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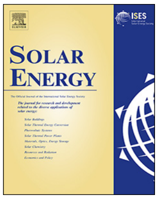

Enhancing photovoltaic performance of perovskite solar cells with silica nanosphere antireflection coatings

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ABSTRACT

Organic-inorganic halide perovskite solar cells have enormous potential to impact the existing photovoltaic industry. As realizing higher power conversion efficiency (PCE) of the solar cell is still the most crucial task, a great number of schemes were proposed to minimize the carrier loss by optimizing the electrical properties of the perovskite solar cells (PSCs). Here, we focus on another significant aspect that is to minimize the light loss by using an antireflection coating (ARC) component to gain a high PCE for PSC devices. In our scheme, silica nanosphere based ARCs are employed to $CH_3NH_3PbI_3$ PSCs for enhancing the device efficiency. SiO_2 nanosphere based ARCs were grown by spin-coating of an aged silica sol. The microstructure and the thickness of the SiO_2 nanosphere based ARC were controlled by changing the spin-coating speed from 400 to 4000 rpm. The effect of SiO_2 nanosphere based ARCs on the photovoltaic performance of perovskite solar cells is systematically investigated. The optimized SiO_2 nanosphere ARC coating on cleaned glass substrate exhibited a maximum transmittance of 96.1% at $\lambda = 550$ nm wavelength, and averagely increased the transmittance by about 3.8% in a broadband of 400–800 nm. The optimized antireflection coating strongly suppressed broadband and wide-angle reflectance in typical PSC solar cells, significantly enhancing the omnidirectional photovoltaic (PV) performance of PSCs. As a result, the power conversion efficiency was improved from 14.81% for reference device without SiO_2 nanospheres to 15.82% for the PSC device with the optimized ARC. Also, the PV performance of the PSC device with the optimized SiO_2 nanosphere ARC revealed less angular dependence for incident light.

1. Introduction

Increasing energy demand, environmental issues and limited availability of fossil fuels are demanding sustainable and renewable energy resources. Solar photovoltaic (PV) technology provides an eco-friendly and renewable energy route to directly convert photon energy into electricity (Kerr, 2007). After decades of research silicon (Si) solar cells have achieved record device efficiencies above 26% (Green et al., 2018). Confirmed terrestrial module efficiencies of crystalline Si solar cells are above 24% and of multicrystalline Si solar cells are close to 20%. However, the efficient establishment of solar cell technology on a global scale requires development in terms of materials and device store to reduce the fabrication cost and increase the power conversion efficiency (PCE). As a new entrant in the PV field, organometallic halide perovskites based photovoltaic device offers the advantages of high-efficiency, low-cost, and facile solution processability, making it one of the most promising next-generation PV technologies for the future. In perovskite solar cells (PSCs), halide perovskites are of the form ABX_3 (A is an organic ammonium ion: $CH_3NH_3^+$ (MA^+) or $NH_2CHNH_2^+$ (FA^+), B is Pb^{2+} or Sn^{2+}, X is the halogen ion: Cl, Br or I) as the efficient light harvester (Green et al., 2018; Kojima et al., 2009; Burschka et al., 2013; Liu et al., 2013; Yang et al., 2017; Zhao and Zhu, 2016). These perovskite materials possess some fascinating properties, such as tunable bandgap (Noh et al., 2013), high absorption coefficients (Xing et al., 2013), long charge carrier (electron–hole) diffusion length (Stranks et al., 2013), and low-temperature solution processability (Wang et al., 2014). Over the past few years, numerous efforts and extensive investigations have been made into the morphology control of each functional layer (Haque et al., 2017; Yang et al., 2016), compositional engineering of perovskite materials (Hao et al., 2014; Jeon et al., 2015), device structure design (Burschka et al., 2013; Lee et al., 2012; Gao et al., 2014; Cao and Xue, 2014; Tan et al., 2014; Zhang et al., 2017), crystallization modulation of perovskites (Jeon et al., 2014; Xiao et al., 2014; Im et al., 2014) and interfacial engineering (Zhou et al., 2014;

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Zhang et al., 2017; Luo et al., 2017; Assadi et al., 2018). Consequently, the power conversion efficiencies (PCEs) of PSC devices have been boosted from 3.8% to 22.7% under 1 sun conditions (100 mW cm^{-2} AM 1.5G) (Green et al., 2018).

The utilization of highly transparent antireflection coating (ARC) in perovskite solar cells provides an important approach to reduce or suppress Fresnel reflection losses, to increase the amount of light entering the PV device and hence, to enhance the power conversion efficiency of PSCs. PSCs typically are formed on transparent conductive oxide (TCO) glasses, such as fluorine doped tin oxide (FTO) and indium tin oxide (ITO) slides. There is a trade-off in PSC devices between the sheet resistance of the TCO and its optical transmittance. Whilst the use of thicker TCOs can significantly reduce the sheet resistance, it will also result in a decrease in transmittance. Thus, the optical transmittance of FTO or ITO slides is limited, which has a considerable impact the efficiency of PSCs. Currently, the ARCs have been widely employed on silicon (Si) or transparent substrates (e.g., glasses, plastics, etc.) in different types of Si based or organic solar cells or modules to reduce the surface reflection losses over a broad range of incident wavelengths and angles (Xu et al., 2008; Sakhuja et al., 2014; Raut et al., 2011; Leem et al., 2014a, 2014b; Ballif et al., 2004).

An important and well-known phenomenon lowering the overall PCE of solar cell devices is Fresnel reflection, which takes place when a refractive index contrast exists between the ambient medium (air) and the solar cell device. The reflectance losses due to the refractive index mismatch between a material and the air can be reduced by applying an ARC with an intermediate refractive index between air and the substrate material (Prevo et al., 2005). The antireflection coatings can either be materials with an intermediate refractive index between the substrate and air or porous or refractive index-graded materials (Mahadik et al., 2015). Most of these antireflection coatings (ARCs) are manufactured by film deposition techniques such as chemical vapor deposition (CVD), sputtering, or evaporation, as well as possible lithography steps. These methods are costly and often involve high-temperature environments that are unsuitable for certain solar cell technologies (Wan et al., 2013; Ali et al., 2014; Lopez et al., 2013). Silicon dioxide (SiO_2) is the most ideal material for ARC due to its low refractive index, good durability and environmental resistance. Especially, SiO_2 nanospheres, which do not require vacuum-based or high-temperature fabrication processes, offer a promising nanophotonic ARC option due to their light trapping and scattering properties (Grandidier et al., 2011, 2012). Further, SiO_2 nanoscale structures are stable with respect to extreme temperatures and light illumination. Different techniques have been used to achieve layers of SiO_2 nanospheres on top of photoactive layers on flexible and rigid substrates, including the Langmuir–Blodgett method (Grandidier et al., 2013; Hsu et al., 2008), sedimentation (Jiang et al., 1999), and controlled evaporation (Mihi et al., 2011). Although these fabrication approaches are simple, scalable and inexpensive and could enable large-scale manufacturability of microresonator-based photovoltaics, the application of nanosphere based ARCs on photovoltaic devices are scarcely reported.

In this study, we demonstrate a facile and simple approach to prepare silica nanosphere based antireflection coatings. SiO_2 nanosphere based ARCs were grown by spin-coating of an aged silica sol. The structure and the thickness of the SiO_2 nanosphere based ARC were finely adjusted by changing the spin-coating speed from 400 to 4000 rpm. The microstructure and the effective refractive index of the SiO_2 nanosphere based ARC were optimized to achieve high transmittance. The effect of SiO_2 nanosphere based ARCs on the photovoltaic performance of perovskite solar cells is systematically investigated. The optimized SiO_2 nanosphere ARC coating on cleaned glass substrate exhibited a maximum transmittance of 96.1% at $\lambda = 550$ nm wavelength, and averagely increased the transmittance by about 3.8% in a broad wavelength range of 300–800 nm, therefore leading to a significant increase in the efficiency of the PSC device from 14.81% to 15.82%, demonstrating over 6.8% enhancement, compared with the reference device without SiO_2 nanospheres. The present process provides a simple and cost effective method for the preparation of antireflection coatings, which benefit the development of PSC solar cells, as well as the light-harvesting scheme applicable to a wide variety of optoelectronic devices.

2. Experimental details

2.1. Preparation of the sols and deposition of the thin films

Tetraethoxysilane (TEOS) was procured from Sigma Aldrich. TEOS was used as a precursor for SiO_2 nanosphere ARC coatings. Ethanol also obtained from Sigma Aldrich was used as the solvent NH_3•$3H_2O$ was purchased from Aladdin Industrial Corporation and used for the catalysis of the sol.

A hybrid silica sol was prepared by mixing the precursor materials (Jun et al., 2008). Initially, 1.5 mL of $NH_3 \cdot 3H_2O$ solution (10 wt%) was mixed with deionized (DI) water (6 mL), ethanol (70 mL) in a 100 mL flask by magnetic stirring. Then, 3 mL of TEOS was added drop by drop to the mixture. The final mixture was magnetically stirred for 30 min. The silica sol obtained was kept at room temperature (RT) for at least 1 day for aging and finally used for deposition on external-facing surfaces of TCO transparent substrates of PSCs.

2.2. Device fabrication

FTO slides (Pilkington TEC 15) were patterned by etching with Zn powders and 2 M HCl. The etched slides were then cleaned with liquid detergent, acetone, ethyl alcohol and DI water for 15 min, sequentially, to remove the organic or inorganic residues, and finally dried in a vacuum oven. Titanium isopropoxide (TTIP) (200 μL) and ethanol (5 mL) were mixed to prepare a clear precursor sol. The precursor sol was spin-coated onto the etched FTO substrate at 4500 rpm, followed by annealing at 500 °C to form a compact TiO_2 (c-TiO_2) layer. Diluted TiO_2 pastes were prepared by mixing TiO_2 paste (Dyesol 18NR-T, 0.1 g) and anhydrous ethanol (444 μL). The mesoporous TiO_2 (p-TiO_2) layer was deposited on the c-TiO_2 layer by spin-coating the diluted TiO_2 pastes at 2000 rpm for 30 s. The layers were then sintered at 500 °C for 30 min in air. After cooling down to the RT, the samples were treated using the $TiCl_4$ aqueous solution at 70 °C for 30 min and dried at 500 °C for 30 min. The perovskite $MAPbI_3$ absorber was grown via a spin-coating process using a GBL/DMSO solution of PbI_2 and MAI. Briefly, a pure perovskite-precursor solution (1.25 mol L^{-1}) was prepared by mixing MAI (0.1975 g) powders and lead iodide PbI_2 (0.5785 g) in GBL (700 μL) and DMSO (300 μL) at 60 °C for 12 h. The formed precursor solution was deposited onto the p-TiO_2/c-TiO_2/FTO sample via a successive two-step spin-coating process, at 2000 rpm for 30 s and at 3500 rpm for 50 s, respectively. Anhydrous diethyl ether was dripped onto the center of the sample in the second spin-stage during the spin-coating process (Luo et al., 2017). The perovskite-precursor coated sample was heated and dried on a hot plate at 110 °C for 30 min. The hole-transport material (HTM) was deposited on the grown perovskite absorber by spin-coating a spiro-OMeTAD solution at 4000 rpm for 30 s. 100 nm thick AgAl alloy film with an active area of 0.1 cm^2 was formed via evaporation on the Spiro-OMeTAD-coated film (Jiang et al., 2016). Finally, SiO_2 nanosphere based ARCs were deposited on the front glass surface of the as-prepared PSC solar cell by spin-coating of the aged silica sol with various spin-coating speeds from 400 to 4000 rpm.

2.3. Characterization

The morphologies of SiO_2 nanosphere based ARC films were characterized by a high-resolution field emission scanning electron microscope (FESEM, Hitachi S4800). Optical spectra of SiO_2 ARCs were examined and characterized by means of ultraviolet–visible light (UV–vis) spectrometer (Hitachi, U-3010). Photocurrent density–voltage (*J-V*)