Journal of Physics: Conference Series

PAPER • OPEN ACCESS

Growth of *a*-axis-oriented Al-doped ZnO thin film on glass substrate using unbalanced DC magnetron sputtering

To cite this article: D Aryanto *et al* 2019 *J. Phys.: Conf. Ser.* **1191** 012031

View the article online for updates and enhancements.

You may also like

- Improved Gas Sensing Performance of ALD AZO 3-D Coated ZnO Nanorods
P. Lin, X. Chen, K. Zhang et al.

- High-density plasma etching characteristics of aluminum-doped zinc oxide thin films in Cl_2/Ar plasma
Liting Zhang, Young-Hee Joo, Doo-Seung Um et al.

- Tunable physical properties of Al-doped ZnO thin films by O_2 and Ar plasma treatments
Young-Hee Joo, Doo-Seung Um and Chang-Il Kim

244th Electrochemical Society Meeting

October 8 – 12, 2023 • Gothenburg, Sweden

50 symposia in electrochemistry & solid state science

Deadline Extended!
Last chance to submit!

New deadline: April 21
submit your abstract!

This content was downloaded from IP address 103.23.101.10 on 10/04/2023 at 08:24

Growth of *a*-axis-oriented Al-doped ZnO thin film on glass substrate using unbalanced DC magnetron sputtering

D Aryanto[1,*], P Marwoto[2], T Sudiro[1], A S Wismogroho[1] and Sugianto[2]

[1] Research Center for Physics, Indonesian Institute of Sciences, Setu, Tangerang Selatan 15314, Banten, Indonesia
[2] Physics Department, Faculty of Mathematics and Science, Universitas Negeri Semarang, Gunungpati, Semarang 50229, Jawa Tengah, Indonesia

*E-mail: didi027@lipi.go.id

Abstract. Al-doped ZnO (AZO) thin films have been deposited on glass substrates using home-made DC magnetron sputtering at room temperature. The effects of Al-doping and sputtering power on the morphology and structure of AZO thin films were studied. X-ray diffraction shows that all AZO thin films have a polycrystalline nature with a hexagonal wurtzite structure. The preferential orientation planes of (100) and (110) AZO thin films were obtained by adding 2 at% Al-doping and prepared using plasma powers of 30 and 40 Watts. Texture coefficient (*TC*), crystalline size (*D*), lattice strain (ε), and lattice constant of the (100) and (110) planes of AZO thin film were calculated. Increasing the plasma power leads to the enhancement of crystal quality, crystalline size and grain size of *a*-axis orientation. Atomic force microscopy analysis shows that the surface of AZO thin film becomes smoother with increasing the grain size. The growth parameters in the deposition of thin films using sputtering technique have significant effect on morphology and structure of the films.

1. Introduction

Aluminium-doped zinc oxide (AZO) thin film is one of ZnO-based materials that has hexagonal wurtzite structure with a wide direct band gap [1]. The AZO thin film has high electrical resistivity and transparency in visible and infrared wavelength [2, 3]. In addition, AZO thin film has high chemical stability, high mechanical stability, and non-toxic and the raw materials are available in abundance [4]. These characteristics are suitable for applications in optoelectronic devices such as light-emitting diodes, flat panel displays, and thin-film solar cells [5]. Many deposition techniques have been used to fabricate AZO thin films, including sol-gel technique [6], spray pyrolysis [7], sputtering [4], pulsed laser deposition (PLD) [8], and metal organic chemical vapour deposition (MOCVD) [9]. Generally, AZO thin films are grown in polar (*c*-axis oriented) and non-polar (*a*-axis oriented) planes [10]. Mostly, however, all works reported the formation of AZO thin films with *c*-axis orientation due to the fact that the material with wurtzite structure has a high growth rate along the *c*-axis.

In the fabrication of light-emitting devices based on ZnO thin films, the *c*-axis-oriented ZnO thin films on the variety of substrates and heterostructures were grown using *c*-axis-oriented ZnO active layer. It generated the spatial separation of electrons and holes in the active layer, causing a reduction in quantum efficiency [10]. Waltereit et al. [11] reported that non-polar layer surface in the light

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd

emitting device structures can improve quantum efficiency. Therefore, many works were carried out to fabricate ZnO thin film with a non-polar orientation, which is expected to increase the ultraviolet light emitting efficiency for ZnO-based heterostructures.

Growth of *a*-axis-oriented ZnO thin film has already been demonstrated using various techniques, such as spray pyrolysis [12], sol gel methods [10], PLD [13], metal organic aerosol deposition [14], single-source chemical vapour deposition (SSCVD) [15], and RF magnetron sputtering [16]. In this paper, we report the deposition and characterization of preferential *a*-axis-oriented polycrystalline AZO thin films on glass substrate by unbalanced DC magnetron sputtering which has not been studied.

2. Experimental Methods

The AZO thin films were deposited on glass substrates using unbalanced DC magnetron sputtering. AZO target was made from high purity ZnO powder (99.999%) doped with desired amount of high purity Al_2O_3 powder (99.999%). The amount of Al_2O_3 added to the ZnO was 1 to 2 at%. Prior to the deposition, the glass substrate was cleaned in an ultrasonic bath with acetone, methanol, and deionized water. After that, substrate was dried using O_2 gun and mounted in the sputtering chamber. Before the deposition, the target was pre-sputtered for about 10 minutes to remove contaminants from the surface. The chamber was initially evacuated to a base pressure of 1×10^{-4} Torr and the deposition was carried out at a working pressure of 4×10^{-1} Torr. The sputtering was carried out at room temperature with high-purity (99.999%) Ar gas as a plasma source. The plasma power was used in the range of 30 to 40 Watts. The distance between target and substrate was kept constant at 45 mm. X-ray diffraction (XRD) and atomic force microscopy (AFM) were used to analyze the structural properties and surface morphology of AZO thin films. The crystalline quality, preferred orientation, and surface morphology of AZO thin films were clarified and discussed.

3. Results and Discussion

The XRD patterns of undoped and Al-doped ZnO thin film recorded in the range of 30-60° are shown in Figure 1. The result shows that all films have a polycrystalline with a wurtzite-type hexagonal structure. No phases related to Al, Al_2O_3, or other impurity phases were found in the AZO thin films within the XRD detection limits. This indicates that Al^{3+} substituted the Zn^{2+} sites or incorporated into interstitial sites of ZnO lattice [17]. As seen in figure 1, the undoped ZnO thin film has a high preferential orientation along the (002) plane. Other diffraction peaks, (100) and (110), with weak intensity were also observed. This indicates that the undoped ZnO thin film exhibits a strong *c*-axis orientation perpendicular to the substrate [18]. The intensity of the (002) diffraction peak decreases and the full width at half maximum (FWHM) of the peak increases at the doped ZnO thin film with 1 at% Al. It suggests that the crystallinity of thin film is deteriorated. Ion Al^{3+} reduces the concentration of the zinc interstitial [19], resulting in suppression of the ZnO grain growth and degradation of the crystal quality. In addition, the intensity of (101) peak appears on the ZnO thin films doped with 1 at% Al. This indicates that some grains grow in other orientations besides the *c*-axis orientation [20]. The significant results are shown in the ZnO thin film with 2 at% Al doping that the intensity peaks of (100) and (110) planes increase and the diffraction peak of (002) plane is not observed. This confirms the improvement of crystal quality of *a*-axis orientation owing to the decrease in the corresponding of FWHM (as shown in figure 1). This result also shows that the grain of thin films grew parallel to the substrate surface. The Al doping causes the decrease in the degree of crystallinity of ZnO thin films [21]. The increase of Al dopant concentration is the reason for a complete change of the preferential orientation of the grains. In the sputtering technique, the crystal orientation of the interfacial grain is less dependent on the growth parameters.

Previous study shows that the intensity ratio of thin films indicates the preferential growth of the films in a particular plane [17]. According to the literature, the preferred orientation of (h k l) plane can be defined as a texture coefficient (TC) (h k l), which is calculated using the following formula [12]

$$TC(hkl) = \left(\frac{I_{(hkl)}/I_{r(hkl)}}{\left[(1/n)\sum I_{(hkl)}/I_{r(hkl)}\right]} \right) \quad (1)$$

where $I_{(hkl)}$, $I_{r(hkl)}$, and n indicate the X-ray diffraction intensities which obtained from the films, intensity of the reference diffraction pattern (ICDD-01076-0704), and considered number of diffraction peaks, respectively. The calculation results of texture coefficient for crystal planes of AZO thin films are shown in the Table 1. The higher value of the texture coefficient of ZnO and AZO thin films is obtained in different diffraction plane. This indicates that the preferred orientation of the film changes with increasing the amount of Al-doping. Rao and Santhoshkumar [12] confirm that the higher value of the texture coefficient indicates the preferred orientation of the films along the diffraction plane. The change in the preferred orientation is possible at the growth of thin film on amorphous substrate.

The nucleation and final growth orientation are believed to affect the crystal orientations. The growth orientation is developed into the crystallographic direction of the lowest surface energy. The grains became larger for the film that grows with a lower surface energy density [12]. This result confirms that the growth parameter in the sputtering system affects the crystal orientations.

The detail of the observed peak intensities from (100) and (110) planes of undoped ZnO and AZO thin films is shown in figure 1. Small shifts toward larger angles are shown on both peak positions. It suggests a decrease of the *a*-lattice parameters of AZO thin films. According to Bragg's Law ($n\lambda = 2d \sin\theta$), the change of lattice parameter d causes a shift of θ when λ is constant [17] and it is possible that the change relates to the strain relaxation in the films. It can be confirmed by the calculation of crystallographic parameters, such as average size of crystalline grain (D), lattice strain (ε), and lattice

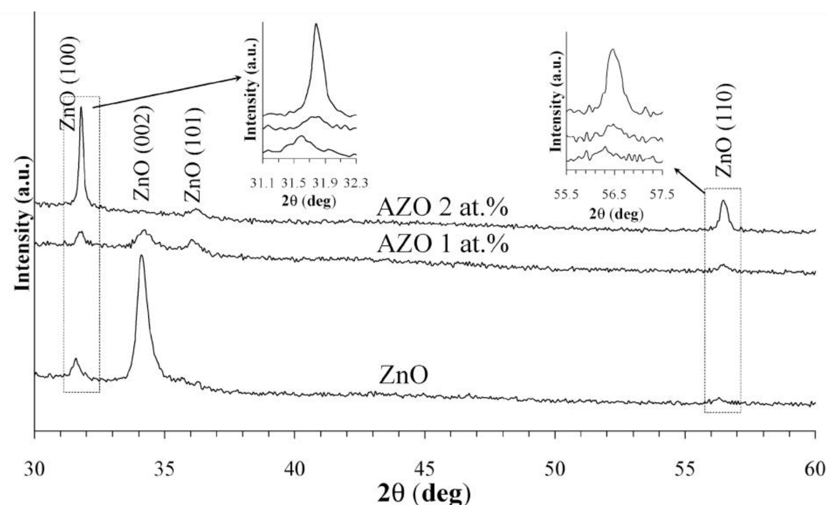

Figure 1. XRD patterns of Al-doped ZnO thin films growth at sputtering power of 30 Watt

Table 1. Texture coefficient of ZnO thin film with variation of Al-dopant.

Al-dopant	Texture coefficient (*TC*)			
	(100)	(002)	(101)	(110)
0	0.54	2.28	-	0.18
1%	1.22	1.73	0.56	0.49
2%	1.93	-	0.20	0.87

constant (*a*). The average size (*D*) of the crystalline grain along the *a*-axis was determined using Debye-Scherrer's formula [12].

$$D = \frac{0.9\,\lambda}{\beta\cos\theta} \tag{2}$$

The lattice strain (ε) was calculated using the tangent formula [22]

$$\varepsilon = \frac{\beta}{(4\tan\theta)} \tag{3}$$

The lattice parameters (*a*) of (100) and (110) plane were determined using the equation

$$a = \frac{\lambda}{\sqrt{3}\sin\theta}\sqrt{h^2 + hk + k^2} \tag{4}$$

where λ is the *X*-ray wavelength of 1.5418 Å, θ is Bragg diffraction angle of *a*-axis peak and β is the FWHM of θ. The crystallographic parameters of AZO thin films growth at sputtering power of 30 Watt are calculated according to (100) and (110) planes and tabulated in Table 2. It is observed that the lattice parameter (*a*) of AZO thin films decreases compared to the ZnO thin film. These results confirm that the shifted angles are affected by *a*-lattice parameters. The replacement of Zn^{2+} ions by Al^{3+} ions also contributes to that case due to the different in radius of Zn^{2+} ions (0.0740 nm) and Al^{3+} ions (0.0535 nm)[17]. However, the addition of Al doping also influences the crystalline size of thin films. Compared to the ZnO thin films, the crystalline size of AZO thin films is smaller. Similar result is shown by Sahay and Nath [22] and Chen et al. [17] that the crystalline size decreases with increasing Al-doping.

Figure 2 shows the XRD patterns of ZnO and AZO thin films at the sputtering power of 40 Watt. It can be seen that two dominant peaks are observed in XRD pattern corresponding to (100) and (110) planes. This result indicates that the film has a polycrystalline hexagonal wurtzite structure with *a*-axis orientation. The crystallographic parameters of AZO thin films growth at sputtering power of 40 Watt are calculated and tabulated in Table 3. However, calculation of texture coefficient shows that the preferred orientation of ZnO and AZO (2 at% Al) thin films changes from (100) to (110) planes compared to ZnO and AZO (2 at% Al) at the plasma power of 30 Watt. In this case, the increase of sputtering power from 30 to 40 Watt influences the dominant of crystal growth in the (110) plane. The intensities of the (100) and (110) diffraction peaks increase and shift to larger angle with the addition of 2 at% Al. It further suggests that the improvement in the crystallinity and the value of *a*-lattice parameter occur. The increase of the peak intensity is also influenced by the grain and crystalline sizes [23]. The ions or ion clusters can obtain more energy with increasing the plasma power. It causes ions or ion clusters to easily adjust their own bond direction and length, so that they obtain an optimum bonding to the adjacent atoms and this helps them to nucleate and grow. Therefore, the crystalline size of AZO (2 at% Al) thin films becomes larger with the sputtering power of 40 Watt. Table 2 and 3 also show that the *a*-lattice parameters decrease with the increase of plasma power. This may be due to the interstitial and substitution of Al atoms or ions in the lattice of AZO thin films [24].

Table 2. Calculated crystalline size, lattice strain, and lattice constants from (100) and (110) planes of ZnO thin film with various Al-doping.

Al-doping	(*h k l*)	FWHM	*D* [Å]	ε [%]	*a* [Å]
0	(100)	0.28	5.219	0.244	3.2686
	(110)	0.30	5.246	0.140	3.2674
1%	(100)	0.35	4.123	0.307	3.2465
	(110)	0.40	3.938	0.186	3.2586
2%	(100)	0.20	7.214	0.097	3.2514
	(110)	0.33	4.750	0.154	3.2586

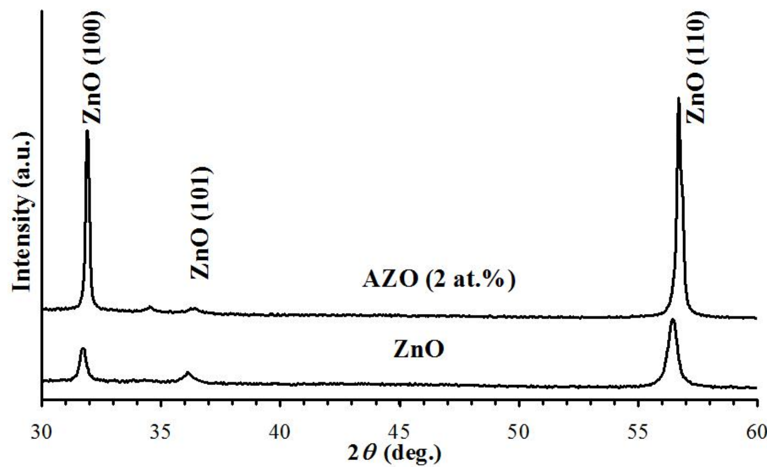

Figure 2. XRD patterns of Al-doped ZnO thin films growth at sputtering power of 40 Watt.

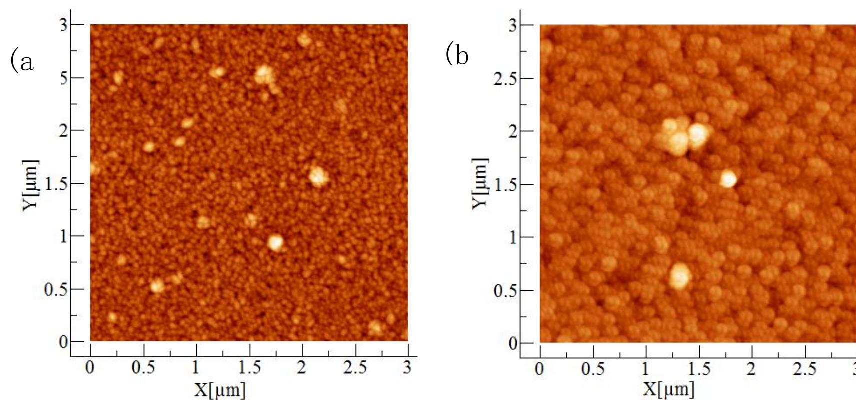

Figure 3. AFM images of AZO (2 at%) thin films growth with the sputtering power of (a) 30 Watt and (b) 40 Watt.

Table 3. Calculated texture coefficient, crystalline size, lattice strain, and lattice constants from (100) and (110) planes of ZnO and Al-doped ZnO (2%) thin film at the sputter power of 40 Watt.

Al-dopant	(*h k l*)	TC	FWHM	*D* [Å]	*ε* [%]	*A* [Å]
0	(100)	0.50	0.30	4.809	0.264	3.2520
	(110)	1.49	0.44	3.580	0.205	3.2586
2%	(100)	0.65	0.22	6.527	0.194	3.2410
	(110)	1.34	0.28	5.705	0.128	3.2469

Figure 3 shows the AFM images of AZO (2 at% Al) at plasma power of 30 and 40 Watts. It can be seen that the film grows with granular grain. The surface morphology of the AZO (2 at% Al) thin films deposited at 30 Watt shows a coarse surface with non-uniform in grain size. The grain size of the AZO (2 at% Al) thin films becomes larger, dense, and uniform in size when the plasma power was increased to 40 Watt. The root mean square (RMS) of roughness for plasma power of 30 and 40 Watts of AZO thin film was 8.546 and 6.999 nm, respectively. This indicates the enhancement in the migration of sputtered atoms on the thin films surface during the growth and the surface roughness reduction. Another reason why the RMS roughness reduces is due to the energy of particles arriving at

the surface increases with increasing plasma power, leaving more energy for surface diffusion caused by collisions with more atoms. The result of AFM analysis is in good agreement with the results of XRD analysis.

4. Conclusions

The AZO thin films have been deposited on glass substrate using unbalanced DC magnetron sputtering system with various doping concentration and plasma power. The films have polycrystalline structure with preferred orientation of (100) and (110). The crystallinity and the a-axis-oriented AZO (2 at% Al) thin films prepared at 40 Watt power plasma give better film quality compared to others sample. The doping concentration and plasma power strongly influence the growth of thin film as well as structure and morphology of the films.

Acknowledgments
This work was supported Ministry of Research, Technology and Higher Education, Indonesia under Grant No. 013/P/RPL-LIPI/INSINAS-2/VII/2018 and 158/P/RPL-LIPI/INSINAS-1/III/2018. The authors are grateful to Laboratory of Thin Film, Physics Department, Universitas Negeri Semarang and Research Center for Physics-LIPI for providing the research facilities.

References

[1] Jeong S H, Park B N, Yoo D G and Boo J H 2007 *J. Korean Phys. Soc.* **50** 622
[2] Chandramohan R, Vijayan T A, Arumugam S, Ramalingam H B, Dhanasekaran V, Sundaram K and Mahalingam T 2011 *Mater. Sci. Eng. B* **176** 152
[3] Kodu M, Arroval T, Avarmaa T, Jaaniso R, Kink I, Leinberg S, Savi K and Timusk M 2014 *Appl. Surf. Sci.* **320** 756
[4] Liu C, Xu Z, Zhang Y, Fu J, Zang S and Zuo Y 2015 *Mater. Lett*. **139** 279
[5] Eshaghi A and Hajkarimi M 2014 *Optik* **125** 5746
[6] Mahroug A, Boudjadar S, Hamrit S and Guerbous L 2014 *Mater Lett* **134** 248
[7] Gahtar A, Rahal A, Benhaoua B and Benramache S 2014 *Optik* **125** 3674
[8] Taabouche A, Bouabellou A, Kermiche F, Hanini F, Sedrati C, Bouachiba Y and Benazzouz C 2016 *Ceram. Int.* **42** 6701
[9] Su J, Tang C, Niu Q, Zang C, Zhang Y and Fu Z 2012 *Appl. Surf. Sci.* **258** 8595
[10] Bhole M P and Patil D S 2007 *Optoelectron. Adv. Mater.* **1** 672
[11] Waltereit P, Brandt O, Ramsteiner M, Trampert A, Grahn H T, Menniger J, Reiche M and Ploog K H 2001 *J. Cryst. Growth* **227-228** 437
[12] Rao T P and Santhoshkumar M C 2009 *Appl. Surf. Sci.* **255** 7212
[13] Nistor L C, Ghica C, Matei D, Dinescu G, Dinescu M and Tendeloo G V 2005 *J. Cryst. Growth* **277** 26.
[14] Neumann U, Grunwald R, Griebner U and Steinmeyer G 2005 *Appl. Phys. Lett.* **87** 171108-1
[15] Dai L P, Deng H, Chen G, Teng C F, Wei M and Li Y 2007 *Vacuum* **81** 969
[16] You J B, Zhang X W, Cai P F, Dong J J, Gao Y, Yin Z G, Chen N F, Wang R Z and Yan H 2009 *Appl. Phys. Lett.* **94** 262105-1
[17] Chen J, Chen D, He J, Zhang S and Chen Z 2009 *Appl. Surf. Sci.* **255** 9413
[18] Zhao L, Shao G-J, Qina X-J and Han S-H-Z 2011 *J. Alloys Compd.* **509** L297
[19] Wang M, Lee K E, Hahn S H, Kim E J, Kim S, Chung J S, Shin E W and Park C 2007 *Mater. Lett.* **61** 1118
[20] Li J, Xu J, Xu Q and Fang G 2012 *J. Alloys Compd.* **542** 151
[21] Sandeep K M, Bhat S and Dharmaprakash S M 2017 *J. Phys. Chem. Solids* **104** 36.
[22] Sahay P P and Nath R K 2008 *Sens. Actuators B-Chem.* **134** 654
[23] Purohit A, Chander S, Sharma A, Nehra S P and Dhaka M S 2015 *Opt. Mater.* **49** 51
[24] Kuo S-Y, Liu K-C, Lai F-I, Yang J-F, Chen W-C, Hsieh M-Y, Lin H-I and Lin W-T, *Microelectron. Reliab.* **50** 730