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Effects of Growth Temperature on Morphological and Structural Properties of ZnO Films

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ABSTRACT

Zinc oxide (ZnO) is one of the most promising oxide possibilities for use in a number of industries due to its unique properties. Because of its broad direct bandgap (3.37 eV) and strong exciton binding energy (60 meV) at ambient temperature, ZnO not only conducts electricity well but also transmits visible light and emits UV light. Here, we investigated the effect of growth temperature on ZnO thin films by changing the growth temperatures from 400 °C to 450 °C. Radio-frequency (RF) magnetron sputtering was used to create ZnO thin films on Si(100) substrates. The atomic force microscopy (AFM) results show that the root-mean-square (RMS) roughness decreases from 6.1 ± 1.0 nm to 4.8 ± 0.6 nm as the growth temperatures increase. The crystalline structures of ZnO films strongly depend on the growth temperature. Our findings indicate that controlling the growth temperature is the critical factor in producing high-quality ZnO thin films.

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1. Introduction

ZnO is a II-VI semiconductor compound that has attracted the interest of scientists in a variety of applications, such as blue optoelectronics, ultra-violet (UV) detectors and emitters, transparent conducting electrodes and chemical and biosensors owing to its properties [1] - [3]. ZnO exhibits a wide direct bandgap of 3.37 eV and large exciton energy of 60 meV at room temperature. Moreover, ZnO has important optical properties such as good visible light transmission, UV light emission and lasing action [2], [4], [5]. Therefore, ZnO has emerged as one of the most promising oxide materials and has been studied in different nanostructures such as nanoparticles, nanotubes, nanorods, and thin films to apply for various purposes in life.

Recently, ZnO thin film has been deposited on silicon substrate for NO_2 and glucose sensors [6, 7], Schottky photodiodes [8-10], piezoelectric generators [11] - [12] and solar cells [13] - [14] applications. However, using ZnO thin films in these devices is difficult because achieving a high-quality ZnO thin film on a silicon substrate is difficult due to lattice mismatch. The differences in lattice constants and crystal structure between the silicon substrate (cubic, a_{Si} = 0.543 nm [15]) and ZnO thin film (hexagonal, a_{ZnO} = 0.325 nm , c_{ZnO} = 0.521 nm [1]) strongly affect the microstructure and morphology of ZnO thin film. Consequently, it will decrease the performance of electronic devices.

Growth temperature provides energy for atoms to occupy the preferential sites. By controlling the growth temperature, ZnO thin films will have differences in crystallinity and surface properties [16-18], stoichiometry (Zn-rich or O-rich) and native defects (oxygen vacancy, zinc vacancy, oxygen interstitial, zinc interstitial) [19]. Moreover, the crystalline quality of ZnO films strongly depends on the deposition method. From the literature, the crystalline structure of ZnO films was decreased with the growth temperature of 600 °C by using the physical vapor deposition method [20] - [21]. ZnO thin films were grown by e-beam evaporation in the range from room temperature to -120 °C [22]. In this work, the RF-magnetron sputtering method was utilized to fabricate ZnO thin films on Si (100) substrates with the growth temperature ranging from 400 °C to 450 °C. The dependence of structural and morphological properties of ZnO thin films on growth temperature was mainly studied.

2. Experimental

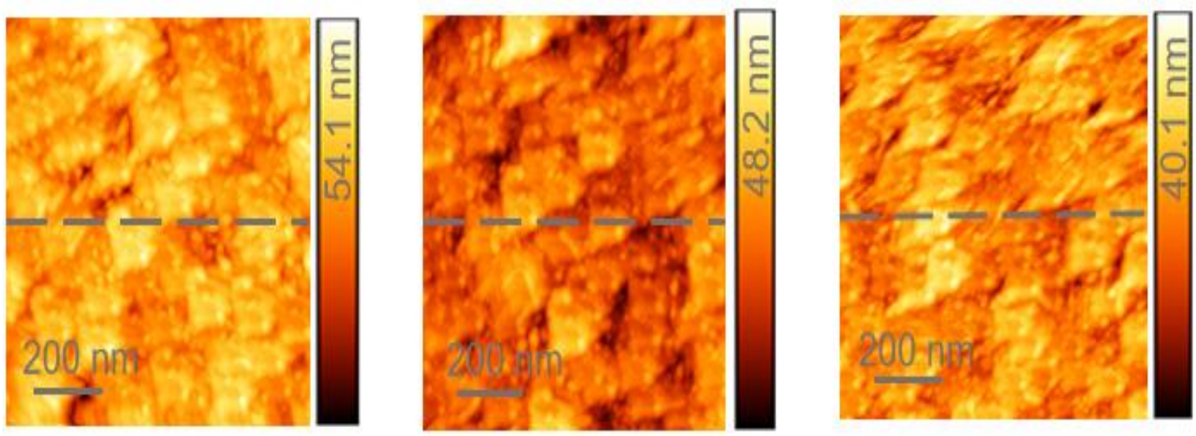
ZnO thin films were prepared by radio frequency (RF)-magnetron sputtering on Si(100) substrates by using ZnO target. The Si(100) substrate was cleaned by rinsing in deionized water (DI), isopropyl alcohol (IPA), H_2SO_4 5%, and HF 20% solution. Si(100) substrate was pre-heated in the vacuum chamber at a pressure of 7.5×10^{-6} Torr to remove contamination and water vapor still clinging to the substrate's surface. The ZnO thin films were deposited on Si(100) substrate at various growth temperatures: 400 °C (Tg400), 425 °C (Tg425) and 450 °C (Tg450).

X-ray diffraction (XRD) was used to analyze the crystalline structure of ZnO thin films. The morphology of ZnO samples was investigated by Atomic Force Microscopy (AFM). The surface properties can be obtained by using ScanAsyst in air mode with a scanning side of 1 µm × 1 µm and a scan rate of 0.7 Hz.

3. Results and discussions

To understand the effect of the growth temperature on the morphology of ZnO samples, the AFM scan was used to measure the root-mean-square (RMS) roughness of ZnO films. The AFM scans of ZnO samples (Tg400, Tg425 and Tg450) are shown in Fig. 1. ZnO thin films exhibit large platelet-like arrays and uneven tiny grains. When the growth temperature increases, the number of small grains decreases and the arrays are more closely packed (see Fig. 1). Sample Tg400 has the roughest RMS roughness, with a RMS roughness of 6.1 ± 1.0 nm, while the RMS roughness values of samples Tg425 and Tg450 decrease to 5.4 ± 1.2 nm and 4.8 ± 0.6 nm. Table 1 summarizes the results obtained using AFM scan for three different growth temperatures of ZnO thin films.

Fig. 2 represents the profile height of samples Tg400, Tg425 and Tg450. All the profile heights of ZnO thin films exhibit many little splitting peaks. The splitting peak phenomenon in these samples shows the existence of many small grains on the platelet-like arrays. These tiny grains are also uneven, which is illustrated by the irregularity of the splitting peaks of the line profile. The peaks of sample Tg400 are the highest compared to the remaining ZnO samples, up to 48 nm high (see Fig. 2a). The high RMS roughness value of 6.1 ± 1.0 nm can result from the high and irregular peaks of ZnO thin films grown at 400 °C.



(a) *(b)* *(c)*

Fig 1. *AFM scans (1µm × 1µm) of ZnO samples (a): Tg400; (b): Tg425 and (c): Tg450. The black dashed line in each AFM scan at 0.5 µm represents the profile height (Fig. 2).*

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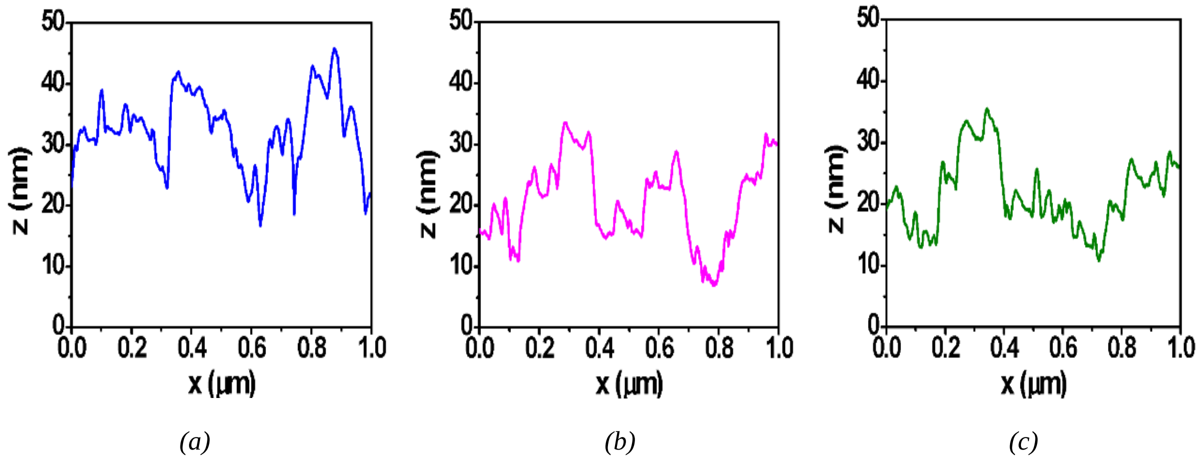

Fig 2. *The profile height of samples (a): Tg400; (b): Tg425 and (c):Tg450.*

Table 1. *Morphology analysis of the AFM scans for samples Tg400, Tg425 and Tg450.*

Samples	Growth temperature (°C)	RMS Roughness (nm)	Peak-to-valley height (nm)
Tg400	400	6.1 ± 1.0	29.1 ± 5.4
Tg425	425	5.4 ± 1.2	26.3 ± 5.4
Tg450	450	4.8 ± 0.6	24.9 ± 3.3

After the morphology characterization, the structural properties of ZnO thin films were measured by using XRD. Fig. 3 displays XRD patterns of samples Tg400, Tg425, and Tg450, respectively.

Tg400, Tg425 and Tg450 samples all exhibit various diffraction peaks such as ZnO(100), ZnO(002), ZnO(103), and ZnO(004). These peaks of ZnO thin films go in line with the hexagonal wurtzite phase [16, 23]. The FWHM values of the diffraction peaks for the (002) plane for samples Tg400 and Tg425 are 0.236° and 0.251°, respectively, which are larger than that of sample Tg450 (0.226°) (see Table 2). The FWHM of ZnO samples reveals that the crystallinity of ZnO thin films is enhanced by increasing the growth temperatures. The growth temperature obviously helps to increase the crystalline quality of ZnO thin films, which agrees with previous reports [16] - [18]. The increased crystallinity of ZnO thin films due to the growth temperature can be explained by the atomic mobility [19]. At higher growth temperatures, the thermal energy increases the mobility of the atoms and helps them occupy the preferential position for good crystallinity [24].

Moreover, there is a shift of the ZnO(002) peaks to higher angles than the bulk values (34.4°) [18] (Table 2). The interplanar spacing of plane (*d*-spacing) values of ZnO thin films are figured out by using the Bragg equation [25]:

$$2d\sin\theta = n\lambda \quad (1)$$

With θ, n, λ is corresponding to diffraction Bragg angle, diffraction order (n = 1), and x-ray's wavelength (λ = 1.54 Å).

When the growth temperature increases, the *d*-spacing values decrease from 2.600 to 2.593 Å, indicating the presence of compressive strain in ZnO thin films [26]. These strains are the reason for the shift of the ZnO (002) peaks of ZnO thin films to higher positions [27].

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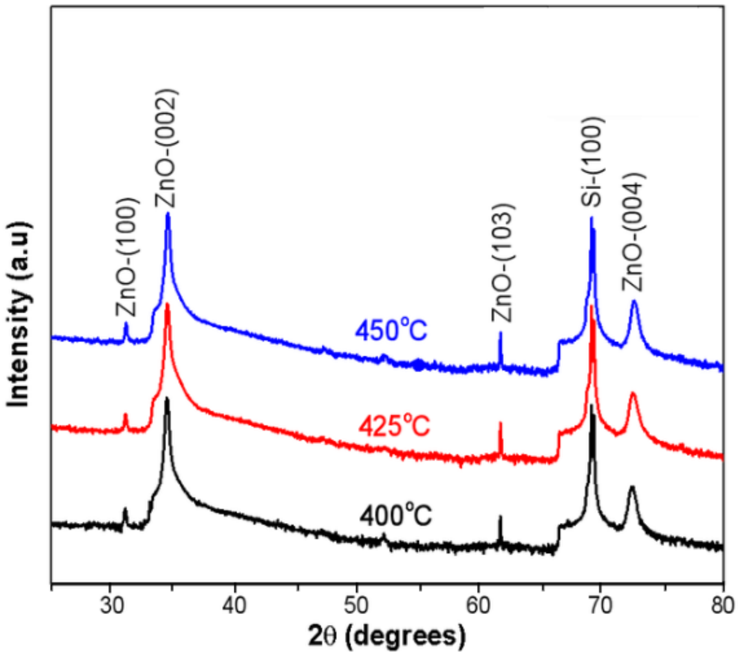

Fig 3. *XRD spectra of samples Tg400, Tg425, and Tg450.*

Table 2. *Information about structural properties obtained from the XRD spectra.*

Samples	(h k l)	(002) peak information		FWHM (°)	*d*-spacing (Å)
		Experimental (°)	Bulk (°)		
Tg400	(0 0 2)	34.46	34.4	0.236	2.600
Tg425	(0 0 2)	34.49	34.4	0.251	2.598
Tg450	(0 0 2)	34.53	34.4	0.226	2.595

4. Conclusions

In summary, ZnO films were deposited on Si(100) substrates using RF-magnetron sputtering. The growth temperatures of ZnO thin films range from 400 ºC to 450 ºC. The effect of growth temperature on the microstructure and morphology of ZnO thin films is investigated. The AFM result reveals that the RMS roughness of ZnO thin film deposited at 450 ºC is the smallest and the platelet-like arrays are more densely packed. In addition, XRD patterns exhibit an improvement in the structure of ZnO films. Our results indicate that the growth temperature of 450 ºC is the optimal condition for growing the high-quality ZnO thin films. It will be suitable for application in the fabrication of nanodevices.

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Vo Doan Thanh Truong graduated in Materials Technology from Ho Chi Minh City University of Technology and Education (HCMUTE), Vietnam with a high GPA. Her research focuses on fabricating semiconductor and magnetic thin films using physical deposition techniques and studying the effects of different factors, such as growth temperature, deposition pressure and power on thin films' properties.

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Thi Kim Hang Pham received her MS. degree from the Institute of Physic, Hanoi, Vietnam in 2011 and then achieved a PhD. degree from Ewha University, Korea in 2019. She has a lot of experience in fabricating and characterizing many magnetic and semiconductor materials using physical deposition techniques such as Fe_3O_4, $IrMn_3$, Mn, Si, FeSi, Fe_2O_3, and ZnO.