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Enhanced electrical and optical properties based on stress reduced graded structure of Al-doped ZnO thin films

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ABSTRACT

Graded structures of aluminum-doped zinc oxide (AZO) multilayered thin film were prepared on quartz glass substrate by sol-gel process, and then sequentially annealed by raped thermal annealing(RTA) and UV laser annealing technologies for transparent conducting oxide (TCO) applications. Different Al mol% (0, 0.17, 0.33,0.5, 0.66, 0.83, 1) doped ZnO graded structures of multilayer thin films were prepared to optimize the lattice parameter to reduce stress, and then the annealing processes were sequentially performed. Introducing graded multilayered thin films, reduced the stress between the layers. The AZO graded structures of multilayer thin films were annealed by RTA followed by a 350 nm nanosecond pulsed UV laser annealing method. The graded structures of multilayered AZO thin films were investigated and analyzed by X-ray diffraction (XRD), field emission scanning electron microscope (FE-SEM), four-point probe, and UV–vis spectrophotometer, respectively. These results show that multilayered graded thin films were well grown with decreased stress, and well crystallized along the c-axis. The optical transmittance of the films is around 94.8% at 400–800 nm wavelength, and the energy band-gap is around 3.27 eV, respectively. The sheet resistance value of 13.2 kΩ/sq shows a 30% improvement.

1. Introduction

Transparent conducting oxide (TCO) has attracted worldwide notice for its high versatility. In particular, organic light-emitting devices (OLEDs) [1–3], optical sensors [4] and thin films solar cells [5] are typical TCO applications. Until the last few years, these applications were future technologies, but due to the rapid evolution of technology, TCO is nowadays widely used in the world. Up to this time, Indium Tin Oxide (ITO) has been used for most of TCO applications, but ITO has many disadvantages. Indium is a toxic metal that threatens the environment, and its resource is limited, because it is a rare earth material. For these TCO applications, aluminum-doped zinc oxide (AZO) can be an alternative to ITO. AZO has many advantages, such as improved electrical and optical properties, high temperature stability, non-toxicity, and low-cost fabrication. However, when functional materials are exposed to electronic devices applications, small weak points of materials can be huge draw backs for device reliability. For instance, thin films were usually used for active channel layer of TFT device. Therefore on-resistance variation may come from the defects of materials traps for free carrier and arise threshold voltage drift, on-resistance variation and imperfect channel saturation which make device uncontrollable and easily breakdown [6]. Therefore, the removal of defects should be carefully considered. In general, ZnO or AZO thin films have many crystal defects that arise during the thin film growing process. These defects in the AZO thin films can affect the electrical and optical properties of AZO thin films, because the defects develop as a result of the co-relation between the stress and strain, which inevitably formed from the lattice mismatch and thermal expansion with substrate. Therefore, reducing the stress is very important to enhance the functionality of thin films or decrease the strains, which result in making a more coherent wave with high transmission.

Many thin film deposition methods have been employed to fabricate the AZO thin films, such as pulsed laser deposition [7–9], magnetron sputtering [10–12], and molecular beam epitaxy [13], which are classified as physical deposition. Also, as chemical vapor deposition methods, atomic layer deposition [14–16], sol-gel spin coating [17], and electrostatic spray-assisted vapor deposition [18–20] have been employed. In this paper, a sol-gel spin coating deposition method which has advantages of large area deposition, high homogeneity of the precursor and enables control of stoichiometry [21], was attempted to deposit graded AZO multilayer thin film structure on glass substrates with RTA and laser annealing process. In this paper, we investigate the effects of graded structure for AZO thin films for TCO applications to reduce the strains coming from the lattice mismatch. By preparing the

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graded AZO multilayer thin film, lattice parameters were slightly modified in the multilayer structure to reduce the stress as much as possible. As a result, the co-relation behavior between the reduced stress and electrical properties is discussed.

2. Experimental details

Graded AZO multilayer thin film structures were prepared by a sol–gel process on glass substrates (Corning EAGLE, JMC GLASS). Zinc acetate dihydrate ($Zn(CH_3COO)_2{\cdot}2H_2O$) and aluminum chloride hexahydrate ($AlCl_3{\cdot}6H_2O$) were used as starting materials. 2-Methoxyethanol and monoethanolamine (MEA) were used as solvent and stabilizer, respectively. Different doping concentrations of Al were employed in the multilayer structure to modify the lattice parameters to reduce the strain. The molar ratio of MEA to zinc acetate dihydrate was maintained at 1.0, and the concentration of zinc acetate dihydrate was 0.7 mol/l. Appropriate amounts of 0, 0.17, 0.33, 0.5, 0.66, 0.83 and 1 mol% of Al were added to ZnO as dopants. The resultant solution was stirred at 60 °C for 2 h. The solution was dropped onto glass substrates by the spin coating method. After deposition by sol–gel spin coating, the films were dried in air at 300 °C for 10 min on a hotplate, in order to remove the solvents and organic compounds by evaporation. First, different Al mol% doped ZnO thin films were deposited on the glass substrates to determine which composition of Al was well matched on the glass substrates. Then, the first thin film layer on the glass substrates was fixed and different mol% Al doped ZnO thin films were prepared on the optimized AZO layer. After the deposition, Rapid thermal annealing process and UV laser annealing process [22,23] were sequentially carried out to crystalize the graded AZO multilayer thin film structure. After the thin film preparation process, different types of characterization processes were carried out to compare the electrical and optical properties. The crystal structures of graded AZO thin film multilayer structure were investigated by X-ray diffraction (XRD). The surface microstructure was examined by field emission scanning electron microscope (FE-SEM). The electrical resistance was measured by four-point probe method. Optical transmittance measurements were carried out by UV–vis spectrophotometry.

3. Results and discussion

Fig. 1(b) explains the geometrical properties of different Al mol% doped AZO thin film layers to prepare the graded AZO thin film multilayer structure. The figure shows that 6 layers of AZO thin film were deposited on the glass substrates by employing the spin coating process. The thickness of each AZO layer was around 50 nm, and therefore 300 nm thickness of AZO thin film [24,25] was prepared to know which composition could be well matched on the glass substrates. Fig. 1(a) shows the measured sheet resistance of different Al mol% doped ZnO thin films on the glass substrates. As shown in the Fig. 1(a), 0.5 mol% of Al doped ZnO thin film showed the lowest sheet resistance of 19.3 kΩ/sq. This means that as thin film form, 0.5 mol% Al doped ZnO thin films have lowest surface defects [26,27]. The free charge carrier can be trapped in defect and it changes electric potential at particular region which can act like potential wall. So the lowest surface defects means enhancement of conductivity and if the minimum free charge carrier trap exists in the thin film, carrier can move easily without disturbance [27].

After fixing the first layer of AZO thin films as 0.5 mol% of Al doped ZnO, the middle layers in multilayer thin film were then changed as shown in Fig. 2(b). The middle layer of thin films was changed from 0.33 mol% of Al doped ZnO thin films to 0.66 mol% of Al doped ZnO thin films to avoid the misfit stress as much as possible. After the graded AZO multilayer thin film deposition, RTA and UV laser annealing processing were sequentially conducted [22,23]. After that, the sheet resistance was measured. The measured sheet resistance was shown in the Fig. 2(a). After the optical annealing process, 1 layer of 0.66 mol% of Al doped ZnO thin film showed the lowest sheet resistance value of 13.2 kΩ/sq. The major purpose of RTA and laser annealing is providing enough optical energy in a short time to remove internal defect in thin film by minimize substrate heating. If substrate heated, thermal expansion occurs and arises inevitable lattice mismatch at substrate-thin film interface. As a result, low sheet resistance which measured after annealing processes, shows enhancement of conductance [23,28,29].

After changing the Al mol% in the middle layer composition in the multilayer structure, then the 3rd middle layer composition was then fixed as 0.66 mol% of Al doped ZnO thin film. Then, the number of 0.66 mol% Al layers was slightly increased, as shown in Fig. 3(b). In

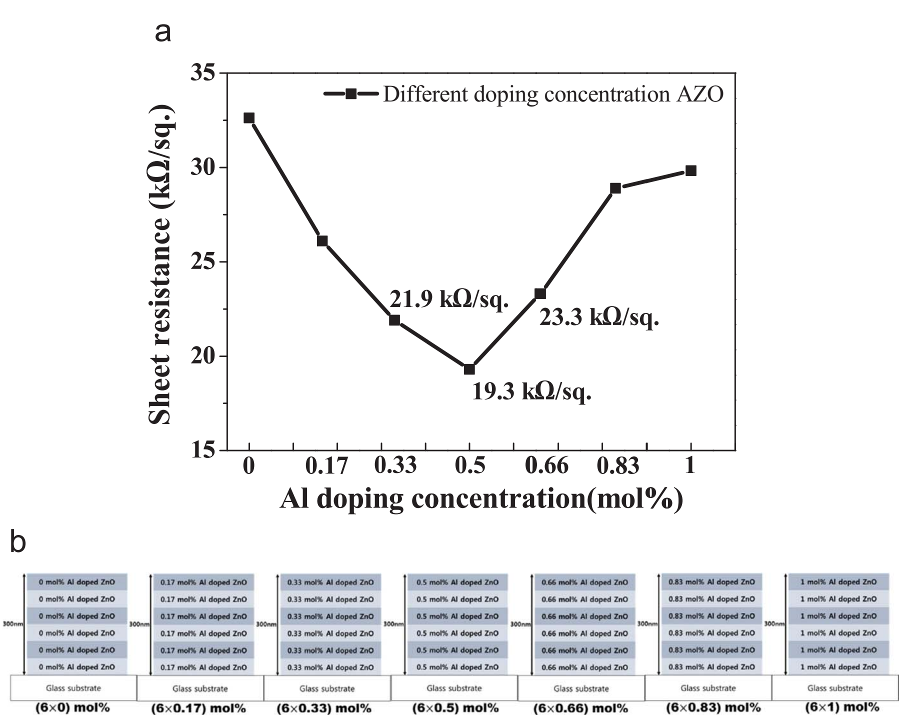

Fig. 1. (a). The sheet resistance for 0, 0.17, 0.33, 0.6. 0.66, 0.83, and 1 mol% of Al doped ZnO thin films. (b). Geometrical properties of different mol% of Al doped ZnO thin film structure.