



The inter-metallic oxide of ZnO/ITO/ZnO tri-layer films using a heat-induced diffusion mechanism

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ABSTRACT

This study presents a bias-crystallization mechanism (BCM) that is based on ZnO/In/ZnO tri-layer film and thermal annealing treatment on ZnO/ITO/ZnO tri-layer films. After biasing (40 V, 0.025 A), the resistivity of the ZnO/In/ZnO sample was reduced to $1.35 \times 10^{-2} \Omega \text{ cm}$. Bias-induced Joule heat and indium ion diffusion were critical factors with regard to decreasing resistivity. When substituted for the metal indium layer, the ZnO/ITO (13 nm)/ZnO thin film demonstrated comparatively better electrical properties and optical transmittance. During thermal annealing, the indium and tin atoms in the ITO structure diffused into the ZnO matrix and improved the conductivity of the tri-layer film. Inter-metallic oxide (IMO) was formed in the interface between the ZnO and the interlayer, and it dominated the crystallization characteristics as well as the optical and electrical properties of the tri-layer films.

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1. Introduction

Transparent conductive oxide (TCO) thin film has received extensive attention due to its potential applications in various optoelectronic devices [1–3]. Indium tin oxide (ITO) has been the usual candidate for TCO film due to its particular optical and electrical characteristics [4]. However, sources of ITO material will be rare and valuable in the future. Also, various ZnO-based thin films have been widely investigated because of their easy preparation and good optoelectronic properties [5,6]. To achieve higher conductivity and transmittance, many studies have combined a metal layer with semiconductor layers to form tri-layer TCO thin films, such as IZO/Al/GZO [7], AZO/Cu/AZO [8], ZnO/Ag/ZnO [9] and ZnO/Cu/ZnO [10] tri-layer films, among others. Notably, most studies have used metal as the interlayer to improve the electrical properties. However, a mirror metal interlayer results in a decrement in optical transmittance. Furthermore, the interface characteristics in tri-layer films system affect the optical and electrical properties of TCO films and related investigations have been few in number.

In this study, a ZnO/In/ZnO tri-layer film using an electrical current method was investigated to understand ion diffusion behavior over a short period of time. An ITO interlayer ($t = 3\text{--}17 \text{ nm}$) was

substituted for the indium layer, and the influences of various ITO thickness on the optoelectronics properties of ZnO/ITO/ZnO system were clarified. The electrical properties of ZnO/ITO/ZnO film were compared with Zn–In–O (IZO), Zn–In–Sn–O (ZITO) and ITO film in order to understand the contribution of ion diffusion mechanisms. The interface characteristics that influence the performance of TCO film are topics worthy of further research.

2. Experimental procedure

ZnO film of 100 nm and an indium (In) layer of 50 nm were sequentially deposited onto a glass substrate to form the ZnO/In/ZnO tri-layer film using RF sputtering. The calibration curve for the current values as a function of varied biasing voltage was measured to acquire a stable biasing condition (Fig. 1(a)). The current value reached a maximum value (0.025 A) at a bias voltage of 35 V and then decreased to 0.02 A when the bias voltage was increased to 43 V. This result was associated with the deterioration of indium crystallization caused by bias-induced Joule heat [11]. According to above results, a bias voltage of 40 V was used in the biasing treatment. Additionally, a determination of the relation of biasing duration and bias-induced temperature was also carried out, as shown in Fig. 1(b). The temperature of the sample surface was raised to 140 °C, and then saturated within 13 min with increasing the bias duration. Accordingly, a DC power supply (KIKUSUI, PAK60-6A) provided an electrical current onto the

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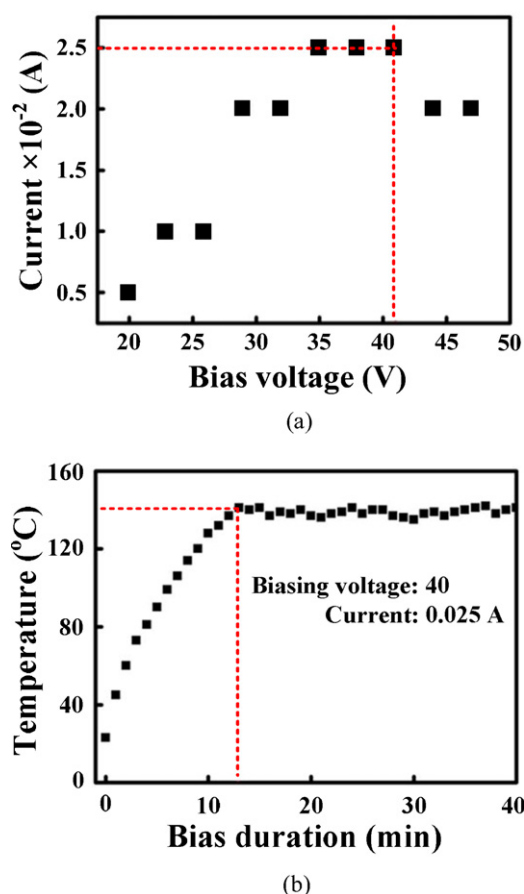


Fig. 1. (a) The calibration curve for the current value as a function of varied biasing voltages and (b) the relation of biasing duration and induced temperature biased at 40 V.

middle indium layer, and the ZnO/In/ZnO tri-layer film was biased at 40 V for 40 min in order to investigate the interface diffusion behavior. Considering the transmittance of tri-layer films, the ITO film substituted for the indium layer to form the ZnO/ITO/ZnO tri-layer film. The thickness of the ITO layer varied from 0 to 17 nm. The resulting ZnO/ITO/ZnO films were subsequently annealed at 500 °C for 1 h in a vacuum.

The interface mechanisms were examined using high-resolution thermal field emission transmission electron microscopy (FE-TEM, JEM-2100F) and energy-dispersive X-ray (EDX). The diffusion behavior at the interface zones was examined using electron spectroscopy for the purpose of chemical analysis (ESCA, PHI 5000 VersaProbe). The crystalline characteristics were examined using multipurpose X-ray thin film diffraction (XRD, Rigaku). The electrical properties and optical transmittance of all samples were measured using a Hall measurement system and UV–visible spectroscopy.

3. Results and discussion

3.1. Bias-induced interface mechanism

Fig. 1(a) shows the bright field image of the ZnO/In(50 nm)/ZnO tri-layer film. Both the ZnO films had a pillar-like crystalline structure and grew with a (002) orientation. From select area electron diffraction (SAED) patterns (Fig. 2(b)–(d)), the structures of the upper ZnO layer (point a) and bottom ZnO layer (point b) were attributed to ZnO wurtzite crystal, and the crystalline lattice was not twisted. These results indicated that the bias-induced thermal

Table 1

The resistivity of ZnO/In/ZnO samples with different treatment environments.

Sample	As-deposited	DC biasing 40 V, 0.025 A	Annealing 600 °C
Treatment time (min)	0	40	40
Resistivity ($10^{-2} \Omega \text{ cm}$)	–	1.35	1.99

diffusion behavior did not affect the crystal lattice. The diffusion distance of the indium ions was not clear.

Fig. 3 reveals the depth profiles of the element distribution for ZnO/In/ZnO tri-layer films intended to lead to an understanding of the diffusion behavior in the interface between the ZnO and indium layers. For an un-biased sample (Fig. 3(a)), zinc and oxygen ions were detected in the initial stage (film surface). With increasing detection time, the intensity of the zinc ions gradually decreased, and the indium gradually increased, which indicated that X-ray had gotten into the interface between the ZnO and indium layers. After biasing at 40 V for 40 min (Fig. 3(b)), some indium ions were detected on the film surface, and meanwhile, the zinc content gradually decreased. After that, the indium content increased, which indicated that the indium interlayer was detected. At that point, the indium content began to decrease, and the zinc content increased (the bottom ZnO layer was detected). The diffusion path of the indium ions was about 50 nm, and these indium ions might integrated into the ZnO to form a Zn–In–O (IZO) structure in the interface.

Electrical measurements were conducted to determine the resistivity of ZnO/In/ZnO tri-layer films under different treatment conditions (Table 1). The resistivity of the as-deposited ZnO/In/ZnO sample could not be acquired due to the fact that its resistivity value exceeded the limit of detection. After biasing for 40 min (40 V, 0.025 A), the resistivity of the ZnO/In/ZnO tri-layer film was $1.35 \times 10^{-2} \Omega \text{ cm}$. In fact, the indium ions were diffused into the ZnO, resulting in the formation of an IZO structure in the interface between the ZnO and indium layers that affected the electrical properties of the ZnO/In/ZnO tri-layer film. Additionally, as compared to a traditional annealed treatment (annealing at 600 °C for 40 min), the biasing sample demonstrated lower resistivity and significantly less energy consumption [11]. In brief, the improvement in ZnO/In/ZnO resistivity was attributed to the formation of inter-metallic oxide (IMO) in the interface. Inserting the metal layer into ZnO films was shown to definitely reduce the resistivity of thin film (Table 2) [9,12,13]. However, a high temperature treatment is necessary to acquire lower film resistivity that wastes energy and is unsuitable for application in flexible substrates. The consumed energy of the biasing treatment greatly reduced, and better film conductivity achieved in a short time [11]. Additionally, the biasing induced thermal energy (140 °C) was low enough that it could be considered as a candidate for applications involving low temperature manufacturing processes.

Based on above results, a schematic illustration of the interface characteristics of the ZnO/In/ZnO tri-layer film with a bias-crystallization mechanism (BCM) is shown in Fig. 4. It can be seen

Table 2

Comparison of the resistivity for different multi-layer structures.

Multi-layer structures	Resistivity ($\Omega \text{ cm}$)	Post-treatment	Reference
ZnO (200 nm)	3.5×10^3	–	[9]
ZnO (200 nm)/Al (20 nm)/ZnO (200 nm)	2.9×10^{-3}	300 °C	[12]
ZnO (35 nm)/Ag (12 nm)/ZnO (35 nm)	9.4×10^{-3}	–	[13]
ZnO (100 nm)/In (50 nm)/ZnO (100 nm)	1.4×10^{-2}	DC biasing 40 V to 0.025 A	Present study

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