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Vapor textured aluminum-doped zinc oxide on cellophane paper for flexible thin film solar cells

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ABSTRACT

Paper-based flexible thin film solar cells are promising power sources for the emerging portable and wearable electronics, for which high conversion efficiency and flexibility are demanded. Herein, flexible thin film silicon solar cells using textured aluminum-doped zinc oxide (AZO) for light scattering, and ultrathin silver electrodes combined with cellophane paper substrates, were demonstrated. A universal route to form textured AZO thin film by chemical etching with hydrochloric acid vapor was proposed. The AZO layers textured in this way exhibited similar light scattering properties as compared with state-of-the-art AZO thin films. Applying textured AZO to solar cells on cellophane paper substrates resulted in current density and conversion efficiency increase from 8.7 to 9.5 mA/cm^2 and from 4.1% to 5.7%, respectively. Thin film solar cells on cellophane exhibited excellent mechanical flexibility, which maintained 90% of their initial efficiency after bending at a radius of 1 mm for 50 cycles.

1. Introduction

The next-generation of low-cost flexible, portable and wearable electronic systems require adapted flexible power sources. Flexible thin film solar cells could serve for this task and development of such devices with appropriate performance is of considerable interest. Among flexible substrates, paper is compelling owing to its low-cost, lightweight, earth-abundant, environmental friendliness, bendable and foldable properties [1–4]. Additionally, paper is thermally stable up to 300 °C which is of advantage for layer and device preparation processes at such elevated temperatures. Therefore, paper-based solar cells are considered promising choice for power sources in flexible electronic systems.

Recently, there was an increasing interest in the development of paper-based thin film solar cells [5–14]. The best reported conversion efficiencies of thin film silicon, organic, and perovskite solar cells on paper were 6.7% [9], 5.94% [10], and 9.05% [11], respectively. Usually these efficiencies are lower than those on their rigid counterparts. One reason might be the coarse surface of paper substrates. It could be overcome by utilizing paper substrates with smoother surface. Nanopaper composed of nanoscale cellulose fibers is an example of a popular option [15–17]. In the present study we propose and apply cellophane as smooth and transparent paper substrate. Compared to nanopaper, though the fabrication processing of cellophane creates a bit of toxic waste, the energy consumption for fabrication of cellophane is much lower, which can potentially shorten the energy payback time [18]. Additionally, cellophane is a commercially available material in wrapping and biomedical applications. These advantages of cellophane make it an attractive substrate for flexible thin film solar cell. To our knowledge, no investigation on cellophane-based solar cells has been reported up to now.

Another reason for the lower conversion efficiencies of paper-based solar cells is seen in the inefficient “light management” structures typically applied so far. Note that with “light management” we summarize the combined effects of layers and structures in solar cells for light scattering, trapping and in-coupling, all of which is mandatory for achieving high conversion efficiency especially in thin film solar cells. By now, the reported light management structures in paper-based solar cells were based on substrate, including nanoimprinting paper surface [9,12] and developing paper with strong light scattering ability [19]. These methods are complex and introduce additional procedures. The existing rigid substrates light management approaches, e.g. textured front electrodes and plasmonic back reflectors, have not been successfully applied to paper-based solar cells. This is due to the

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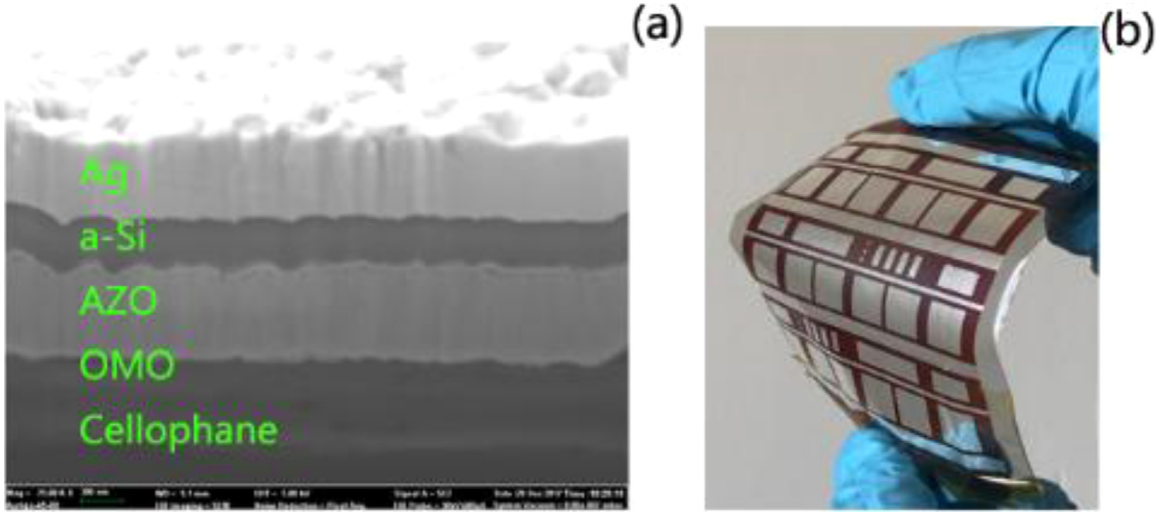

Fig. 1. (a) Cross sectional FIB-SEM image and (b) picture of a-Si:H solar cells on cellophane substrate.

incompatibility of existing approaches with the paper substrates, mainly arising from temperature and solvent restriction of paper. Therefore, developing a substantially simple and paper-compatible method for light management is of great importance.

In the present work, amorphous silicon (a-Si:H) thin film solar cells were developed and investigated on flexible cellophane paper substrates. A cross-sectional FIB-SEM image of solar cell is shown in Fig. 1a. ZnO/ultrathin silver(Ag)/ZnO (referred to OMO–oxide/metal/oxide) tri-layers were used as flexible transparent electrodes [20–22]. Aluminum-doped zinc oxide (AZO) thin films were textured by a newly developed hydrogen chloride (HCl) vapor method, in order to introduce a rough surface as light management structure. The optical properties of such structured AZO layers on flexible cellophane substrates were investigated and compared to corresponding layers on rigid glass substrates. The performance of resulting solar cells was studied in the as-deposited state as well as after multiple bending cycles. The solar cell efficiency improved from 4.1% to 5.7% using the structured AZO on flexible cellophane and retained ~90% of initial efficiency after bending at curvature radius of 1 mm for 50 cycles.

2. Experimental section

2.1. Thin films and solar cells

Layers and devices were prepared on 25 µm cellophane™ foils (Mosodo Environmental Materials Company, P. R. China) as well as on rigid glass (Corning Eagle XG) substrates with size of 10 * 10 cm^2. For ZnO/Ag/ZnO tri-layer electrodes, ZnO and Ag thin films were prepared by magnetron sputtering with 110 W RF and 3 W DC energy sources, respectively. Both processes were performed at room temperature and pressure was kept constant at 0.2 Pa. The resulting ZnO (40 nm)/Ag (12 nm)/ZnO (40 nm) possessed sheet resistance and average transmittance in the range 400–800 nm of ~8 Ω/□ and ~80% (see the transmittance curve in Fig. S1), respectively.

AZO thin films to be used as light management layer were prepared by radio-frequency magnetron sputtering with a ZnO:Al_2O_3 (99:1 wt%) target at a pressure of 0.47 Pa, power of 100 W and a substrate temperature of ~135 °C [23]. Thickness of as-deposited AZO films was ~780 nm. The texturing of the AZO was achieved by exposing the film surface to the vapor of a 25% HCl solution for times of 1 min, 2 min, 3 min, and 4 min (Fig. S2). The textured AZO film was cleaned by ethanol solution to remove residual surface contaminants.

Amorphous silicon (a-Si:H) based solar cells were prepared in p-i-n deposition sequence by plasma-enhanced chemical vapor deposition in a multichamber deposition system. Substrate temperature was kept constant at around 140 °C. Additional details can be found in Ref. [24]. Evaporated silver pads were used as back contact, which defined the area of solar cell as 1 cm^2. Total thickness of silicon layers was around 390 nm. After deposition, solar cells were annealed at 120 °C for 2 h in air [25]. For a-Si:H solar cells on glass, glass/AZO/p-i-n-Si/Ag stack was prepared to test the light scattering ability of textured AZO. Solar cells with textured state-of-the-art AZO prepared at 300 °C were used as reference [26]. For a-Si:H solar cells on cellophane, a cellophane/OMO/AZO/p-i-n-Si/Ag stack was prepared, in which OMO and textured AZO layers served as flexible electrode and light management structure, respectively. The picture of the fabricated solar cells on cellophane is shown in Fig. 1b. During bending procedure, solar cells on cellophane were wined around a cylindrical stick. Thus the bending radius was defined by the cylinder radius. The bending cycle was conducted manually.

2.2. Characterization

Transmittance and reflectance of the materials were measured using a UV/VIS/NIR spectrophotometer (LAMBDA 950, PerkinElmer, USA). The transmittance haze of the films was defined as diffused transmittance divided by total transmittance. The surface morphology of the AZO films was measured by a scanning electron microscope [SEM (X-Max 51 XMX0023, Oxford Instruments, England)]. The sheet resistance of the AZO films was measured by an electrometer with four-point-probe. The current density–voltage (*J–V*) curves of the solar cells were measured using a class A sun simulator which provided AM 1.5G illumination with a power density of 100 mW/cm^2. External quantum efficiency (EQE) was determined by measuring the differential spectral response of the solar cells.

3. Results and discussion

3.1. Wet-chemical etching of AZO thin films by HCl vapor

Wet chemical etching of AZO thin films is a widely used approach to introduce a light scattering texture for application in solar cells. Technically this is easily done by immersing as-deposited AZO thin films in 0.5% HCl solution [27]. For cellophane substrate, which is composed of cellulose, this approach is not applicable as the cellophane will react with the watery solution leading to deformation of the cellophane surface. Thus, we developed a new etching procedure for AZO thin films on cellophane, where the surface of the AZO layer on the