Applied Surface Science 280 (2013) 104–108

Contents lists available at SciVerse ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Study of working pressure on the optoelectrical properties of Al–Y codoped ZnO thin-film deposited using DC magnetron sputtering for solar cell applications

Feng-Hao Hsu[a], Na-Fu Wang[b], Yu-Zen Tsai[b], Ming-Chieh Chuang[b], Yu-Song Cheng[b], Mau-Phon Houng[a,*]

[a] Institute of Microelectronics, Department of Electrical Engineering, National Cheng Kung University, No. 1, Dasyue Road, East District, Tainan City 701, Taiwan
[b] Department of Electronic Engineering, Cheng Shiu University, 840 Chengcing Road, Niaosong District, Kaohsiung City 833, Taiwan

ARTICLE INFO

Article history:
Received 27 August 2012
Received in revised form 19 April 2013
Accepted 23 April 2013
Available online 1 May 2013

Keywords:
Heterojunction solar cell
Electrical properties
Sputtering
Thin-film
Working pressure
ZnO

ABSTRACT

Low cost transparent conductive Al–Y codoped ZnO (AZOY) thin-films were prepared on a glass substrate using a DC magnetron sputtering technique with various working pressures in the range of 5–13 mTorr. The relationship among the structural, electrical, and optical properties of sputtered AZOY films was studied as a function of working pressure. The XRD measurements show that the crystallinity of the films degraded as the working gas pressure increased. The AZOY thin-film deposited at a working pressure of 5 mTorr exhibited the lowest electrical resistivity of $4.3 \times 10^{-4}\ \Omega$ cm, carrier mobility of 30 cm^2/V s, highest carrier concentration of 4.9×10^{20} cm^{-3}, and high transmittance in the visible region (400–800 nm) of approximately 90%. Compared with Al doped ZnO (AZO) thin-films deposited using DC or RF magnetron sputtering methods, a high carrier mobility was observed in our AZOY thin-films. This result can be used to effectively decrease the absorption of near infrared-rays in solar cell applications. The mechanisms are attributed to the larger transition energy between Ar atoms and sputtering particles and the size compensation of the dopants. Finally, the optimal quality AZOY thin-film was used as an emitter layer (or window layer) to form AZOY/n-Si heterojunction solar cells, which exhibited a stable conversion efficiency (η) of 9.4% under an AM1.5 illumination condition.

1. Introduction

In the past few years, transparent conductive oxide (TCO) thin-film has been widely used as a transparent electrode in several optoelectronic devices [1–3]. The most commonly used TCO thin-films include Sn-doped In_2O_3 (ITO) thin-films, F-doped SnO_2 (FTO) thin-films, and Al-doped ZnO (AZO) thin-films. Among these TCO thin-films, AZO thin-films have attracted considerable attention as substitutes for other TCO materials because of their high transmittance in the visible region, low resistivity, low cost and abundancy, nontoxic nature, easy fabrication, and high stability in hydrogen plasma [1,3–6]. AZO thin-films can be prepared using several techniques, such as thermal evaporation, chemical vapor deposition (CVD), sol–gel, spray pyrolysis, pulsed laser deposition (PLD), and magnetron sputtering [6–10]. RF magnetron sputtering using a sintered ceramic target (ZnO:Al_2O_3 (98:2 wt.%)) is the most extensively used method in the industry because it can achieve excellent orientation and uniform films at low substrate temperatures [7,8]. In 2005, GfE Metalle und Materialien GmbH developed a novel low fabrication cost AZOY ceramic target (ZnO containing 2.0 wt% Al_2O_3 and 0.33 wt% Y_2O_3). They indicated that the AZOY ceramic target can be easily deposited using direct current (DC) and pulsed-DC sputtering because of the higher conductivity of the target [9]. In our previous study [10], we used the current DC magnetron sputtering system to prepare AZOY thin-films, and the result showed that these films had higher mobility than that of AZO thin-films because of the higher conductivity of the AZOY target, resulting in fewer arc events in the continuous DC sputtering process. In addition, the faster deposition rate (15 nm/min) for AZOY thin-film deposition is optimal for industry applications. Based on these results, AZOY thin-films may be a superior TCO material for application in various optoelectronic devices. However, no studies have addressed the effects of the working pressure on the properties of AZOY thin-films using the DC magnetron sputtering method. Therefore, in this study, transparent conducting AZOY thin-films were fabricated at various working pressures on glass substrates using DC magnetron sputtering, and the dependence of structural, electrical, and optical properties on the working pressure were investigated. In addition,

* Corresponding author. Tel.: +886 6 2757575x62342; fax: +886 6 2345482.
E-mail address: mphoung@eembox.ncku.edu.tw (M.-P. Houng).

0169-4332/$ – see front matter
http://dx.doi.org/10.1016/j.apsusc.2013.04.103

the AZOY/n-Si heterojunction solar cells were fabricated by sputtering the AZOY film on an n-Si substrate as the emitter layer (or window layer) to confirm that the AZOY film was a promising TCO material for solar cell applications.

2. Experimental

AZOY thin-films were deposited on glass substrates using DC magnetron sputtering with a ZnO ceramic target containing 2.0 wt% Al_2O_3 and 0.33 wt% Y_2O_3. The glass substrates were first cleaned with standard cleaning procedures (acetone and methanol) and subsequently rinsed in deionized water. Before deposition, the sputtering chamber was initially evacuated using a turbo molecular pump to a base pressure of approximately 2×10^{-6} Torr. Prior to film deposition, pre-sputtering was performed for 10 min to remove any contamination on the target surface. A 90 W power sputtering source was used for the AZOY thin-film deposition, and the growth temperature was fixed at 250 °C. To investigate the effects of the working pressure in the AZOY films, the working pressure was varied in the range of 5–13 mTorr with Ar-ambient gas.

An AZOY thin-film of approximately 800 nm was deposited on (1 0 0)-oriented n-type silicon substrate with a resistivity of 1–10 Ω cm for the formation of a heterojunction solar cell. Back contact Al electrodes were thermally deposited with a thickness of approximately 300 nm, and a finger-shape Al layer served as the front electrode.

The thickness of the AZOY thin-films was determined using a profilometer (Ambios Technology, XP-1), and the deposition rate was not varied substantially (approximately 15 nm/min) when the film was deposited at various working pressures. All films were fixed at the same thickness (800 nm) for all measurements. The 3D images and surface root-mean-square (RMS) roughness of AZOY were estimated using atomic force microscopy (AFM, BASO-SPM). The structural properties of the AZOY thin-films were measured using X-ray diffraction (XRD, Philips PW3710). The electrical properties of the AZOY thin-films were obtained using Hall effect measurement (HL 5500IU) using Van der Pauw's method at room temperature. The transmittance spectra of the AZOY thin-films were measured using a UV-Vis spectrophotometer (Hitachi U-4100) in the wavelength from 300 to 900 nm. The photovoltaic characteristics of the device were tested using an AM 1.5 standard Newport #96000 solar simulator (Peccell PEC-L11) with an illumination intensity of 100 mW/cm^2. The current density–voltage (*J*–*V*) characteristics were collected using a Keithley 2400 source-measurement unit.

3. Results and discussion

3.1. Optoelectrical properties of AZOY thin-films deposited on glass substrate

Fig. 1 shows the atomic force microscopy (AFM) 3-D images and surface root-mean-square (RMS) roughness of the AZOY thin-films as a function of working pressures. As shown in the figure, the surface morphology of AZOY grains is found to be continuous and dense. In addition, the surface RMS roughness of the AZOY thin-films increased from 7.51 to 9.39 nm with the increase of working pressure. Fig. 2 shows the X-ray diffraction (XRD) spectra of AZOY thin-films deposited at various working pressures. The XRD measurement showed a strong and narrow ZnO (0 0 2) peak for all investigated samples. In addition, the ZnO (0 0 2) diffraction angle at 34.45° was not significantly dependent on the working pressure, indicating that bombardment of the growing films with high energetic argon ions was irrelevant in the working pressure range [11]. In addition, the intensity of the ZnO (0 0 2) diffraction peak decreased gradually with the increase of the working pressure, and the full width at half maximum (FWHM) of ZnO (0 0 2) diffraction peak increased gradually from 0.21° to 0.24° with the increase of the working pressure. According to the Debye–Scherrer formula [12], the grain sizes of these AZOY thin-films were estimated from 39.6 to 34.7 nm. The result shows that a smaller grain size may be attributed to the lower kinetic energy of sputtered particles. When the films are deposited at higher working pressure, the collision probability between sputtered particles and gas molecules are intensified. The increase of collision probability may decrease the mean free path of the sputtered particles, leading to a reduction of the energy of the particles. Subsequently, the particles have insufficient surface mobility to aggregate and grow. Thus, the crystal growth of films change gradually from 2-D to 3-D growth results to larger surface RMS roughness and smaller grain size [13].

The electrical resistivity (ρ), carrier concentration (n), and carrier mobility (μ) of AZOY thin-films deposited at various working pressures are shown in Table 1. As shown in the table, the carrier

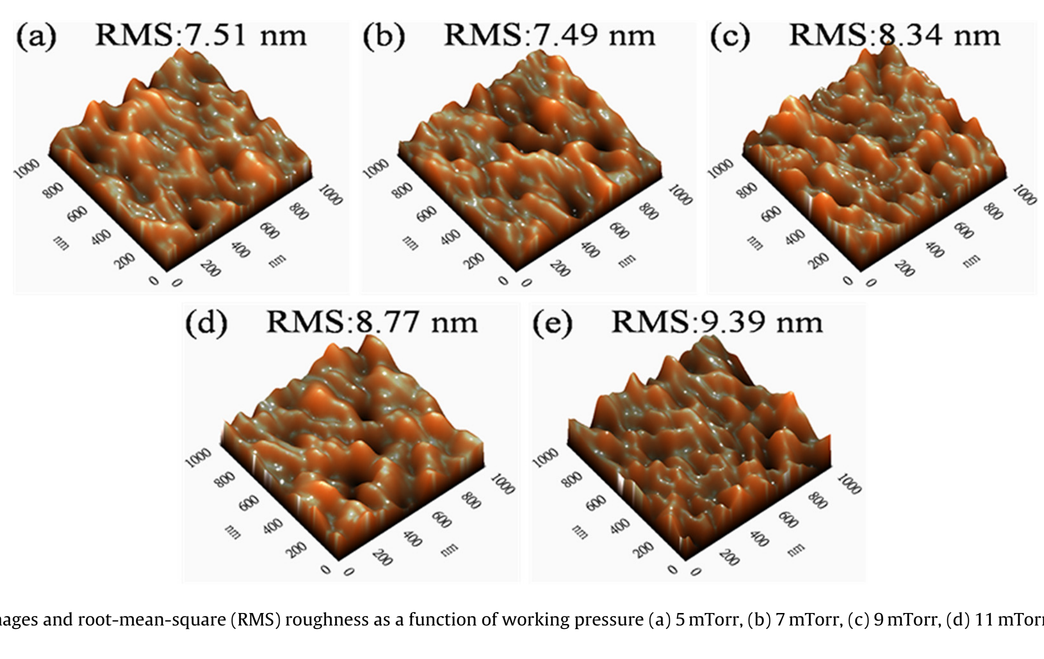

Fig. 1. AFM 3D images and root-mean-square (RMS) roughness as a function of working pressure (a) 5 mTorr, (b) 7 mTorr, (c) 9 mTorr, (d) 11 mTorr, and (e) 13 mTorr.