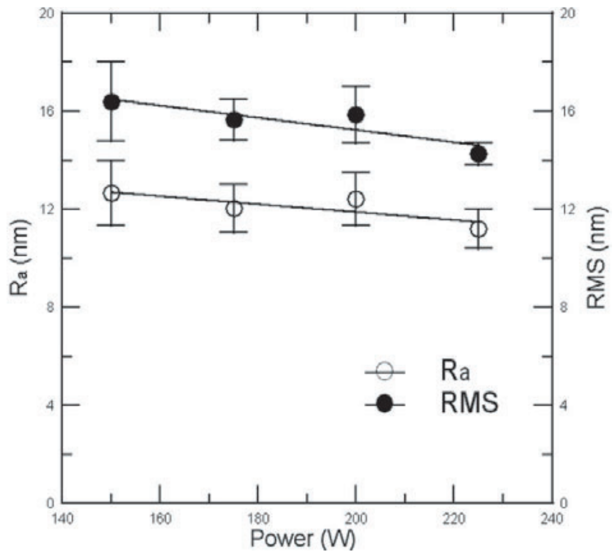

Figure 4. Variation of average surface roughness (R_a) and root-mean-square surface roughness (RMS) at various sputtering powers.

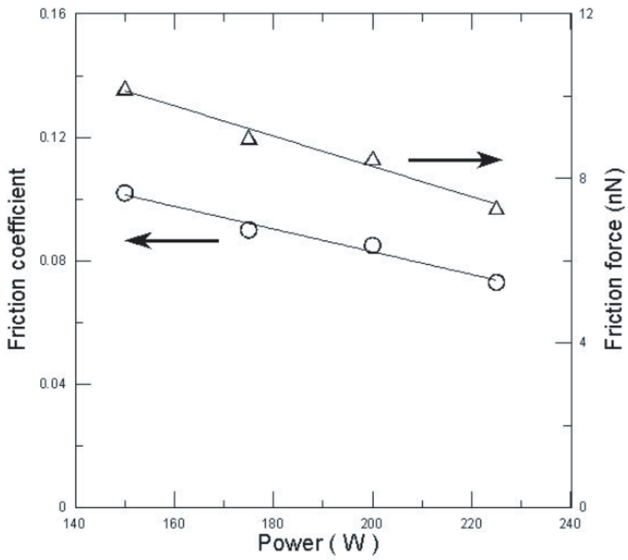

Figure 5. Variation of friction force and friction coefficient at various sputtering powers.

analysis at a sputtering power of 225 W is 2.03312. They both are at the lowest and therefore the closest to a perfect surface. In addition in figure 1 the XRD patterns show the best orientation at a sputtering power of 225 W. As can be seen in figures 4 and 6, both the results of surface roughness and fractal dimension decreases as the sputtering power increases and it is observed that the surface becomes slightly smoother.

According to the above-mentioned experimental records the XRD results clearly display that the deposited films have larger grain size and lower surface roughness as the sputtering powers increase, moreover the fractal dimension is smaller at higher sputtering powers. Our results are in agreement with a previous study [19], which shows lower fractal dimension of

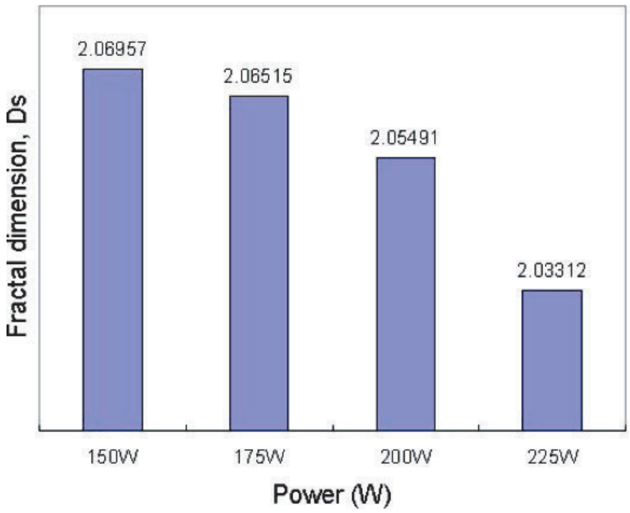

Figure 6. Fractal dimensions of ZnO thin films at various sputtering powers.

GaN films which exhibit larger grain size, smoother surface and lower fractal dimension. Using these techniques the features of films can be accurately characterized.

3.4. Nanowear tests

Typical wear marks for ZnO thin films at the sputtering power of 150W with different loadings of 82.2, 110 and 137 μN are shown in figure 7. In addition, nanowear tests found out that the wear depths are 31.75 nm, 40.32 nm and 48.28 nm for the applied loadings of 82.2 μN, 110 μN and 137 μN, respectively. Thus, the changes in wear depth is assumed to be due to structural changes of the ZnO thin films. The relationship between the average wear depth and sputtering powers at a 110 μN applied loading is shown in figure 8. Clearly increasing the sputtering power is beneficial for wear resistance. In terms of this, we can obtain the information that the higher the sputtering power, the better the quality of the film.

4. Conclusion

ZnO thin films were deposited by RF magnetron sputtering system at various sputtering powers. XRD indicated that the crystalline structures and the orientations of the films had a strong *c*-axis. A SPM apparatus was used to conduct nanofriction, nanowear and fractal investigations to determine the nanotribological characteristics of the deposited films. In this paper, considering that the distribution of the grain is random, fractal analysis is essentially applied to characterize quantitatively the surface roughness of films measured by AFM. Furthermore, fractal analysis has provided a valuable description of the irregularities found on ZnO thin film surfaces and it has been shown that both roughness and the fractal dimension D_s decrease with increasing sputtering powers. The higher sputtering power leads to larger crystallite size and more pronounced preferential orientation, a smoother surface and lower fractal

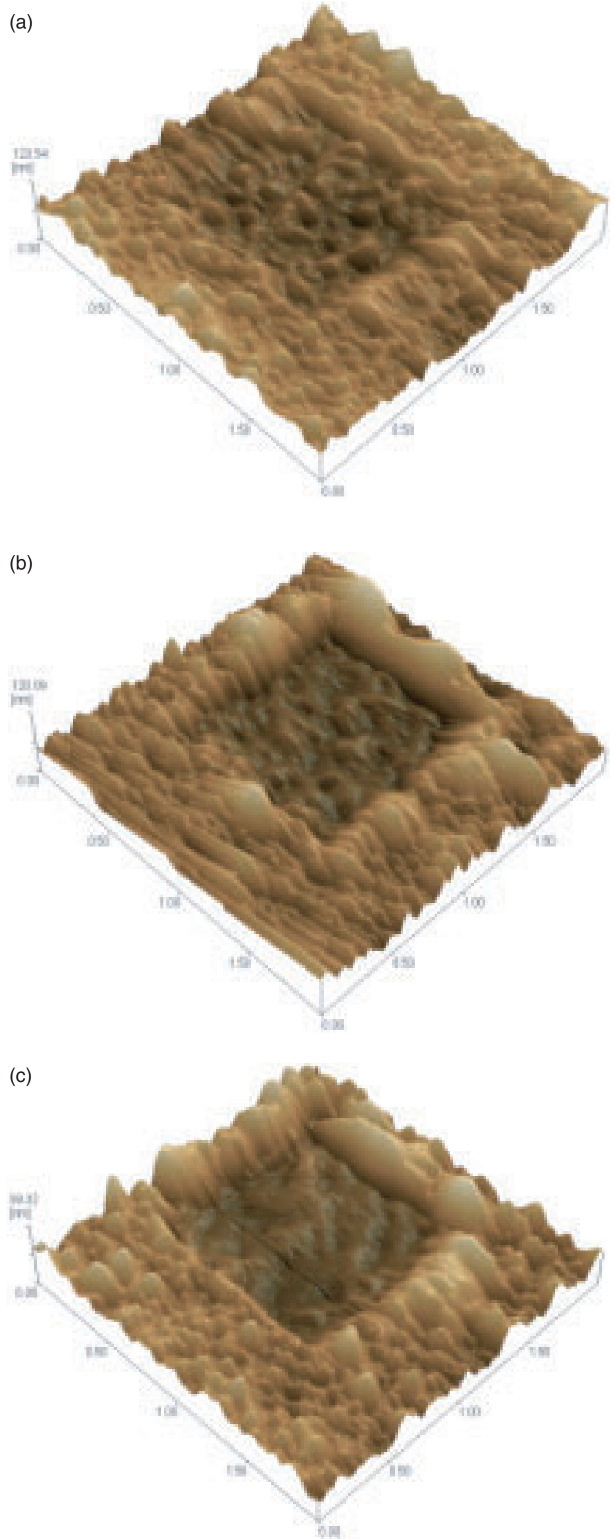

Figure 7. Surface topography of ZnO thin films at a sputtering power of 150 W at the different loads of: (*a*) 82.2 μN, (*b*) 110 μN, (*c*) 137μN.

dimension. Fractal analysis could be used to obtain suitable ZnO films for applications, such as SAW devices. To sum up, the results confirm that various sputtering powers play an important role in the nanotribological characteristics

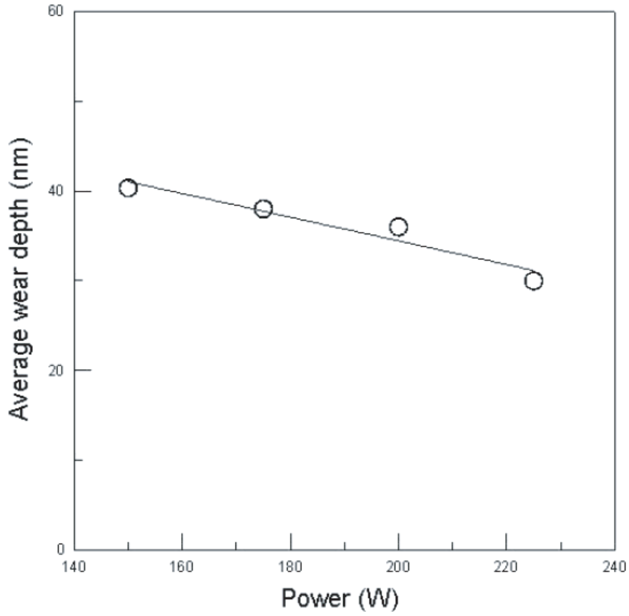

Figure 8. Variation of average wear depth at various sputtering powers using a constant load of 110 μN.

and fractal analysis of ZnO thin films deposited on silicon substrate.

Acknowledgments

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