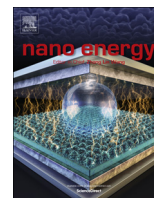




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High performance thin film solar cells on plastic substrates with nanostructure-enhanced flexibility

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ABSTRACT

Plastic substrates possess conspicuous advantages for flexible thin film solar cell applications due to their superior flexibility and light weight characteristics. However, there are several challenges of using plastic substrates for high performance thin film solar cells since they usually have low melting/softening temperature and high coefficient of thermal expansion (CTE). In this work, we demonstrated a low cost process to achieve regular nanocone arrays on polyimide (PI) substrates which have unique photon management property and excellent mechanical flexibility. To leverage these benefits of the nanocone substrates, flexible amorphous Si solar cells were fabricated on the structures. Intriguingly, it was discovered that properly designed nanocones can significantly improve solar cell device performance via light management. And the fabrication yield of properly designed nanocone solar cells is much higher than that of planar devices. In addition, the nanocone plastic solar cells possess much improved bendability and robustness verified by both experiment and mechanical modeling, showing unique stress release mechanism originated from three-dimensional nanostructure design. This property is of practical significance for flexible electronics not limited to solar cells.

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1. Introduction

Photovoltaic (PV) devices, which directly convert solar energy into electricity, have gained considerable interest worldwide as promising candidates for harvesting clean and renewable solar energy. Global cumulative photovoltaic capacity has grown exponentially for more than two decades, sufficient to supply 1 percent of global electricity demands by the end of 2014 [1]. Thin film solar cells are tremendously attractive for low cost applications, due to small material consumption and low temperature processes, as compared with the conventional crystalline Si-based PV devices. Particularly, light-weight and mechanically flexible thin film solar cells enable a wide range of potential applications from building-integrated PV generation to portable energy sources [2–4]. There are a number of choices of substrates for flexible thin film

solar cells, such as metallic foils, thin glasses and plastics [2,5–13]. With the superior flexibility and light weight characteristics, plastic substrates possess conspicuous advantages for wearable and aerospace/space applications [4,10,12,14]. However, there are several challenges of using plastic substrates for high performance thin film solar cells. For example, they usually have low melting/softening temperature and high coefficient of thermal expansion (CTE). Low melting/softening temperature limits the processing temperature for high efficiency solar cells based on materials including Si, CdTe, CIGS and perovskite thus leading to moderate energy conversion efficiency of the devices [15–20]. Meanwhile, higher CTE of the supporting plastic substrate than the atop photovoltaic active layers may incur stress and strain accumulation in the thin films thus leading to device failure or fast performance degradation. In this work, we demonstrate a low cost and scalable approach to achieve regular nanocone arrays on polyimide (PI) substrates, on which thin film hydrogenated amorphous silicon (*a*-Si:H) solar cells were directly fabricated with decent device performance. It is worth pointing out that among all plastic materials, PI has high thermal stability and excellent mechanical flexibility thus is promising as a substrate material for flexible

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electronic devices [9,15,17,21,22]. In our process, liquid PI solution was used to cast nanocone-structured films without relying on complicated and costly lithographic techniques. Several unique advantages of utilizing PI nanocones for thin film solar cells have been discovered in this work. Intriguingly, it was found that properly designed nanocones can significantly improve solar cell device performance. Particularly, device power conversion efficiency (PCE) was found almost doubled on the optimized nanocone structures, as compared with the device fabricated on a flat substrate prepared with the same solution process. Even compared with the devices fabricated on commercial flat PI substrates, the nanocone devices still outperform by 48%. Meanwhile, it was discovered that the fabrication yield of properly designed nanocone solar cells is much higher than the flat devices. The large improvement on device performance and fabrication yield can be rationalized by a combinational effect of superior light trapping capability and the thermal strain/stress release. Moreover, it was found that the nanocone plastic solar cells possess much improved flexibility and robustness as opposed to the flat devices. Experiment and mechanical modeling showed that the nanotexturized substrate helps to significantly reduce the stress and strain inside a solar cell device induced by mechanical bending. Note that this phenomenon has not been reported before and it is of practical importance for flexible electronics not limited to solar cells. Overall, this work demonstrates a viable and convenient route toward low cost fabrication of efficient and robust flexible thin film solar cells. Although *a*-Si:H is used as the model material in this work, the developed nanocone plastic substrates can also be used for other types of thin film PV devices, such as organic solar cells as well as emerging high performance perovskite solar cells.

2. Materials and methods

2.1. Materials

Aluminum foil (0.25 mm thick, 99.99% purity) was purchased from Alfa Aesar, liquid polyimide solution (PI-2525) was purchased from HD Microsystems, commercial flat polyimide film (Kapton[®] B, 25.4 μ m thick) was provided by DuPont. All other chemicals are products of Sigma-Aldrich.

2.2. Flexible nanocone PI substrates assembly

An electrochemically polished clean Al foil was mechanically imprinted using a silicon stamp with hexagonally ordered nanopillars with height of 200 nm and tunable pitch of 500 nm–2 μ m to produce a nanoindentation array on the surface of the Al foil. Thereafter, the i-cone array was fabricated by a multi-step anodization and wet etching process on the imprinted Al foil in an acidic solution with a proper direct-current (DC) voltage [23–25]. Afterwards, a 100 nm thick SiO₂ film was deposited on the as-obtained i-cone AAO template by PECVD at 150 °C as an anti-sticking layer between the i-cones AAO and PI for easy peeling off of PI film from the template subsequently. Thereafter, liquid PI solution (PI-2525) was spin coated onto the SiO₂-coated template at 50 rpm for 1.5 mins, followed by three baking steps (30 mins for each step) with temperatures of 100 °C, 150 °C and 170 °C on a hot plate to solidify the PI solution in air. After the sample cooled down to room temperature, the solidified PI film was carefully peeled off from the AAO template and then sandwiched in between silicon wafers and placed in an oven for higher temperature curing. The curing temperature gradually rose up to 200 °C with a ramping rate of 4 °C/min, then maintained at 200 °C for 30 mins, followed by increasing the temperature to 350 °C with a ramping rate of 2.5 °C/min. Then the PI film was curing at 350 °C for 4 hrs

before natural cooling. An argon flow was maintained at 400 sccm during the curing process in the oven. Finally, the fully cured PI substrates were immersed in buffered oxide etch (BOE) solution for 10 mins to remove potential SiO₂ residue on the surface.

2.3. Fabrication of *a*-Si:H solar cells

A Ag reflector layer was firstly deposited on a PI substrate by direct current (DC) magnetron sputtering at room temperature in argon plasma atmosphere. Then an AZO spacer layer was sputtered by radio frequency (RF) magnetron sputtering of a 2 wt% Al₂O₃ doped ZnO ceramic target (99.99% purity) under argon plasma at a base temperature of 150 °C. Subsequently, a stack of *n*-*i*-*p* *a*-Si:H layers were successively deposited in a multi-chamber PECVD system consisting of three PECVD chambers. All these chambers have identical capacitively coupled parallel-plate electrode configurations and serve for the deposition of intrinsic, *n* doped and *p* doped layer, respectively. The *a*-Si:H absorber layers were prepared from a mixture of SiH₄ and H₂ gases at an excitation frequency of 40 MHz. Doping was achieved by gas mixture of hydrogen-diluted phosphine (PH₃) for deposition of *n* layer and hydrogen-diluted diborane (B₂H₆) for *p* layer at an excitation frequency of 13.56 MHz. After device fabrication, an 80 nm thick ITO layer was deposited by RF sputtering as top electrode.

2.4. Device characterization

SEM images of the thin film *a*-Si:H PV devices were obtained by a JEOL JSM-6700F SEM working at 5 kV. Angular and wavelength dependent absorption spectra of all devices were performed with a home-built Ultra-violet/Visible measurement system. All the *J*-*V* curves of thin film *a*-Si:H solar cells were carried out using a solar simulator (Newport corporation, 91150V) under 1 sun illumination. The EQE measurements were characterized by Oriel QE-PV-SI (Newport Corporation).

3. Results and discussion

The fabrication procedure of thin film *a*-Si:H solar cells on nanocone PI substrates is schematically illustrated in Fig. 1a1–a4. Firstly, a highly ordered anodic aluminum oxide (AAO) template with an inverted nanocone (i-cone) array was fabricated (Fig. 1a1) via a multi-step anodization and wet etching process on the imprinted Al foil [23–25]. Note that the pitch and aspect ratio of the i-cones fabricated with this approach are largely tunable, as we have reported recently [24–30]. A 100 nm thick SiO₂ layer was then deposited on the as-obtained i-cone AAO template by plasma enhanced chemical vapor deposition (PECVD) at 150 °C as an anti-adhesion layer followed by spin coating of PI solution on the surface, as shown in Fig. 1a2. Then the PI solution coated AAO template was partially cured on a hot plate to solidify the PI solution in air with gradual increased curing temperature up to 170 °C. Thereafter, the solidified nanocone PI film (Fig. 1a3) was carefully peeled off from the i-cone AAO template. Afterward, the nanocone PI film (~100 μ m thick) was further cured in an oven up to 350 °C for 4 hrs. Finally, a thin film *a*-Si:H *p*-*i*-*n* junction solar cell was fabricated on the fully cured nanocone PI substrate, as shown in Fig. 1a4. The detailed fabrication process can be found in the Experimental Section and Fig. 1a4 indicates each layer in the device. Note that, a Ag layer was used at the bottom of device as a back reflector and also bottom electrode. An 80 nm thick aluminum doped zinc oxide (AZO) layer serves as a buffer layer to reduce the metal diffusion into the silicon layer [31–33]. Fig. 1b–d shows scanning electron microscopy (SEM) images of the as-obtained thin film *a*-Si:H devices based on flat PI substrates prepared

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