CRYSTALLOGRAPHIC CHARACTERISTICS OF ZnO THIN FILMS DEPOSITED ONTO GLASS SUBSTRATES BY DC-MAGNETRON SPUTTERING

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ABSTRACT

CRYSTALLOGRAPHIC CHARACTERISTICS OF ZnO THIN FILMS DEPOSITED ONTO GLASS SUBSTRATES BY DC-MAGNETRON SPUTTERING. High quality ZnO thin films were deposited onto glass substrates by dc-magnetron sputtering. It was found that the characteristics of ZnO films depend on the deposition parameters obtained. Those parameters are temperature, pressure, gas mixture, and magnetron variation. By adjusting these parameters, ZnO thin films with an extremely sharp X-ray diffraction peak with an 0.484^0 full width at half-maximum were obtained. The resistivity of the deposited ZnO films from target ZnO on substrate temperature 430^0C is $0.307.10^3 \Omega$.cm, and the resistivity of the deposited ZnO films from Zn target with 20% O_2 at substrate temperature 430^0C is $0.106.10^2 \Omega$.cm.

The crystallographic properties of ZnO thin films formed by the magnetron sputtering technique were investigated using X-ray diffraction, SEM (scanning electron microscopy) and UV-spectrophotometer.

As the substrate temperature increases, the (002) diffraction peak becomes progressively more dominant and at temperature near 430^0C the films are strongly textured with preferential orientation along the (002) axis. This result indicates that the c-axis of the ZnO thin films is oriented almost normal to the substrate. When the sputtering gas pressure is $6x10^{-2}$ Torr, the (002) diffraction peak is very strong and when gas pressure is high (up to $10x10^{-2}$Torr) the diffraction peak is very weak. The crystallites reorient themselves so that the c-axis is normal to the substrate as new surface layers are formed.

Keywords: ZnO Thin Films, crystallographic, dc-magnetron sputtering

INTRODUCTION

So far, ZnO is considered as a promising material for many different applications such as solar cells, gas sensors, ultrasonic oscillators and transducers [1]. ZnO thin films are well-known materials used for the piezoelectric excitation and detection of the vibration motion of the resonator. ZnO is an attractive material to use because it combines a high piezoelectric coefficient with an IC-based deposition process [2] and [3]. Special application is in micromechanical sensors, which measure mechanical quantities such as force, pressure and acceleration [4].

Several techniques, such as rf or dc sputtering, ion plating and chemical vapor deposition [5], have been developed in recent years to form

ZnO thin films on many kinds of substrate. The essential point in the formation of ZnO thin films is to control its c-axis orientation, which is usually normal to the substrate within a certain distribution angle. Many workers have reported that if the sputtering method is used to form ZnO thin films, the c-axis orientation is influenced by the formation conditions, such as the location of the substrate, the substrate temperature, the deposition rate, and the sputtering gas pressure. However, there have been very few reports on the relation between the c-axis orientation and the growth of the film.

This report is concerned with the influence of the films thickness and sputtering gas pressure on the c-axis orientation and other crystallographic characteristics of ZnO thin films. The mechanism of crystal reorientation is also considered.

ZnO has wurtzite-type crystal, the form is hexagonal, and a 6-mm symmetry as shown in Fig. 1. The wurtzite structure has a rather open character, as is evident from the figure. As a result one might expect that the zinc ions in the crystal could readily exist in the interstitial sites as well as the normal lattice sites. Because of the large size of the oxygen ions, one would not expect the existence of oxygen in interstitial sites to be energetically feasible. The calculations presented previously with regard to the energy of formation of Schottky defects indicated that the energy was sufficiently low, so that this could well be the predominant type of defect in most crystals, unless some particular characteristics of the compound allowed some other defects to be energetically more favorable. The structure of ZnO fits this particular situation. Experimental information must be obtained, however, to decide what defect type is predominant [6].

The simplest imperfection is a lattice vacancy, which is a missing atom or ion, also known as a Schottky defect. A lattice vacancy is often indicated in illustrations and in chemical equations by a square. A Schottky defect is created in a perfect crystal by transferring an atom from a lattice site in the interior to a lattice site on the surface of the crystal. In thermal equilibrium a certain number of lattice vacancies are always present in an otherwise perfect crystal, because the entropy is increased by the presence of disorder in the structure [7].

In metals with close-packed structures the proportion of lattice sites vacant at temperatures just below the melting point is of the order of 10^{-3} to 10^{-4}.

The probability that a given site is vacant is proportional to the Boltzmann factor for thermal equilibrium: $P = exp(-E_v /k_B T)$, where E_v is the energy required to take an atom from a lattice site inside the crystal to a lattice site on the surface. If there are N atoms, the equilibrium number n of vacancies is given by the Boltzmann factor

$$\frac{n}{N-n} = \exp(-E_v / k_B T) \ . \qquad (1)$$

If n << N, then

$$n/N \cong \exp(-E_v / k_B T). \qquad (2)$$

If $E_v \approx 1$ eV and $T \approx 1000$ K, then $n/N \approx e^{-12} \approx 10^{-5}$.
The equilibrium concentration of vacancies decreases as the emperature decreases.

The ZnO is a simple yet it is a complex material. ZnO is a versatile material with unique properties. Sputtered ZnO films are applied as piezoelectric transducers, planar optical wave guides, detectors of oxidizing and reducing gases, and as transparent electrodes in a wide variety of devices.

Controlling the properties can be quite troublesome. The deposition parameters, substrate types, pretreatment of the substrates and post deposition treatment greatly influence the properties of the layers (as with all thin-films materials). Moreover, environmental parameters such as moisture content and light conditions affect the electric, piezoelectric, optical, and chemical properties. This can cause stability problems. However, surmounting all these problems by proper materials research and engineering yield many opportunities for a material with a simple chemical formula, but, with rather complex properties [8].

Any calculation of the intensity of a diffracted beam must always begin with the structure factor. From the intensity information, the position of the atom can be determined. There are six factors affecting the relative intensity of the diffraction lines on a powder pattern:

1. Polarization factor
2. Structure factor
3. Multiplicity factor
4. Lorentz factor
5. Absorption factor
6. Temperature factor.

And the width of the curve at half its maximum height is *W* and if *V* is the mean pulse size, then the resolution *R* of the counter is [9]:

$$R = \frac{W}{V} \qquad (3)$$

the smaller *R*, the better the resolution.

In this report, we investigated the effect of deposition parameters on the properties of dc-magnetron sputtered ZnO films and showed the correlation of crystal reorientation change with microstructural evolution.

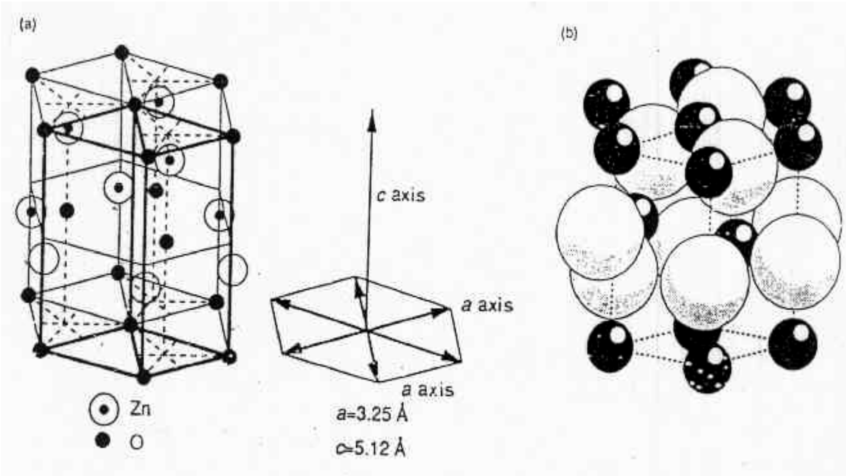


Figure 1. ZnO Crystal Structure: (a) Wurtzite crystal structure with 6-mm symmetry and (b) ZnO model with zinc atoms represented as smaller balls, oxygen atoms as larger balls [6].

EXPERIMENTS

Sample preparation

In this study, dc magnetron sputtering equipment was used to form the ZnO thin films. Figure 2 is the schematic diagram of the equipment. The glass substrate is positioned obliquely above the target. The substrate heater is positioned in the anode, and the substrate temperature is controlled by a thermocouple. The substrate makes thermal contact with the anode through the copper substrate holder, and the target-anode distance is kept at about 30 mm.

The glass substrate has been used during this research, including microscope slides. The substrate was cleaned thoroughly prior to deposition. Zinc oxide deposition was carried out using magnetron sputter source coupled to 60 W dc power supplies. The vacuum was evacuated by an Edwards 306 pumping system, the vacuum chamber was exhausted by an oil-diffusion pump and rotary pump, and this system achieved base pressures of $2x10^{-6}$ Torr in around 30 minutes. The 'sputter up' arrangement and inverted top-hat substrate holder allowed substrate heating up to 450^0C. Zinc oxide was sputtered from metallic zinc and ceramic zinc oxide targets in an atmosphere of argon and oxygen. The total pressure was varied from $4x10^{-2}$ to $9x10^{-2}$ Torr and the dc power varied from 30 to 60 W. Films were deposited at substrate temperatures between ambient temperature and $450\,^0$C. The gas composition of the sputtering atmosphere was constantly measured using flow gas meter. Sputtering was carried out in a mixture of Ar (70-100%)+O_2 (5-30%), the gas mixture being admitted through a variable leak valve and controlled by the main valve of the diffusion pump.

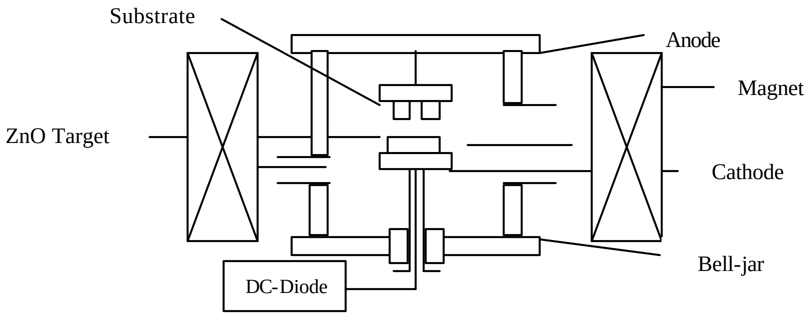


Figure 2. Schematic Diagram of Sputtering Equipment.

Examination Method

In this paper, the examination using X-ray diffraction, Co-Ká radiation, SEM (scanning electron microscopy), EDAX at BATAN Serpong, and UV-spectrophotometer for measuring the thickness of ZnO thin films at Faculty of Science of Department of Physics, University of Indonesia Jakarta.

RESULT AND DISCUSSIONS

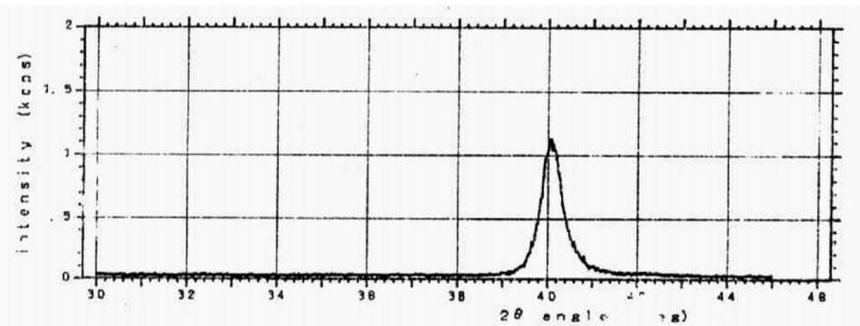
The influence of experimental parameters such as growth temperature, growth pressure, growth rate, morphology, optical transmittance and crystallographic orientation of ZnO films were studied as an aim of optimizing the technological process and to obtain uniform thin films of such a quality necessary for micromechanical transducers.

X-ray diffraction results

Influence of the growth temperature

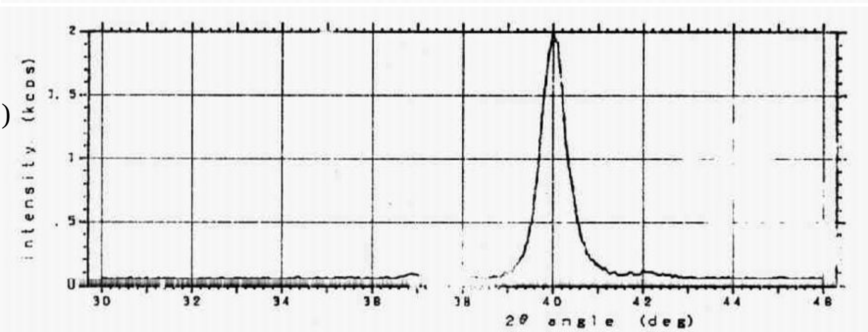
ZnO films were grown in the temperature region from 200^{0}C up to 450^{0}C (hereafter referred as the substrate temperature). It was found that the deposition rate increased from the result examination of resistance and thickness of thin films with increasing temperature. The results of X-ray diffraction measurements shown in Fig. 3 They indicate that the films produced at low temperatures have only a weak (002) orientation which is associated by the X-ray diffraction table for ZnO materials from the Joint Committee on Powder Diffraction Standards Associateship at the National Bureau of Standards. As the growth temperature increases, the (002) diffraction peak becomes progressively more dominant and at temperature near 430 ^{0}C, the films were strongly textured with preferential orientation along the (002) axis. This result indicates that the c-axis of the ZnO thin films is oriented almost normal to the substrate. Fig. 3 shows the line profiles of the (002) diffraction peaks of the ZnO thin films for several temperatures. The same tendency was observed by other authors [1] and [10]. The test by SEM indicates that ZnO crystal seeds seem big also. This resistance shows a high value because ZnO deposition on substrate is more thick than others.

a)



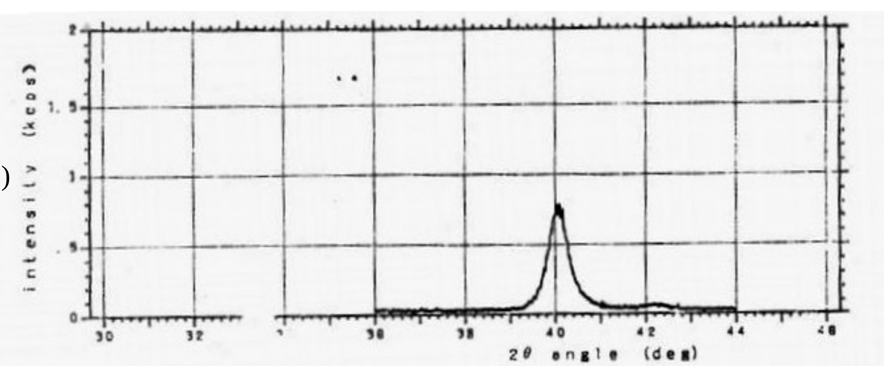
< ZnO 450 ^{o}C >

b)



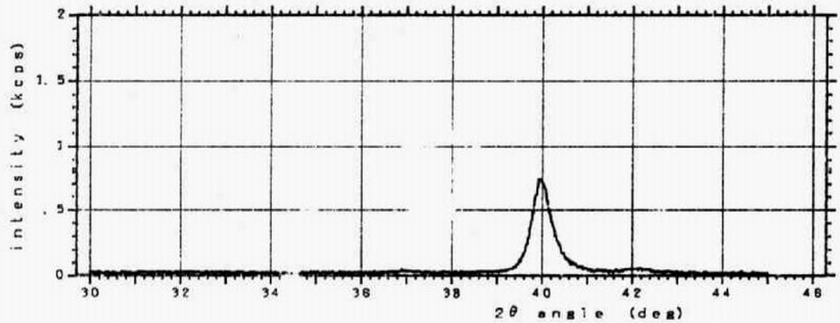
< ZnO 430^{o}C >

c)



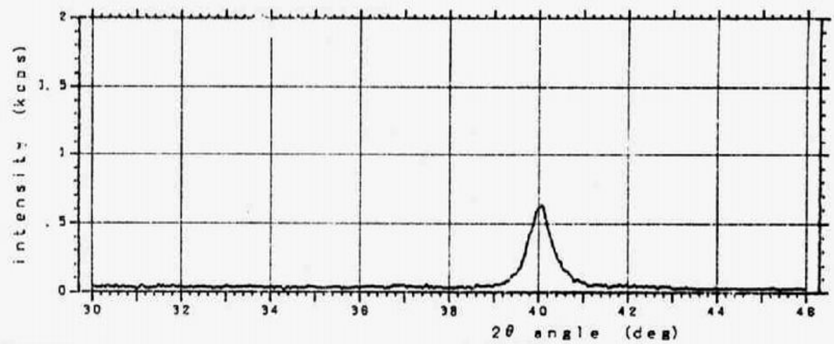
< ZnO 400^{o}C >

d)



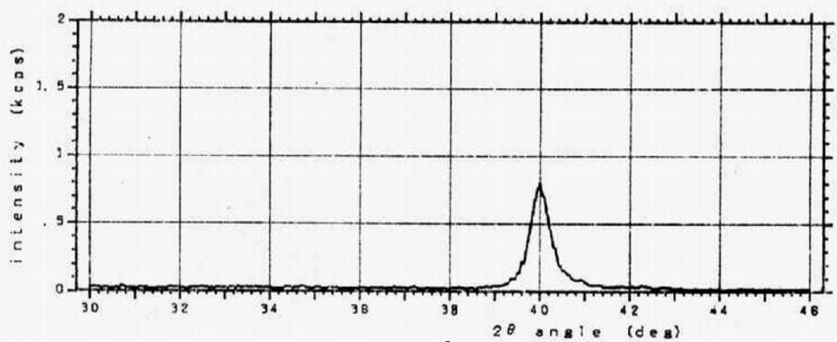
< ZnO 350^{o}C >

e)



< ZnO 300^{o}C >

f)



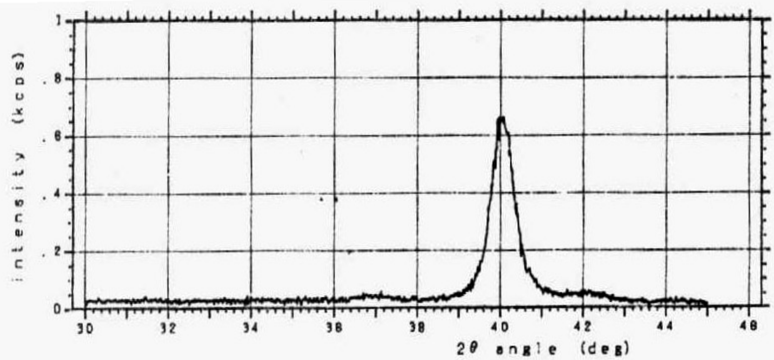
< ZnO 250^{o}C >

Figure 3. Line profiles of the (002) diffraction peaks of the ZnO thin films for several substrates temperatures (250 – 450)^{o}C, with X-ray condition: Co 30kV 30mA and split ; (SS) 1 deg.

Influence of the pressure growth.

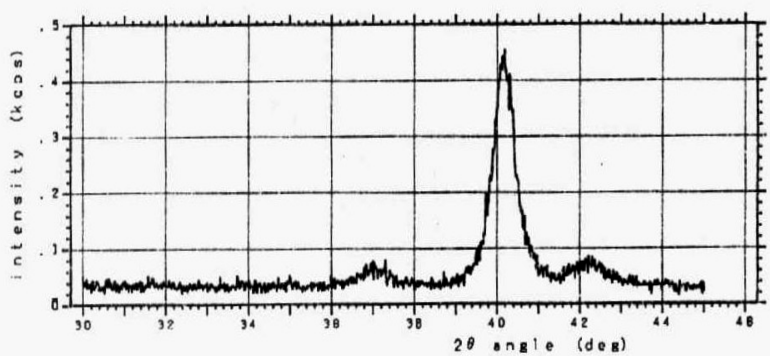
X-ray diffraction results for deposition pressure variation ($6x10^{-2}$ to $100x10^{-2}$ Torr) indicated that only (002) diffraction peak of ZnO was observed in the ZnO thin films formed at a sputtering gas pressure between ($6x10^{-2}$ to $10x10^{-2}$) Torr. This result indicates that the c-axis of the ZnO thin films is oriented almost normal to the substrate. The results of X-ray diffraction measurements shown in Fig. 4, X-ray diffraction for ZnO materials which is referenced from the Joint Committee on Powder Diffraction Standards Associateship at the National Bureau of Standards. When the growth pressure increases, the (002) diffraction peak becomes weak. The same tendency was observed by other authors [11].

a)



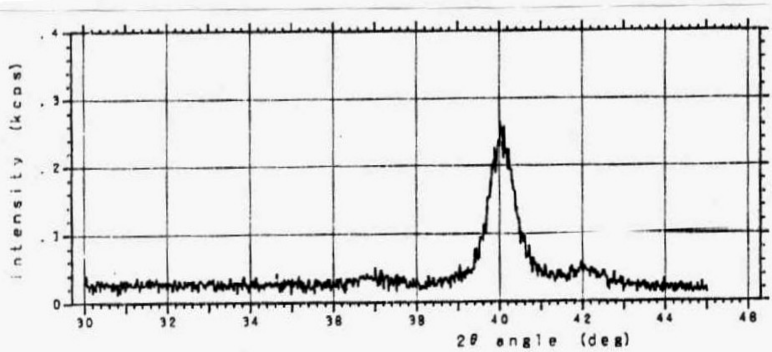
< ZnO 60 mTorr >

b)



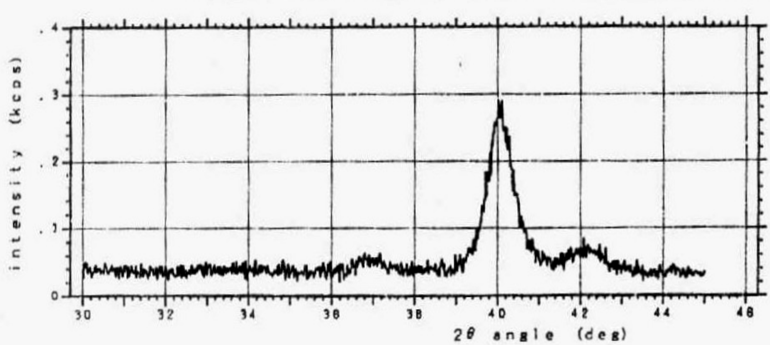
< ZnO 70 mTorr >

c)



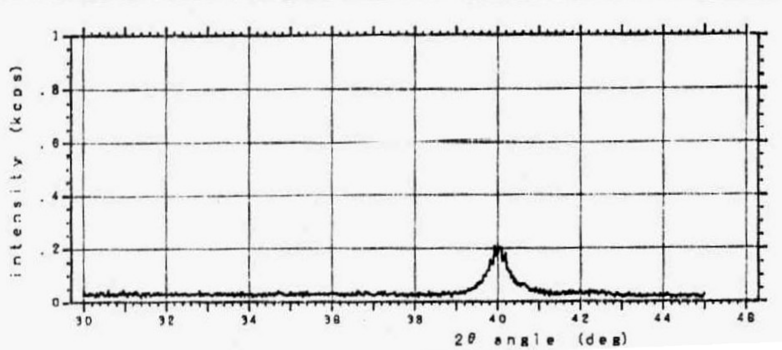
< ZnO 80 mTorr >

d)



< ZnO 90 mTorr >

e)



< ZnO 100 mTorr >

Figure 4. Line profiles of the (002) diffraction peaks of the ZnO thin films for several pressures (60 – 100)m Torr, with X-ray condition: Co 30kV 30mA and split ; (SS) 1 deg.

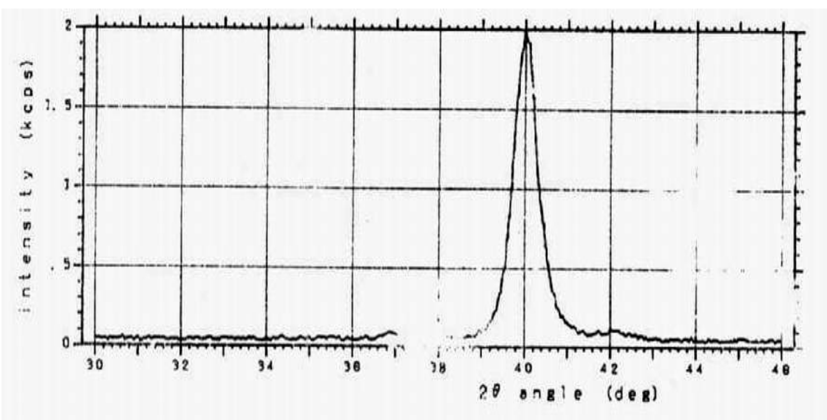
ZnO films were grown in the pressure region from $6x10^{-2}$ to $100x10^{-2}$ Torr. It was found that the diffraction rate increases with decreasing pressure.

Influence of the gas mixture

The energetic oxygen species were generated in front of the target as negative ions during the sputtering of oxides by Ar^+ ions, which were accelerated in the cathode fall region, and then neutralized in transit to the anode through charge exchange with Ar gas molecules. This study clearly shows the effect of the energetic oxygen species on the orientation and the microstructure of the films. The results of X-ray diffraction measurements shown in Fig. 5 show the variation of XRD patterns of the films deposited from zinc oxide target at different oxygen partial pressures at a total pressure of 6.10^{-2} Torr and substrate temperature of $430\,^0$C, to test the films thickness 1 μm using SEM. The ZnO films deposited in pure argon, perpendicular surface of substrate oriented normal to the substrate. With increasing oxygen partial pressures to 20%, the XRD pattern is shown to be weak. The same tendency was observed by other authors [12]

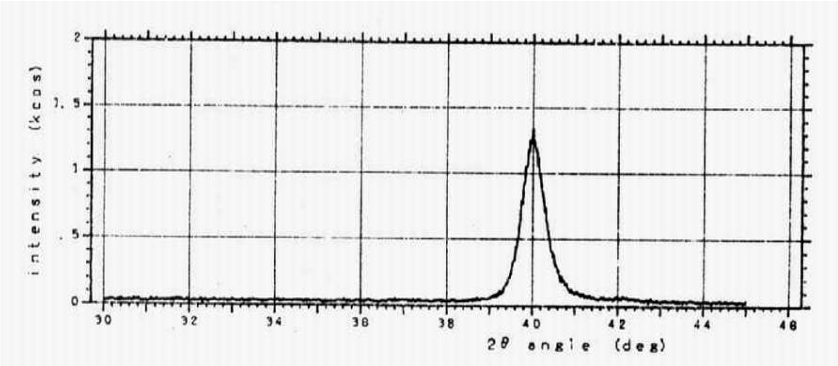
The results of X-ray diffraction measurements shown in Fig. 6, show variation of XRD patterns of the films deposited from zinc metal target at different oxygen partial pressures at a total pressure of 6.10^{-2} Torr and substrate temperature of $430\,^0$C. The X-ray diffraction shows three peaks (100), (002), and (101) respectively at 100% Ar. The same previous peaks appeared when applying a mixture of 10% O_2 with Ar, nevertheless those peaks seemed to be very weak in this case. The result of the X-ray peak diffraction when implementing 20% O_2 with Ar, seemed to be strong only in (002) orientation.

a)



< ZnO 100%Ar >

b)



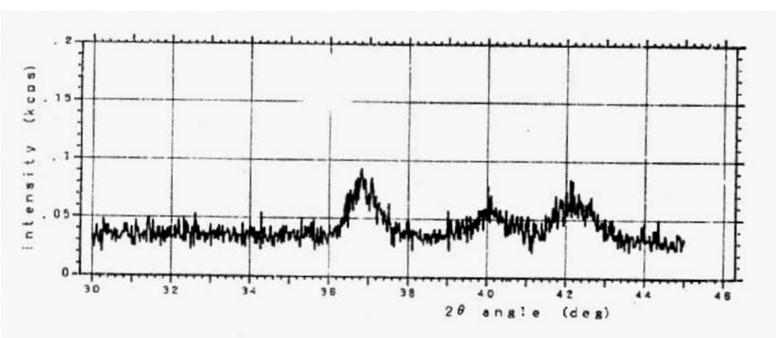
< ZnO 20%O_2 >

Figure 5. XRD patterns of the ZnO films from Zinc Oxide ZnO as a function of the oxygen partial pressure: (a) Ar/O_2 ==100/0, (b) Ar/O_2 =80/20, with X-ray condition: Co 30kV 30mA and split; (SS) 1 deg.

Influence of Zn Sputtering

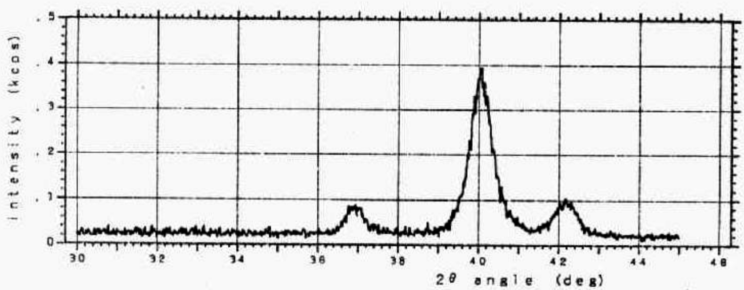
Fig. 6.a shows the Zn thin films of the gas sputtering Ar (100%) has three weak peaks [100] [002] [101]. Fig 6.b shows the Zn thin films of gas sputtering mixture result of Ar (90%) + O_2 (10%) also has the same three weak peaks but the crystal orientation more clearly. Fig. 6.c shows the Zn thin of gas sputtering mixture result of Ar (80%) + O_2 (20%) has strong peak [002], and perpendicular with the glass substrate respectively by comparing them with peaks mentioned which is detected by X-ray diffraction for Zn materials which is referenced from hand book Joint Committee on Powder Diffraction Standards Associateship at the National Bureau of Standards powder diffraction standards [13].

a)



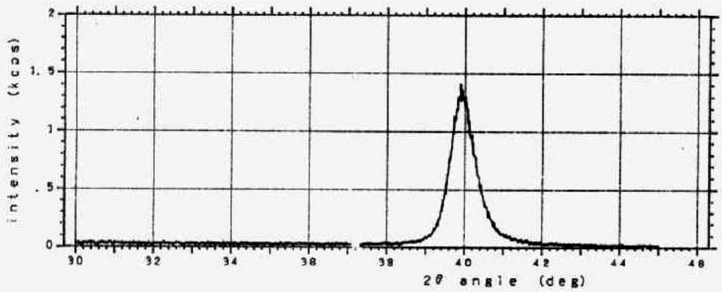
< Zn 100% Ar >

b)



< Zn 10%O_2 >

c)



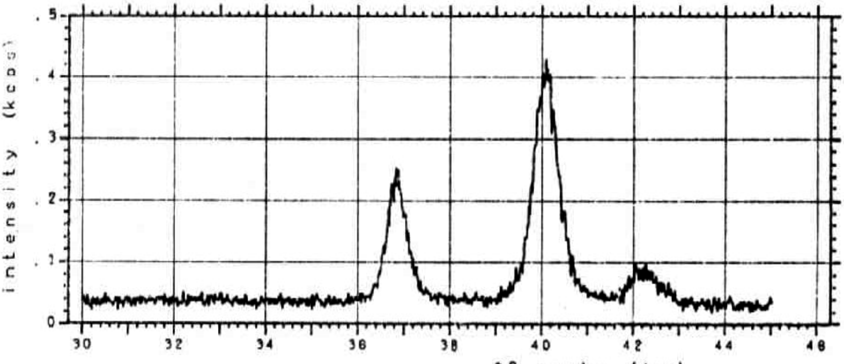
< Zn 20%O_2 >

Figure. 6. XRD Patterns of the ZnO films from Zinc metal (Zn) as a function of the oxygen partial pressure; (a) Ar/O_2 ==100/0, (b) Ar/O_2 =90/10 (c) Ar/O_2 =80/20, with X-ray condition: Co 30kV 30mA and split ; (SS) 1 deg.

Influence of magnetic field

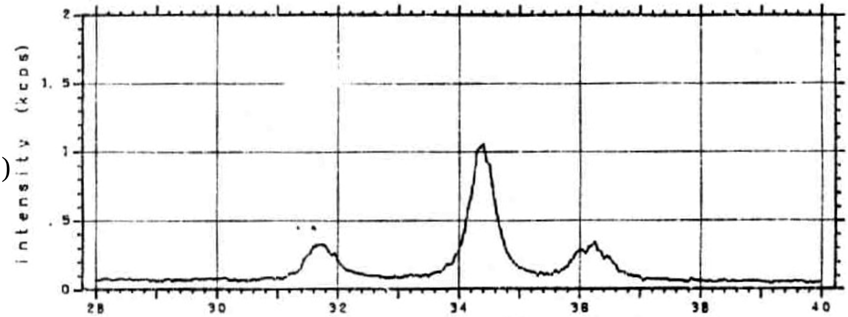
Magnetron sputtering has many other advantages. The primary advantages are; (1) high deposition rates, (2) ease of sputtering of any metals, alloys or compounds, (3) high-purity films, (4) extremely high adhesion of films, (5) excellent coverage of steps and small features, (6) ability to coat heat-sensitive substrates, (7) ease of automation, (8) excellent uniformity on large-area substrates [14], (9) work at low pressure and (10) at low current (according to this research) yield the same thin film.

a)



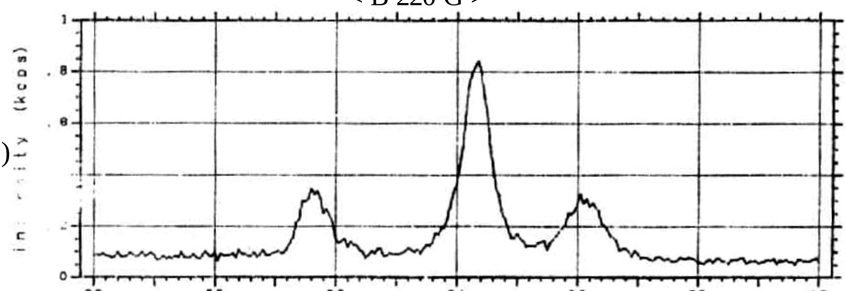
< B 0 G >

b)



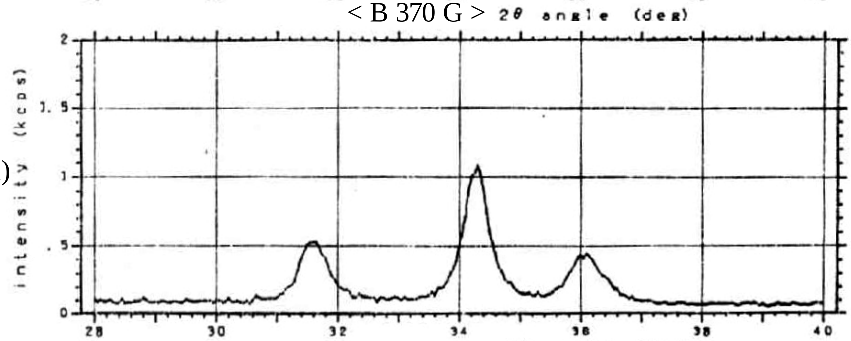
< B 220 G >

c)



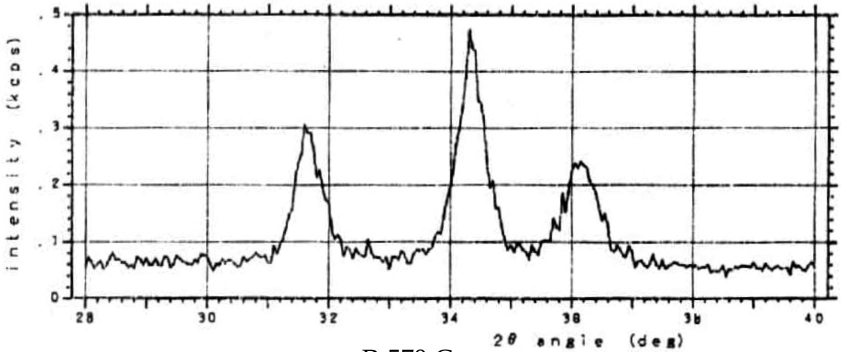
< B 370 G >

d)



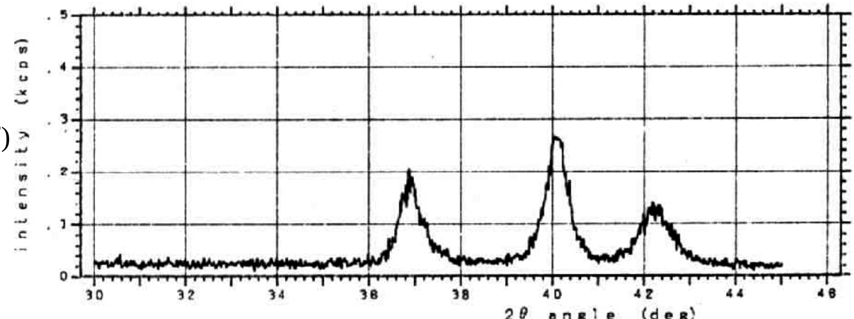
< B 470 G >

e)



< B 570 G >

f)



< B 670 G >

Figure 7. X-Ray Diffraction Spectrum of the ZnO thin films repared by magnetron variation (0 – 670) G, with X-ray condition: Co 30kV 30mA and split ; (SS) 1 deg.

The results of X-ray diffraction measurements shown in Fig. 7 show the X-ray diffraction results for deposition magnetron variation (0 G and 670 G) at ambient temperature corresponding to the planes [100], [002], and [101] respectively by comparing them with the peaks mentioned in which is detected by the table X-ray diffraction for Zn-O materials which is referenced from the Joint Committee on Powder Diffraction Standards Associateship at the National Bureau of Standards powder diffraction standards [13]. And at high temperature, only (002) diffraction peak of ZnO was observed in the ZnO thin films formed at a magnetron sputtering. This result indicates that the c-axis of the ZnO thin films is oriented almost normal to the substrate. From fig. 7, we can see that the optimum results (the homogeneous of thin layer and the high velocity of deposition movement) of the dc magnetron sputtering device on X-ray diffraction spectrum can be achieved at magnetic field 370G.

SEM observations

The microstructure of the ZnO films was determined using scanning electron microscopy (SEM). Fig. 8 shows a typical SEM micrograph taken from the perpendicular films surface of 1 µm thick oriented normal to the substrate. This shows that the structure is dense and without porosity. No cracks or voids were observed. The oriented films have a columnar structure with columns perpendicular to the substrate.

Fig. 8 shows a SEM micrographs of ZnO thin films formed at a sputtering gas pressure of $6x10^{-2}$ Torr. A columnar structure can be observed. These results on the crystallographic characteristics of ZnO thin films and Zn thin films were obtained with 100% Argon sputtering gas and a substrate temperature of 430^0C. Thin films formed at a gas mixture with oxygen (Ar/O_2) 20% O_2, are normal and correspond with the results of other workers [11].

(a)

(b)

(c)

(d)

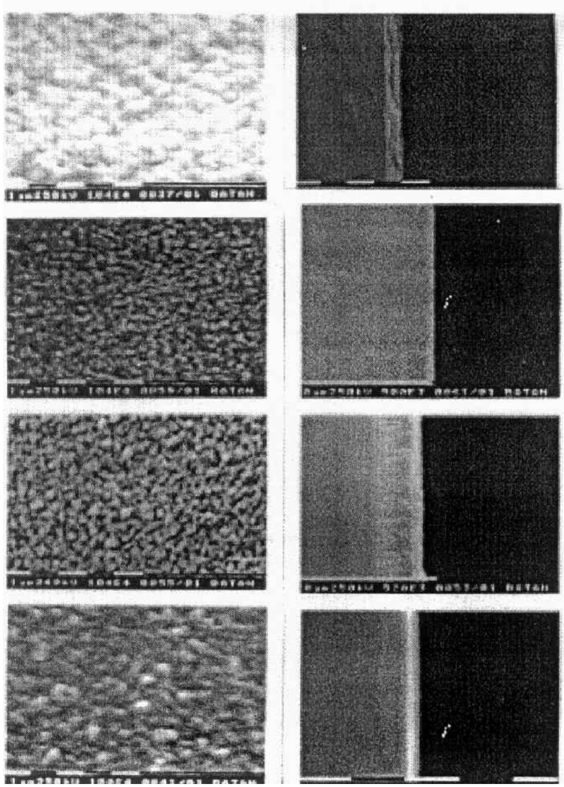


Figure 8. Scanning Electron Micrographs of Zn and ZnO films, deposition at a temperature of 430^0C under $6x10^{-2}$ Torr, as a function of Oxygen partial pressure: (a) Ar/O_2 =100/0 with ZnO films, (b) Ar/O_2 =100/0 with Zn films, (c) Ar/O_2 =80/20 with ZnO films, (d) Ar/O_2 =80/20 with Zn films.

Films Thickness Determination

Many references on this subject can be found in a recent review. These interferometric techniques can be divided into two general categories. In the most commonly used technique the radiation is reflected from the sample films in a spectrophotometer with fringes being formed as a function of wavelength. This technique has been named CARIS (constant-angle reflection interference spectroscopy) by Reizman and Van Gelder. In the other technique, interference fringes are formed by varying the angle of observation, and it is therefore called VAMFO (variable-angle monochromatic fringe observation). In both techniques, the films thickness can be given approximately as follows :.

$$d = \frac{1}{2n_1\left(\frac{1}{l_1} - \frac{1}{l_2}\right)} \qquad (4)$$

with

$$n_1 = \frac{(1+n_0) + \sqrt{(1+n_0)^2 - 4n_0T_c}}{2\sqrt{T_c}} \qquad (5)$$

$$T_c = 100\% - \%R$$

where: d = thickness

l = wavelength

ë1, ë2, … = the arbitrary nearer wavelength

n_1 = refractive index

n_o = glass refractive index = 1.5

T_c = minimal transmission

%R = reflection

The data collected from transmission spectrum is shown in Fig. 9:

Name : Zn, mixture gas Ar 60% + O_2 40%

Threshold : 0.200

ë1 = 569 nm

ë2 = 666 nm

n_1 = 1.546

Thus d = 1263 nm = 1.263 μm (film thickness)

T_c = 83%

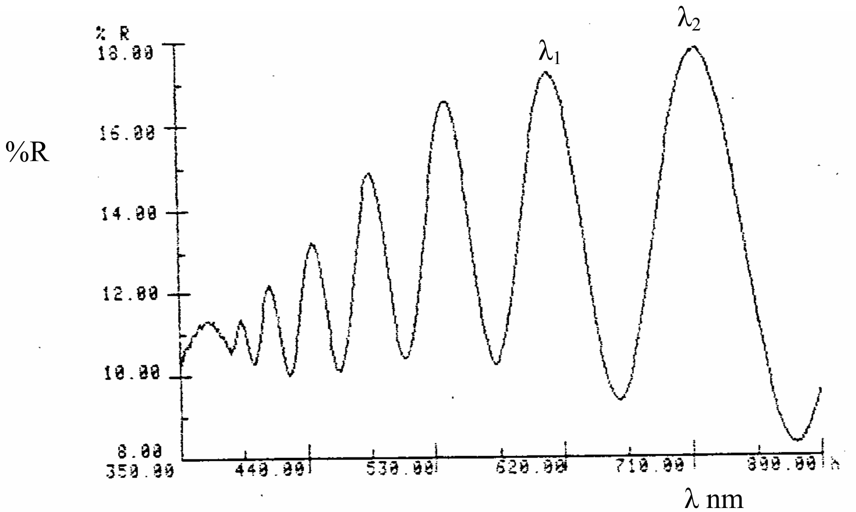


Figure 9. UV-Spectrophotometer transmission spectrum of the ZnO thin films prepared by dc-magnetron sputtering

The properties of the ZnO thin films prepared by the dc-magnetron sputtering under the appropriate conditions were examined. Figures 3,4,5,6 and 7 show the X-ray diffraction spectrum of the ZnO thin films. In this X-ray diffraction spectrum, the peaks corresponding to the planes [100], [002] and [101] [16] of the hexagonal phase of ZnO were present. The peak at $2\theta = 40.5^0$, which corresponds to the diffraction from the [002] plane, was very strong. This film shows good crystalline character and exhibits a preferential orientation with the c-axis perpendicular to the substrate. The same preferred orientation of the films is found in the case of ZnO films prepared using other methods [8] and [16].

Table 1 shows the powder diffraction standards of Zinc Oxide [12]. The quantity I/I_1 is the scattered intensity ratio while hkl is the Miller indices.

Table 1. The powder diffraction standards of ZnO

d(Å)	**I/I_1**	**hkl**
2.816	71	100
2.602	56	002
2.476	100	101
1.911	29	102

Fig. 8 shows the scanning electron micrographs of the ZnO thin films on a glass substrate. It was observed that hexagonal particles were closely packed on the substrate. The hexagonal particles seem to grow from the surface of the substrate. The thickness of the films was 1 μm and the hexagonal particles were about 1 μm long. The grain sizes of the ZnO thin films are almost the same as the sizes obtained with SEM, and are independent of the sputtering gas pressure and temperature.

The resistivity of the ZnO films was measured to be 10^{-2} Ω cm, by using four-point probe at 430^0C. The effect of the film thickness on the orientation of crystal growth is well known and that the orientation of the crystals in the film is strongly dependent on the crystallography of the substrate. However, preferential orientation along the [002] plane has been observed for ZnO films even on glass substrates, suggesting that in this material grain growth orientation takes place inherently. Then, it is expected that the XRD peak intensity should increase in proportion to the films thickness [17].

CONCLUSIONS

Zinc oxide has been deposited using a dc-magnetron sputtering system, from metallic zinc and ceramic zinc oxide targets. In this experiment we have shown that thin films are ~1 μm for 2 hours and the deposition time for 1 hour is 0.5 μm.

The crystalline ZnO films with a good quality and a good (002) texture can be grown under considerably different process parameters. After a series of optimization experiments, crystalline films of c-axis-oriented ZnO were deposited at varying parameters of temperature, pressure, electrode distance, and magnetic field. The extremely preferred grain growth along the [002] plane was observed in the ZnO films post-heated at temperatures of $(200\text{-}450)^o$C.

The degree of the crystal orientation was low when the thickness of the films was small indicating that there is a critical thickness that offers highly oriented crystal growth. In addition to the above, it can be observed that, in case of applying a magnetron source, the experimental conditions at low pressure and low current field, yield the same thin films thickness. We also observed that in order to minimize the vacancies occurring in the crystal structure of the thin film, the sample should not be taken out of the sputtering chamber until the device is cooled to the ambient temperature. This is attributable to the Schottky defect occurring in the crystal structure.

In this paper we described Zn thin films which were made from target Zn, and ZnO thin films which were made from target Zn composed with active gas O_2 (20%). The result of ZnO thin films that mixtured with 20% O_2 is close to the result of the thin film which made from ZnO target.

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