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Effect on structural, optical and electrical properties of aluminum-doped zinc oxide films using diode laser annealing

Wen-Tse Hsiao [a,*], Shih-Feng Tseng [a], Chien-Kai Chung [a], Donyau Chiang [a], Kuo-Cheng Huang [a], Keh-Moh Lin [b], Liang-Yan Li [b], Ming-Fei Chen [c]

[a] Instrument Technology Research Center, National Applied Research Laboratories, No. 20, R&D Rd. VI, Hsinchu Science Park, Hsinchu City 30076, Taiwan
[b] Department of Mechanical Engineering, Southern Taiwan University, No. 1, Nan-Tai Street, Yungkang Dist., Tainan 71005, Taiwan
[c] Department of Mechatronics Engineering, National Changhua University of Education, No. 1, Jin-De Road, Changhua 50007, Taiwan

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ABSTRACT

This study investigated the laser annealing characteristics of aluminum-doped zinc oxide (AZO) films using a diode laser source (808 nm) combined with moving stage with varying parameters, including laser fluence and speed of moving stage in air atmosphere. The commercial AZO thin films were prepared by RF magnetron sputtering on glass substrates. The films characteristics were systematically analyzed using a field emission scanning electron microscope, an atomic force microscope (AFM), an X-ray diffraction (XRD) equipment, an ultraviolet–visible–near-infrared (UV–vis–NIR) spectrophotometer, a four points probe instrument, and a Hall effect measurement system. The experimental results indicate that varying the laser fluence and annealing speed affected the optical, electrical, and structural characteristics of the AZO films. After annealing, approximately 90% of transmittance spectra exhibited slight changes in the visible region. All resistivity values of the laser-annealed AZO films decreased substantially from $4 \times 10^{-2}\ \Omega$ cm to $2.8 \times 10^{-2}\ \Omega$ cm. The absorption band edge moved toward shorter or longer wavelengths, depending on the annealing laser fluence and annealing speed. The optical energy band gap of the annealed AZO films increased because the carrier concentration of the annealed AZO films increased. The grain size increased in conjunction with the annealing speed. The AFM-derived root mean square (RMS) values decreased as the annealing speed increased, and the corresponding RMS values ranged from 1.4 to 1.9 nm.

1. Introduction

The demand for digital products, such as multi-touch-screen smart phones, digital cameras, and e-books, has increased, and novel computer, communication, and consumer-electronics products are highly valued items. Display products feature an electrode layer comprising conductive oxide films. However, applying heat treatment to these films is a particularly challenging and crucial step in growing transparent conductive oxide thin films. The advantages of mechanism for laser material processes are including: non-contact processing, no traditional tool wear, high speed and high accuracy processes, and a little absence of residual thermal effect and residual stress; therefore, there are many applications for laser processing such as cutting [1], welding [2,3], marking [4,5], drilling [6], surface treatment [7,8], and so on.

Oh et al. [9] proposed a post-deposition annealing process for annealing aluminum-doped zinc oxide (AZO) films in a hydrogen atmosphere. The annealing temperature and time were 573 K and 10–120 min, respectively. The experimental results indicated that the full-width at half-maximal (FWHM) values of ZnO (002) peaks ranged from 0.27° to 0.25°. The average optical transmittance in the visible light wavelength range exceeded 90%. Moreover, a blueshift in the absorption edges was observed when the annealing time was increased to 60 min. Cho et al. [10] employed rapid thermal annealing to anneal AZO films in an oxygen atmosphere at temperatures ranging from 300 °C to 500 °C. The X-ray diffraction (XRD) intensity of the main peak (002) increased in conjunction with the annealing temperature. Moreover, the FWHM values of the AZO films decreased from 0.477 to 0.258 as the annealing temperature increased from 300 °C to 500 °C. The resistivity of the films decreased as the annealing temperature increased. Yang et al. [11] used annealed AZO films in various atmospheres. The AZO films exhibited strong (002) diffraction peaks at an annealing

* Corresponding author. Tel.: +886 3 5779911x594; fax: +886 3 5773947.
E-mail addresses: wentse@itrc.narl.org.tw (W.-T. Hsiao), tsengsf@itrc.narl.org.tw (S.-F. Tseng), ckchung@itrc.narl.org.tw (C.-K. Chung), dony@itrc.narl.org.tw (D. Chiang), huangkc@itrc.narl.org.tw (K.-C. Huang), kemo@mail.stust.edu.tw (K.-M. Lin), 49814026@stust.edu.tw (L.-Y. Li), chenmf@cc.ncue.edu.tw (M.-F. Chen).

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temperature of 600 °C. The optical transmittance decreased slightly as the annealing temperature increased. Kuo et al. [12] discussed how varying the molar concentration of thin films and the composition of the atmosphere used in the annealing processes affects the characteristics of AZO films. The XRD analysis results indicated that the AZO thin films exhibited the greatest diffraction peak intensity at an annealing temperature of 850 °C. The grain size and sheet resistivity increased in conjunction with the annealing temperature. Lee et al. [13] used a krypton fluoride (KrF) excimer laser with a 248-nm wavelength to anneal AZO films, and then analyzed their structural, surface morphological, and optoelectronic properties. The pulse energy, pulse repetition rate, and pulse numbers were fixed at 160 mJ/cm^2, 10 Hz, and 600–6000 shots, respectively. Scanning electron microscopy was performed to show that the gaps decreased as the temperature increased, and XRD was used to measure the crystalline properties. The results indicated that the pulse number increased in conjunction with the diffraction peak. Tsang et al. [14] used a sol–gel spin-coating method to prepare AZO films on glass substrates. KrF excimer laser irradiation was applied to determine the structural, electrical, and optical properties at various laser energies. When the laser energy was fixed, the diffraction peak value increased in conjunction with the pulse number. The laser energy and number of pulses affected the grain structure. Chen et al. [15,16] used laser-beam shaping technology to anneal indium-doped tin oxide and fluorine-doped tin oxide thin films. The beam profile, energy distribution, overlapping rate, and operation mode affected the optoelectronic properties of the thin films. Hsiao et al. [17] proposed a hybrid processing (i.e., patterning and annealing) for AZO films in a one-step process that involved using a diode pumped solid state ultraviolet laser system. The patterning and annealing parameters (i.e., pulse repetition frequency and speed) influenced the optical and electrical properties of the AZO films. Xu et al. [18] used an Nd:YAG laser with a 1064-nm wavelength for annealing AZO samples under power densities of 18.5, 27.8, 36.4, and 41.8 W/mm^2. After laser-annealing, the carrier concentration, mobility, and resistivity of the AZO films were markedly changed as the laser power density increased. Li et al. [19] employed a nanosecond-pulsed laser with a 532-nm wavelength treatment of AZO films that were deposited on FTO films to improve their photoelectric properties. The experimental results indicated that the laser fluence and scan speed affected the AZO/FTO film properties.

Compared with the above study, this study investigated the effects of laser-annealing on the structural, electrical, and optical properties of AZO films by using a portable diode laser system. The results show that the structural and optoelectronic properties of the AZO films depended on the laser fluence and speed of moving stage. Moreover, the film characteristics were systematically analyzed using a field emission scanning electron microscope (FE-SEM), an atomic force microscope (AFM), an XRD equipment, an ultraviolet–visible–near-infrared (UV–vis/NIR) spectrophotometer, and a Hall effect measurement system.

2. AZO thin film preparation and laser annealing system

Commercial AZO films were deposited on Corning Eagle 2000 glass substrates by using radio-frequency magnetron sputtering. The thickness of the deposited AZO films was approximately 200 nm. To ensure that the dimensions of all specimens were uniform, the glass substrates were diced to dimensions of 30 mm^2 by using a diamond wheel machine. Subsequently, the machined specimens were cleaned in an ultrasonic cleaner with a solution of alcohol (75 vol%) and distilled water (25 vol%).

The surface roughness and morphological views of the as-deposited AZO films on the glass substrates were recorded using an atomic force microscope (AFM, Veeco di Dimension 3100) and FE-SEM (Hitachi S-4300). In addition, the optical transmittance was measured using a UV–vis/NIR spectrophotometer (Jasco V-670). The average transmittance ranged from 400 to 800 nm. The structural properties of the AZO films were measured using XRD equipment (Rigaku D/MAX 2500) with Cu-Kα radiation. Moreover, the electrical properties were measured using the Hall effect measurement system (ECOPIA HMS-3000) and the four point probe instrument (QUATEK, 5601Y). Table 1 summarizes the optical transmittance and electrical properties of the as-deposited AZO films on the glass substrates in this experiment.

A diode laser-annealing system comprising a laser source, laser-beam collimation optics, and dual-axis moving stage was employed for the thermal annealing process. The fiber-optic diode laser source had a wavelength and maximal average power of 808 nm and 30 W, respectively. The laser beam was transmitted using a 400-μm-diameter fiber core with a numerical aperture of 0.22. A cooling system was used to cool the laser source and driving circuits. Table 2 lists the complete specifications of the diode laser-annealing system.

Fig. 1 shows a schematic diagram of the designed anneal path planning process for the AZO films. The annealing area was 40 × 40 mm^2, covering the entire specimen. A positive defocused diode laser beam with a diameter of 3 mm was used to anneal the AZO films. Fig. 2 shows the optical transmittance spectra of the as-deposited and annealed AZO films. Approximately 85.7% of the as-deposited AZO films exhibited transmittance in the visible region.

The diode laser combined with a dual-axis moving stage was employed to anneal the AZO films in ambient air. Adjustment of the annealing parameters is detailed as follows.

To obtain favorable annealing results (i.e., to prevent micro cracks and damage to the substrate), the laser source was fixed in the continuous wave operation mode. The laser fluence varied among 20, 40.1, and 60.1 mJ/cm^2, and the dual-axis moving stage speed varied among 5, 10, and 20 mm/s. To yield favorable annealing quality, a crosshatch path with equal spacing of 1 mm was used for each annealing path.

3. Results and discussion

Fig. 2 shows the transmittance spectra of AZO films under various annealing parameters. Fig. 2(a-1) and (a-2) shows the optical transmittance when the laser fluence was fixed at 20 and 40.1 mJ/cm^2, respectively. The measurement results indicate that the transmittance value decreased as the annealing speed increased. At high annealing

Table 1
Optoelectric properties of as-deposited AZO films on glass substrates.

Substrate	Film thickness (nm)	Transmittance (400–800 nm) (%)	Resistivity (Ω cm)	Surface roughness (Rms) (nm)
Corning Eagle 2000	200	85.7	4×10^{-2}	1.16

Table 2
Specification of the diode laser annealing system.

Parameters	Values
Wavelength (nm)	808
Maximal power (W)	> 30
Spatial mode	TEM_{00}
Beam diameter (mm)	3
Operation mode	Continuous wave