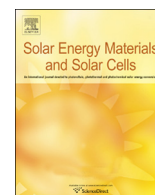




Contents lists available at ScienceDirect

## Solar Energy Materials and Solar Cells

journal homepage: [www.elsevier.com/locate/solmat](http://www.elsevier.com/locate/solmat)

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

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## ARTICLE INFO

## Keywords:

Transparent electrodes  
Perovskite solar cells  
Sheet resistance  
Transmittance

## 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<sup>2</sup> 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<sub>2</sub> 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<sub>2</sub>: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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Received 8 May 2018; Received in revised form 9 July 2018; Accepted 10 July 2018

Available online 20 July 2018

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