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High-performance ZnO:Ga/Ag/ZnO:Ga multilayered transparent electrodes targeting large-scale perovskite solar cells

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ABSTRACT

We investigated the thermal stability of multilayered transparent electrodes composed of ZnO:Ga (GZO)/Ag/GZO layers for the application of large-scale perovskite solar cell (PSC) modules, which require a post-annealing temperature of 450 °C. The sheet resistance and transmittance of the GZO/Ag/GZO (50 nm/12 nm/50 nm) electrodes were encouragingly improved by an annealing test with a temperature variation from 25° to 450°C. The optimum value of the sheet resistance was 4.48 Ω/square at an annealing temperature of 450 °C. By a simulation of PSC module performance, we confirmed that PSC modules with our GZO/Ag/GZO multilayered transparent electrodes (4–5 Ω/square) suffer less performance degradation compared to those with conventional FTO transparent electrodes (8–15 Ω/square).

1. Introduction

The performance of perovskite solar cells (PSCs) has dramatically improved over half a decade, and a record efficiency of 22.7% for a cell with an active area of less than 0.1 cm^2 has been achieved [1–4]. However, the upscaling of high-efficiency PSCs in a series interconnection is essential for viability in the commercial market. To achieve high-efficiency PSC modules, design parameters such as the active-layer width and dead region width must be optimized, while the sheet resistance of the contact layers should be minimized [5–7].

So far, transparent conducting oxide (TCO) thin films on glass substrates have received a considerable amount of attention as a contact layer in planar-type PSC configurations [8]. In conventional thin-film solar cells, the TCO should possess a high transmittance in the visible wavelength region and a low sheet resistance. However, for a PSC configuration, the thermal stability is another crucial factor because a high-temperature annealing process is required for the dense TiO_2 hole blocking layer deposited onto the TCO layer during fabrication of the PSC [9–11]. Various TCO candidates such as indium tin oxide (ITO), fluorine-doped tin oxide (FTO, SnO_2:F), and aluminum-doped zinc oxide (AZO, ZnO:Al) have been investigated. Among them, FTO thin films have been widely used because of the high thermal stability of FTO; unlike FTO, the sheet resistances of ITO and AZO significantly increase with an annealing temperature over 300 °C. However, the sheet resistance of conventional FTO (8–15 Ω/square) is still high for PSC module applications; thus, other candidates are required.

To date, ITO/Ag/ITO multilayered electrodes have been widely employed in thin-film solar cell structures owing to the low-temperature process, flexibility, low sheet resistance, and easy control of the optical properties [12,13]. However, the PSC and its application are limited by the low thermal resistance of the ITO/Ag/ITO structure. The oxidation of ITO and Ag diffusion result in optical and electrical degradation at annealing temperatures above 300 °C [14–16]. In addition, ITO is well-known to suffer from a material cost issue due to the use of indium.

In contrast, the optical and electrical properties of AZO/Ag/AZO multilayered electrodes are maintained up to an annealing temperature of 400 °C [17]. Further, other research groups have improved the performance of AZO/Ag/AZO electrodes with an annealing temperature of 500 °C [18]. This implies that ZnO-based oxide/metal/oxide (OMO) multilayered electrodes have a better thermal stability than ITO-based OMO electrodes, and they could be a good candidate for the bottom contact layer in high-efficiency PSCs.

It was reported that ZnO:Ga (GZO) thin films have an enhanced chemical and thermal stability compared with AZO thin films [19–21]. Therefore, in this study, we investigated GZO/Ag/GZO multilayered transparent electrodes that have a very low sheet resistance (< 5 Ω/square) and high transmittance (> 85%) in the visible wavelength range for PSC module applications. Moreover, we explored the thermal

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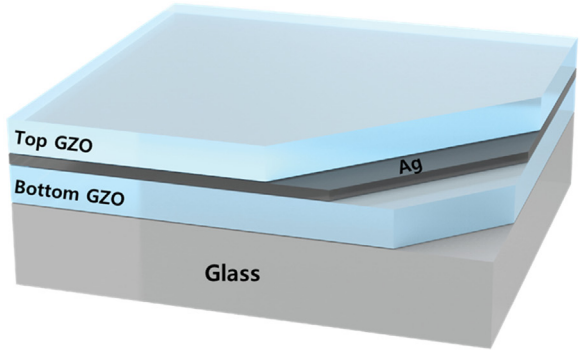
stability of GZO/Ag/GZO transparent electrodes considering the annealing temperature and time used in the fabrication of high-efficiency PSCs. In addition, we calculated the changes in the efficiency of PSC modules with respect to the sheet resistance of the TCO by an analytical simulation method. The employment of GZO/Ag/GZO transparent electrodes reduced the performance degradation in a PSC module compared with conventional FTO transparent electrodes, as the active-area width increased. Therefore, we expect that our GZO/Ag/GZO electrode is a promising candidate for large-scale PSC modules.

2. Experiments

GZO/Ag/GZO transparent electrodes were deposited onto glass substrates at room temperature by radio-frequency (RF) magnetron reactive sputtering with a Ag target (99.99% purity, diameter: 10.16 cm) and ZnO ceramic target (99.99% purity, diameter: 10.16 cm) with 5-wt% Ga_2O_3. In the deposition process, Ar gas was used as the sputtering gas at a flow rate of 150 sccm. The pressure within the chamber during the deposition process was 5 mTorr, and the RF plasma power was 300 W. A schematic and transmission electron microscopy (TEM) image of a GZO/Ag/GZO electrode are presented in Fig. 1. The multilayered structures consist of bottom GZO, Ag, and top GZO layers. To evaluate the thermal stability of GZO/Ag/GZO transparent electrodes, we used a thermal furnace system. The GZO/Ag/GZO electrodes were annealed in an air atmosphere, and the annealing temperatures and times were varied.

For characterization of the films, we measured their sheet resistances using a four-point probe and their film thickness using a profiler (Alpha-Step IQ, KLA-Tencor). The transmittance was measured by ultraviolet–visible-light (UV–Vis) spectroscopy (LAMBDA 750, PerkinElmer). The figure of merit (FOM) for a GZO/Ag/GZO electrode was defined by FOM $= \frac{188.5}{R_{sheet}(\frac{1}{\sqrt{T}}-1)}$ [22], where R_{sheet} and T denote the sheet resistance and transmittance at a wavelength of 550 nm, respectively. The crystal structures of the films were analyzed using X-ray diffraction (XRD, D/MAX-2500, RIGAKU) in the θ–2θ mode. The depth profiles of the films were measured by X-ray photoelectron spectroscopy (XPS, MXP10, ThermoFisher Scientific.). Cross-sectional images of the GZO/Ag/GZO electrodes were measured by TEM (JEM-2100F HR, JEOL).



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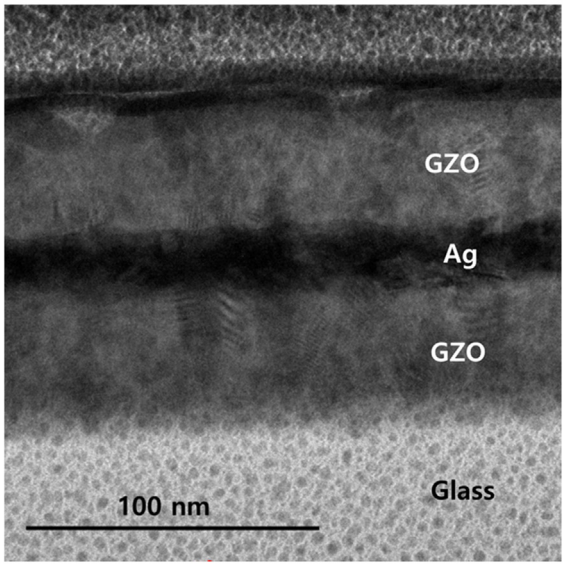


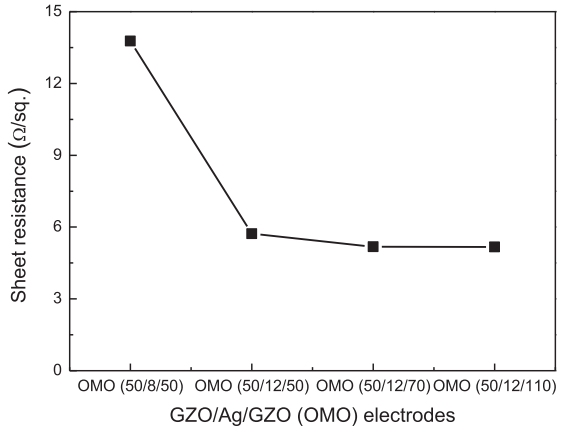
(b)

Fig. 1. (a) Schematic and (b) TEM image of a fabricated GZO/Ag/GZO (50 nm/12 nm/50 nm) transparent electrode.

3. Results and discussion

3.1. Performance optimization of GZO/Ag/GZO electrodes

Fig. 2 shows the electrical and optical performance of GZO/Ag/GZO electrodes deposited at room temperature with different layer thicknesses. In this study, the thicknesses of the Ag and top GZO layers were varied while the bottom GZO layer thickness was fixed at 50 nm. The



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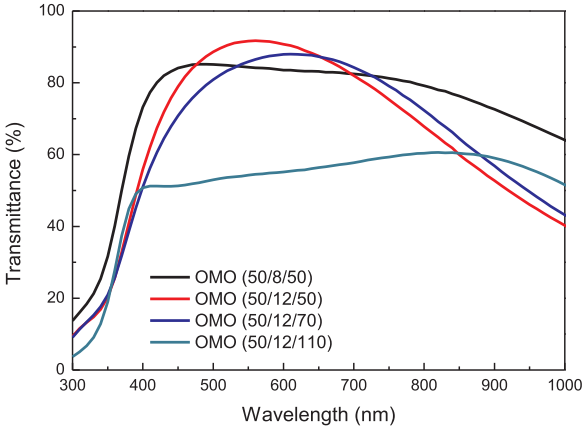

(b)

Fig. 2. (a) Sheet resistances and (b) transmittances of GZO/Ag/GZO (oxide/metal/oxide, OMO) transparent electrodes with various layer thicknesses; "OMO" (e.g., 50/12/110) denotes the layer thicknesses of the top GZO, Ag, and bottom GZO layers (50, 12, and 110 nm, respectively).