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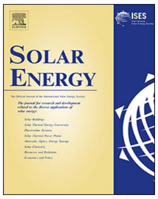

Flexible p-type PEDOT:PSS/a-Si:H hybrid thin film solar cells with boron-doped interlayer

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ABSTRACT

We reported highly flexible a-Si:H thin film solar cells with p-type poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) window. We firstly adopted the substrate-type cell structure on 75 μm-thick polyimide (PI) film unlike earlier studies in which the superstrate-type cell structures were utilized and the deformation of PEDOT:PSS was inevitably caused by subsequent deposition processes. We clearly demonstrated that the performance of the hybrid a-Si:H thin film solar cells with the substrate-type structure was superior to that of the superstrate-type cells. A highly boron-doped interlayer (IL) of 5 nm thickness was introduced at the hetero-interface between the p-type PEDOT:PSS and intrinsic a-Si:H to enhance built-in potential and form a homogeneous p/i-junction in the cell, which led to further improvement in the cell performance. The efficiencies of the cells with the PEDOT:PSS/IL window on glass and PI substrates were 7.40% and 6.52%, respectively, which were considerably higher than that of the cell with a conventional p-type microcrystalline (μc-) Si:H window. Also, the degradation of the cell with PEDOT:PSS window by bending was much smaller than the cell with p-type μc-Si:H window, particularly at bending radius < 10 mm. The present work demonstrates that PEDOT:PSS with a proper interfacial layer is a promising p-type window for substrate-type a-Si:H thin film solar cells and also for enhancing the flexibility of inorganic light absorbing materials-based solar cells on flexible film substrates.

1. Introduction

Flexible and light thin film solar cells fabricated on metal foils or plastic sheets have been extensively studied with the goal of achieving low cost and high throughput PV manufacturing for various applications, such as building- and vehicle-integrated PVs, and wearable electronic devices, etc. (Chae et al., 2014; Meillaud et al., 2015; Yoon et al., 2011). Among thin film solar cells, hydrogenated amorphous silicon (a-Si:H) thin film solar cells have numerous advantages in flexible applications due to their low processing temperature and mature process technologies, and also have intrinsic advantages, including naturally abundant materials, non-toxicity, low temperature coefficient, and chemical stability (Akhmad et al., 1997; Massiot et al., 2012). a-Si:H thin film solar cells also require much less material than crystalline silicon (c-Si) due to the high absorption coefficient of a-Si:H in the visible region (Shah et al., 2002). The a-Si:H thin film solar cells can be fabricated with two different structures, based on the deposition sequence of layers: the conventional superstrate-type, which is also called pin-configuration, on a transparent substrate; or an inverted structure on an opaque substrate, i.e., the substrate-type (nip-configuration). Regardless of their structure, the p-type layer of the a-Si:H thin film solar cells, which is used as a window, is very important for obtaining high performance because the lifetime of holes is much shorter than that of electrons (Ramanujam and Verma, 2012; Schade, 2010).

Substrate-type a-Si:H thin film solar cells with inorganic p-type windows have recently been attracting growing interest because they can be used for flexible applications on stainless steel (SS) and polyimide (PI) substrates, which have no or low transmittance of visible light compared to glass (Cho et al., 2011; Ostfeld et al., 2016; Rath et al., 2010; Söderström et al., 2008). This inverted structure can be a better choice for introducing various p-type materials than conventional superstrate-type cells, because the p-type window layer would not be exposed to the subsequent deposition process of the light-absorbing layer.

In this work, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) was adopted as a window layer of a substrate-type a-Si:H thin film solar cell because of its high transparency in the visible region, and reasonable stability in air. PEDOT:PSS has been often used as a hole transporting layer in organic solar cells and also a transparent

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conducting electrode because its conductivity could be enhanced by several techniques (Nagamatsu et al., 2014; Yeon et al., 2015; Yeon et al., 2017). The energy bandgap (E_g) of PEDOT:PSS is 1.5–1.6 eV (Nagamatsu et al., 2014; Williams et al., 2005), which is higher than that of p-type microcrystalline (μc-) Si:H film (E_g = 1.1–1.2 eV). The p-layer of larger energy bandgap is more advantageous because it must induce lower light-absorption loss.

Despite the notable advantages of PEDOT:PSS as a window layer, studies on substrate-type a-Si:H thin film solar cells hybridized with a p-type PEDOT:PSS window have not yet been carried out. Williams et al. (2005) had reported that PEDOT:PSS could act as an electrically ideal p-type window in superstrate-type a-Si:H thin film solar cells. However, the observed efficiency was as low as 2.1%. Jung et al. (2013) introduced a PEDOT:PSS window containing gold nanoparticles for superstrate-type a-Si:H thin film solar cells fabricated using a plasma-enhanced CVD (PECVD) system. The reported V_{OC} and efficiency of the cell on a glass substrate were 0.56 V and 5.49%, respectively (Du et al., 2006). The V_{OC} and efficiency were much lower than those of conventional a-Si:H thin film solar cells. These earlier studies taught us that the inferior performances might be due to degradation of the PEDOT:PSS films during the following plasma-assisted process, used for depositing the 450 nm-thick light absorbing layers (Du et al., 2006), and that the superstrate-type structure is not appropriate for p-type organic polymer window materials such as PEDOT:PSS, which can be degraded by subsequent fabrication processes.

In the present work, unlike the earlier studies reported on the superstrate-type cells on glasses, p-type PEDOT:PSS film was firstly adopted as the window layer of substrate-type a-Si:H thin film solar cells (called as the nip-configuration to describe the deposition sequence; n-, i-, and p-layer) on flexible PI with opaque back-reflector. With this configuration, the PEDOT:PSS can avoid any damage by subsequent thermal or plasma-assisted deposition processes.

In addition, the insertion of a highly boron (B)-doped Si intermediate layer (IL) at the interface between the p-type PEDOT:PSS and intrinsic (i-) a-Si:H layers is proposed to lower the barrier possibly caused by an imperfect and heterogeneous p/i-interface, thereby improving carrier collectivity.

In this work, the effects of an additive and H_2-plasma treatment on the adhesion of PEDOT:PSS to the a-Si:H film were also intensively investigated to improve the adhesion of PEDOT:PSS to the Si:H surface, because good adhesion at the organic/inorganic hybrid interface is critical to achieving a high performance device, especially on more flexible substrates.

2. Experimental details

Fig. 1 illustrates the structure of the substrate-type a-Si:H thin film solar cell fabricated in this work. PEDOT:PSS was used as a p-type window, and a very thin p-type Si:H IL was inserted at the PEDOT:PSS/i-Si:H interface to improve the p/i-junction properties for some samples.

The substrate-type a-Si:H thin film solar cells were fabricated in a single chamber PECVD system. The cells were deposited on Ga-doped ZnO (GZO)/Ag back reflector-coated PI and glass substrates with an rms roughness of approximately 30 nm at 200 °C. SiH_4 (100%), H_2 (100%), B_2H_6 (0.5%, diluted in H_2) and PH_3 (1.5%, diluted in H_2) were used as the source and doping gases. The thicknesses of the i-Si:H and n-Si:H were 300 and 30 nm, respectively.

The top electrode, 130 nm-thick GZO film, was deposited using an RF magnetron sputtering process. In order to minimize plasma-induced damage to the PEDOT:PSS during the GZO deposition, a two-step deposition was carried out at room-temperature. The 30 nm-thick GZO was deposited at a low RF-power of 100 W (power density of 1.23 W/cm^2), and 100 nm-thick GZO was deposited at 150 W. The total deposition time of GZO was only 2.8 min.

Then, an Al grid was formed on the GZO top electrode using the e-beam evaporation method at room temperature. The cell area was 20 mm^2. Because good adhesion between the PEDOT:PSS and inorganic Si:H surface is essential to fabricate the PEDOT:PSS/a-Si:H thin film solar cells, the effects of added Triton X-100 and H_2 treatment on the adhesion were investigated.

The PEDOT:PSS (Clevious PH1000, Heraeus) solution was mixed with Triton X-100 (Aldrich) and sonicated for 30 min, and filtered using a 5 μm pore size filter paper prior to spin-coating. The spin-coated PEDOT:PSS films were annealed at 120 °C. The adhesiveness was indirectly determined by measuring the wettability of the PEDOT:PSS solution on the a-Si:H film using a contact-angle analyzer (Phoenix 300, S.E.O). The thickness of the PEDOT:PSS film was measured using a profilometer (Alpha-step IQ, KLA-Tencor). To fabricate the solar cells, the PEDOT:PSS with added Triton X-100 was used as a window.

The optical transmittance of the PEDOT:PSS was evaluated using UV–Vis spectroscopy (LAMBDA 750, Perkin-Elmer) and compared to that of p-type Si:H. The influence of the subsequent plasma-assisted process on the PEDOT:PSS film was studied using X-ray photoelectron spectroscopy (XPS) (ESCALAB 200R, VG Scientific) with an Al Kα X-ray source and Fourier-transform infrared (FT-IR) spectroscopy (Nicolet iN10MX, Thermo Scientific™). The performance of the a-Si:H thin film solar cells was measured using a solar simulator with an AM 1.5G spectrum and 100 mW/cm^2 illumination, and the quantum efficiency (QE) curve was also obtained with a QE measurement system (IQE-200, Oriel).

To investigate the effect of bending on the performance of the flexible PEDOT:PSS/a-Si:H thin film solar cells on 75 μm-thick PI film, a home-built bending setup was utilized, and the results were compared to the behavior of a conventional a-Si:H thin film solar cell with a p-type μc-Si:H window on PI film.

3. Results and discussion

In a-Si:H thin film solar cells, the window layer absorbs a portion of the incoming light, resulting in a reduction in cell efficiency, and consequently light loss in the window layer must be minimized. To address this issue, the light absorption loss by p-type PEDOT:PSS was estimated in comparison to a p-type μc-Si:H film, which has been conventionally used as a window layer in a-Si:H thin film solar cells (Lee et al., 2015; Oh et al., 2014). The results are shown in Fig. 2. The light absorption loss by the PEDOT:PSS combined with highly boron (B)-doped Si:H IL was also estimated because the window structure would be also investigated to determine whether the insertion of the IL in the interface between the window layer and i-Si:H would have an effect on carrier collection.

In Fig. 2, the sum of specular transmittance and reflectance of PEDOT:PSS (90 nm), PEDOT:PSS (90 nm)/IL (5 nm), and p-type μc-Si:H (25 nm) films formed on very flat glass substrates are plotted to compare the absorption losses by the different window layers in the wavelength range of 300–700 nm. This range was selected because the bandgap energy of the i-Si:H layer is approximately 1.8 eV (690 nm) and light wavelengths longer than 690 nm cannot be utilized for photo-induced carrier generation. In Fig. 2, the calculated reflectance values with the thicknesses and refractive indices of PEDOT:PSS, IL, and p-type μc-Si:H were used.

The average sum values of transmittance and reflectance of the 90 nm-thick PEDOT:PSS and 25 nm-thick p-type μc-Si:H films were 89 and 71% in the wavelength range of 300–700 nm, respectively. The optical losses of the p-type μc-Si:H (29%) were approximately two times higher than that of the PEDOT:PSS film (11%). In contrast, the PEDOT:PSS/5 nm-thick IL film showed an average sum value of transmittance and reflectance of 86%, indicating the IL resulted in only a 3% reduction. This result suggests that using a PEDOT:PSS or PEDOT:PSS/IL window allows the i-a-Si:H layer to utilize much more light compared to the p-type μc-Si:H.

Prior to applying the PEDOT:PSS film as a window layer, it was