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Room-Temperature Deposition of ZnO Thin Films by using DC Magnetron Sputtering

Putut Marwoto^{1,3,a*}, Sulhadi^{1,3,b}, Sugianto^{1,3,c}, Didik Aryanto^{2,3,d},
Edy Wibowo^{3,e} and Kiki Wahyuningsih^{3,f}

¹Department of Physics, Faculty of Mathematics and Science, Universitas Negeri Semarang,
Gunungpati, Semarang 50229 Jawa Tengah, Indonesia

²Department of Physics, Faculty of Mathematics and Science Education, IKIP PGRI
Semarang, Semarang 50125 Jawa Tengah, Indonesia

³Materials Research Group, Thin Film Laboratory, ¹Department of Physics, Universitas Negeri
Semarang, Gunungpati, Semarang 50229 Jawa Tengah, Indonesia

*^apmarwoto@yahoo.com, ^bsulhadipati@yahoo.com, ^csugianto_fis@yahoo.com,
^ddidik_phys@yahoo.co.id, ^eedywibowo86@yahoo.com, ^fwahyuningshunnes@gmail.com

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Abstract. ZnO thin films have successfully been deposited using DC magnetron sputtering at room temperature by means of plasma power variation. XRD results show that films were grown at a plasma power of 30 W and 40 W are polycrystalline, while at 20 W is considered as amorphous. The optical bandgap of films are shrinkage by increasing the plasma power. The broadest transmittance range is belongs to ZnO film growth at plasma power of 40 W. The electrical conductivity of ZnO films increase from $4.02 \times 10^{-7} (\Omega\text{cm})^{-1}$ to $8.92 \times 10^{-7} (\Omega\text{cm})^{-1}$ once the plasma power is increased. Based on the electrical and optical properties of the films it clearly be seen that ZnO film grown at plasma power of 40 W has highest transmittance and lower electrical resistivity therefore it appropriate for transparent conductive oxide (TCO).

Introduction

Transparent conductive oxide (TCO) has various applications; it is most commonly used for transparent electrode, window material for display, surface acoustic wave devices, sensors, and electronic transducer. One of the candidate materials of TCO is zinc oxide (ZnO); it is considered as an alternative material of indium tin oxide (ITO). ZnO material is considering as low cost material and it can be deposited at relatively low temperature [1]. ZnO has low electrical resistivity, high optical transmittance, and wide optical band gap (3.3 eV) [2-3]. Hence, ZnO has gaining interest for substitute material of ITO and SnO₂ as TCO. ZnO thin films were deposited through various methods such as molecular beam epitaxy (MBE), metal-organic chemical vapor deposition (MOCVD), sputtering, pulsed laser deposition (PLD) and chemical vapor deposition (CVD) [1-3]. By using our method, we success growth ZnO thin films at room temperature and low plasma power; it decreases the power overheads during deposition processes. The properties of grown films were comparable to the ZnO films growth by other sputtering method at relatively higher temperature and higher plasma power as well as other advanced method that previously reported [1-5].

Experimental

The ZnO thin films were deposited on corning glass substrates using dc magnetron sputtering [4]. ZnO target was made from high purity ZnO powder (99,99%). The corning glass substrates were cut into 1x1 cm² and washed using acetone and methanol in ultrasonic bath for 10 min and 5 min, respectively. Then, they were blown dry with oxygen before admitted to the sputtering system.

The ZnO films were deposited at room temperature and argon pressure of 500 mTorr by adjusting the plasma power of 20 W, 30 W, and 40 W, respectively. The deposition time was 1 h for all the films. The crystallinity and the preferred crystal orientation of ZnO films were analyzed by X-ray diffraction (XRD) with a $\text{CuK}\alpha$ radiation ($\lambda = 0.15406$ nm). The transmittance of films were analyzed by using UV-Vis spectrometer. The conductivity of the films were analyzed by I-V measurement.

Result and Discussions

Figure.1 shows the XRD patterns of ZnO thin films deposited at different plasma power. The spectra revealed that ZnO thin films grown at plasma power of 30 W and 40 W are polycrystalline with wurtzite structure, while at plasma power of 20 W, no sharp peak was observed therefore it was considered as amorphous structure. The amorphous spectrum of sample grown at plasma power of 20 W was therefore considered belongs to the glass substrate. It means ZnO film was not deposited yet on the surface of substrate. Based on XRD spectra, it can be notified that the peak intensity of (100), (101) and (110) are decrease, while the peak intensity of (002) plane orientation is increase by increasing the plasma power. The higher plasma power produce greater energy that can help film deposition. Atoms with higher energy have a high surface mobility therefore lead to higher deposition process on the surface of substrate. Moreover, the widening of diffraction peaks leads to lattice strains in ZnO film configuration. The value of lattice strain of ZnO films deposited at room temperature with variation of plasma power is around 0.179 to 0.269. The lattice strain of ZnO thin films can be determined by using Eq. 1 [5].

$$\varepsilon = \frac{\beta}{4 \tan \theta} \quad (1)$$

where ε is the lattice strain, β is full width at half maximum peak (FWHM), and θ is the angle of diffraction. Furthermore, by using Scherrer equation the average crystallite size of ZnO film can be determined. The average crystallite size of the ZnO thin films was obtained around 43-64 nm.

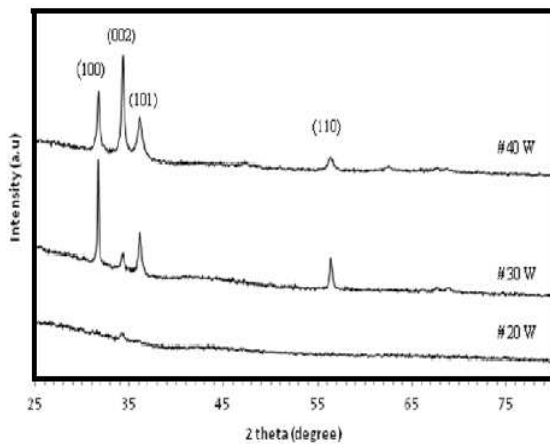


Figure 1. X-ray diffraction spectra of ZnO films formed at various plasma power.

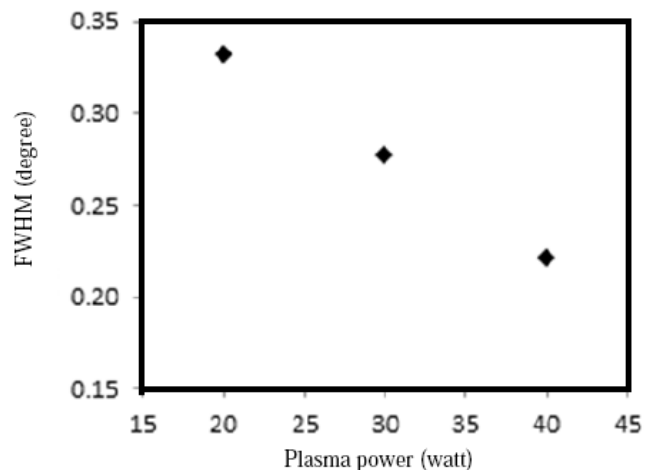


Figure 2. The FWHM of (002) peak's ZnO thin films prepared at various plasma power.

Figure. 2 shows the FWHM of (002) plane orientation of ZnO thin films. It can be clearly seen that the FWHM is decrease by increasing the plasma power. ZnO thin film is grown at plasma power of 40 W has narrowest FWHM. Film with narrow FWHM indicated that it is grown with small lattice strain [6]. Lattice strain influenced the quality of the crystal. The crystal's qualities of the films decrease when the strain of crystal is increase. It means that ZnO film is grown at plasma power of 40 W has better crystalline degree than others. The optical properties of ZnO thin films have been analyzed using room temperature UV-vis spectrometer in wavelength range of 200 nm to

1000 nm. Figure 3. shows the transmittance spectra of ZnO thin films deposited at room temperature with variations plasma power. Based on the transmittance spectra, it can be stated that the grown films have a finite potential barrier and showed the interference fringe pattern. Interference fringe on the transmittance spectra indicated that the surface of film has a high reflectivity level therefore when incident photon come to surface film it is not too much scattered or slightly absorbed. It can also be seen that transmittance spectra of all films is not uniform. It is due to the differences of surface topography of each grown film therefore the interference of reflected light on the surface each film is also different. Moreover, transmittance spectra of all films decreased sharply in the ultraviolet region (~ 280 nm) which is associated to the absorption of electrons in the valence band. All samples showed the transmittance values of 76% to 92% with the highest transmittance at the plasma power of 40 W. Increasing of plasma power leads to the increasing of films' transmittance too. Transmittance value of ZnO films in this study is in good agreement to the experiment results of other researchers that previously reported [1-3].

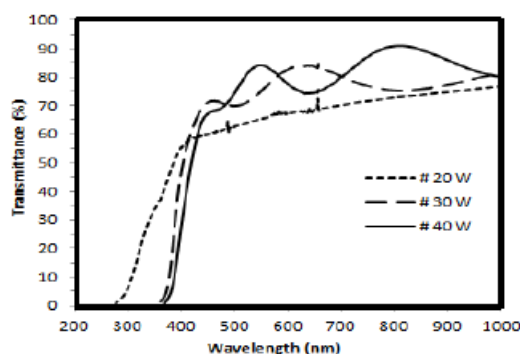


Figure 3. Transmittance spectra of ZnO thin film at various plasma power.

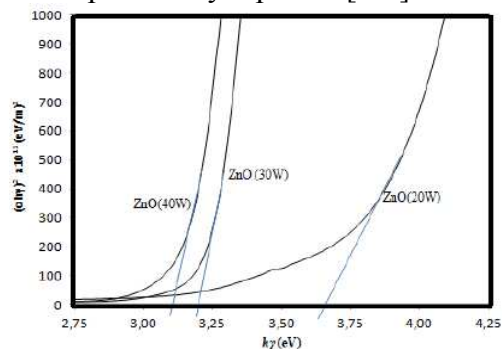


Figure 4. Relation $(ah)^2$ with the bandgap energy ($h\gamma$) ZnO films deposited at room temperature with a plasma power of 20, 30, 40W.

Figure. 4 shows ZnO thin films deposited at room temperature with a plasma power of 20 W, 30 W and 40 W, respectively. The energy gap of those films is 3.65 eV, 3.20 eV, and 3.10 eV, respectively. It clearly be seen that ZnO film grown at plasma power of 20 W has energy gab of 3.65 eV that is considered correlate to the energy gab of glass substrate [4-7]. Therefore, this result confirms the XRD result that ZnO film that grown at plasma power of 20 W did not deposited on the surface of substrate yet. The shrinkage of the energy gap of films was considered due to the large value of crystallite size and small lattice strain as the observed by X-ray diffraction previously. It can be noticed that the energy gap of grown films are qualifies as a transparent conducting oxide (TCO) because it value above 3.10 eV.

Table 1. The electrical resistivity and conductivity of ZnO thin films deposited by using DC magnetron sputtering.

Number	P (mTorr)	Plasma power (watt)	Resistivity (Ωcm)	Conductivity (Ωcm) ⁻¹
1	500	20	8.64×10^5	1.16×10^{-6}
2	500	30	2.49×10^6	4.02×10^{-7}
3	500	40	1.21×10^6	8.92×10^{-7}

The electrical resistivity of the grown films were analyzed by using I-V measurement. The electrical resistivity of ZnO thin films is inversely proportional to the its conductivity. The electrical conductivity of ZnO thin films arise due to the presence of non-stoichiometric composition caused by excess zinc ions (Zn^{2+}) [9-10]. Because the crystal electricity is neutral so that the two abundance of cations in the Zn^{2+} configuration will be offset by the two negative charge of electrons [9]. Then, these electrons are able to move freely in the crystal configuration under influence of the external field. The electrical resistivity and conductivity of ZnO thin films is shown Table 1. Increasing the plasma power from 30 W to 40 W leads to decreasing the electrical resistivity of

films. Consequently, the electrical conductivity of films is increase. Barker, et al (1997) reported that electrical resistivity of ZnO deposited by rf method magnetron sputtered at room temperature was $3 \times 10^7 \Omega\text{cm}$ [2], while Tokumoto, et al (2002) reported that electrical resistivity of ZnO thin film synthesized by pyrosol process at room temperature was around $10^{12} \Omega\text{cm}$ [3]. Therefore it can be noted that the electrical conductivity of ZnO thin film growth by using our method is better.

Summary

ZnO thin films have successfully been deposited by using DC magnetron sputtering at room temperature by means of plasma power variation. XRD result showed that ZnO films were grown at plasma power of 30 and 40 W are polycrystalline; while at plasma power of 20 W is considered do not growth yet. Transmittance performance of films at the wavelength of higher than 400 was 76-92%. The broadest transmittance range was belongs to ZnO film growth at plasma power of 40 W. The optical bandgap of films were shrinkage by increasing of plasma power. The electrical conductivity of ZnO films was increase from $4.02 \times 10^{-7} (\Omega\text{cm})^{-1}$ to $8.92 \times 10^{-7} (\Omega\text{cm})^{-1}$ when the plasma power was increased. Based on the electrical and optical properties of the films it clearly be seen that grown film are suitable for transparent conductive oxide (TCO).

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