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Roll-to-toll Growth of Ga-doped ZnO Transparent Conducting Films by Using Plasma-assisted Molecular Beam Deposition

Tsutomu Muranaka[1], Yuhei Tsuchiya[1], Taiga Aoki[1], Norio Onojima[1], Takashi Matsumoto[1], Satoshi Hiraki[2], Hiroshi Kono[3], Kazuhiro Kijima[3] and Shigeru Hagihara[3]

[1] Univ. of Yamanashi
4-3-11, Takeda, Kofu 400-8511, Japan, Phone: +81-55-220-8476, E-mail: tmuranaka@yamanashi.ac.jp
[2] Nakaya Corporation
1641-8, Tsuijiarai, Showa-cho, Nakakoma-gun, Yamanashi 409-3853, Japan, Phone: +81-55-275-5533
[3]Yamanashi Industrial Technology Center
2094 Otsumachi, Kofu 400-0055, Japan, Phone: +81-55-243-6111

Abstract

Novel growth method of Ga-doped ZnO (GZO) transparent conducting films (TCFs) was demonstrated by using plasma-assisted "roll-to-toll" molecular beam epitaxy system. The GZO TCFs on PEN and PET sheets also showed excellent transparency higher than 85 %. The resistivities of GZO TCFs as low as 10^{-4} Ωcm order have been successfully achieved on roll plastic sheets after optimization of growth conditions.

1. Introduction

Transparent conducting films (TCFs) are key components of optoelectronic devices such as solar cells, image sensors, light-emitting diodes, and flat panel displays. Indium tin oxide (ITO) is the most commonly used material for TCFs. Ga-doped ZnO (GZO) is a promising alternative to ITO because of its low resistivity, high transparency, nontoxicity, and resource abundance. The deposition technique widely used in industry is sputtering. During sputtering, high-energy particles may damage the TCF itself and devices such as solar cells underlying the TCF. Various techniques, such as sol–gel, chemical splay, metal organic chemical vapor deposition (MOCVD), and mist CVD, have been developed for the damage-free deposition of ZnO TCFs. However, these methods require relatively high growth temperatures or high-temperature post-annealing process. In this paper, we first report on novel roll-to-roll (RTR) growth technique of GZO TCFs by using a low damage growth technique at temperatures close to room temperature [1-3], and resistivities as low as 10^{-4} Ωcm order have been successfully achieved on roll plastic sheets.

2. Experimental

All the growth procedures of the GZO films were carried out in our plasma-assisted RTR molecular beam epitaxy (MBE) system shown in **Fig. 1**. The low temperature growth, below 60ºC, on roll PEN and PET sheets were performed with no heating process. Width and thickness of these plastic sheets were 250 mm and 100 μm, respectively. The rolled sheet was correctly wound up from a bottom roller to a top roller after growth. Rolling speed during growth was changed from 4 to 18 mm/min. Zinc (Zn) vapor was supplied on sheet by heating of metallic Zn from 350ºC to 395ºC, and oxygen was excited in a microwave cavity and only neutral atomic radicals were supplied onto sheets. Therefore, grown film is free from ion bombardment. Gallium (Ga) vapor was also supplied by heating of metallic Ga at 850ºC. GZO films were characterized by Hall measurements, X-ray diffraction (XRD), optical transmittance.

3. Results and Discussion

Contour plots of thickness of the GZO films prepared with different zinc source temperatures, T_{Zn}, are shown in **Fig. 2**. Thickness of the GZO films versus process time is also shown in **Fig. 3**. Large decrease of film thickness was observed at high temperatures. However, this decrease was improved by optimization of growth conditions. **Figure 4** shows XRD spectra of 2θ/θ scan of the (002) plane reflection from the GZO films grown on PEN, PET sheets and glass substrates as reference. Both samples show a (002) single peak, which indicates that the films are highly c-axis orientated polycrystalline films with wurtzite structure. Optical transmittance spectra of the GZO films grown on PEN, PET, and glass substrates as reference are shown in **Fig. 5**. The GZO TCFs on PEN and PET sheets also showed visible transparencies as good as that of the GZO films on glass substrate, and the average transmittance in the visible region was higher than 85 %. Relationship between resistivities of the GZO films and film thickness under various growth conditions is shown in **Fig. 6**. In this study, various growth parameters were tried for optimization of growth conditions. The resistivities of GZO TCFs as low as 10^{-4} Ωcm order have been successfully achieved on roll plastic sheets after optimization of growth conditions.

4. Conclusions

Novel growth method of GZO TCFs was demonstrated by using plasma-assisted RTR MBE system. The GZO TCFs on PEN and PET sheets also showed excellent transparency higher than 85 %. The resistivities of GZO TCFs as low as 10^{-4} Ωcm order have been successfully achieved on roll plastic sheets after optimization of growth conditions.

References

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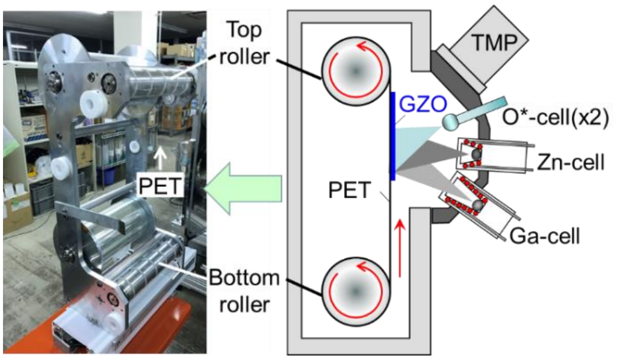

Fig. 1 Photographic and schematic views of the roll-to-roll (RTR) growth system for the Ga-doped ZnO (GZO) films in this study.

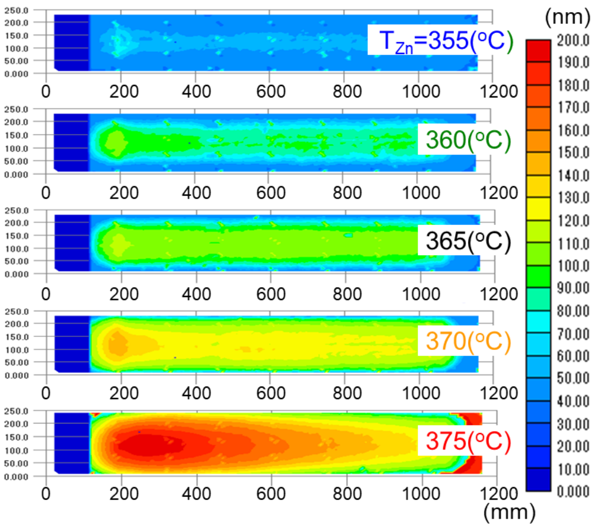

Fig. 2 Contour plots of thickness of the GZO films prepared with different zinc source temperatures, T_{Zn}.

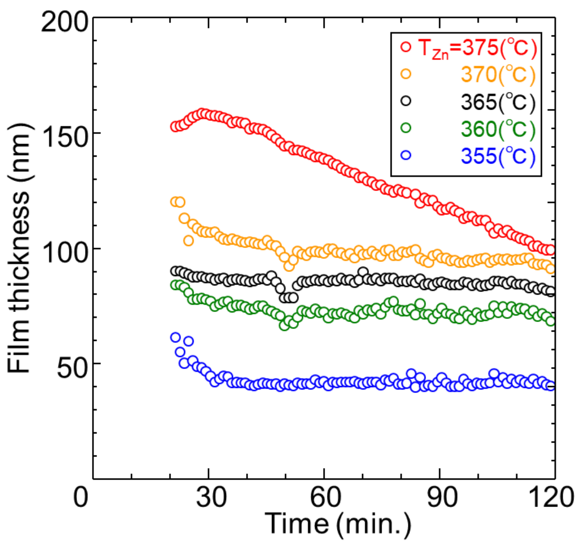

Fig. 3 Thickness of the GZO films versus RTR process time.

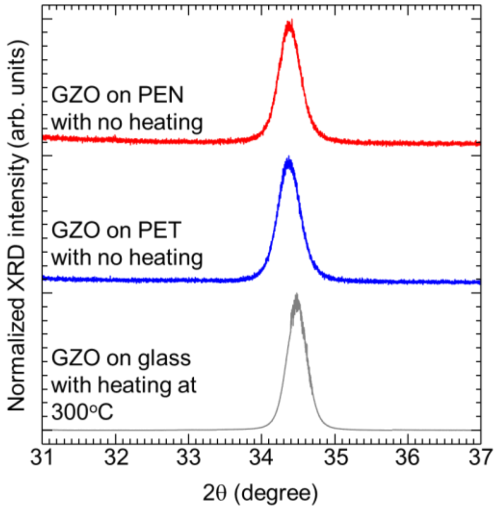

Fig. 4 XRD spectra of (002) reflection from the GZO films grown on PEN, PET, and glass substrates as reference.

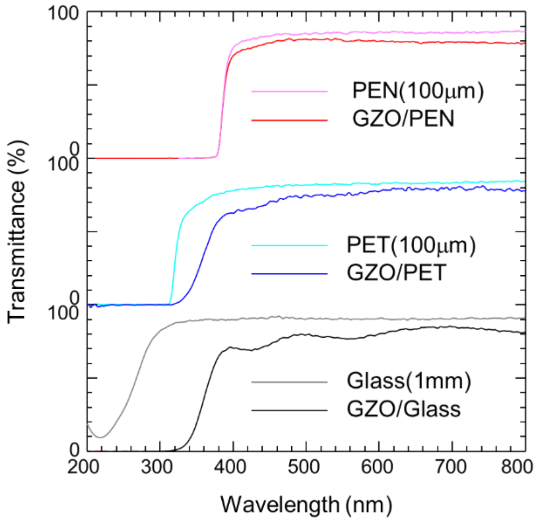

Fig. 5 Optical transmittance spectra of the GZO films grown on PEN, PET, and glass substrates as reference.

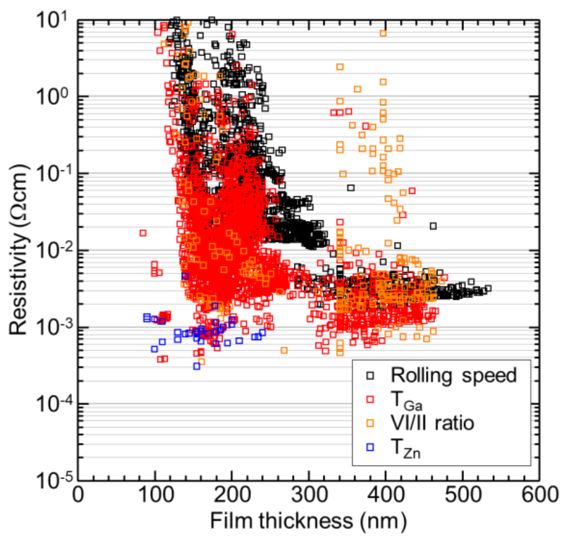

Fig. 6 Relationship between resistivities of the GZO films and film thickness under various growth conditions.