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Strategies for high performance perovskite/crystalline silicon four-terminal tandem solar cells

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ABSTRACT

In this work, we report systematic studies on improving the optical and electrical properties of four-terminal perovskite/c-Si tandem solar cells. Light harvesting power of the device is significantly enhanced due to the complementary absorption spectra of the perovskite and c-Si absorber materials. To obtain high power conversion efficiency (PCE) for the device, careful engineering of optoelectronic properties of the devices are accomplished through: 1. Oxygen annealing treatment for reducing defect density of perovskite materials; 2. Optical engineering of the transparent electrode (MoO_3/Au/MoO_3) to obtain high transmission at long wavelengths for the tandem solar cell applications; and 3. Enhancement of light harvesting power achieved by using the novel biomimicking elastomeric petals as the light trapping layer. The individual perovskite solar cell (PSC) with MoO_3/Au/MoO_3 electrode with or without light trapping layer yields an average PCE of 16.6% and 16.0% respectively. By combining c-Si bottom cell with perovskite top cell mechanically, an overall PCE of 22.4% is achieved for the averaged value, which is a promising result for future development of perovskite based tandem solar cells.

1. Introduction

The development of metal halide perovskite, a low-cost material with a crystal structure of calcium titanate, has recently made tremendous progress towards photovoltaic (PV) applications in terms of efficiency [1,2] and stability [3–5]. In 2009, Kojima *et al.* [6] reported PV properties of perovskite and utilized it as sensitizers in photoelectrochemical cells, yielding a PCE of 3.8%. The first all solid-state PSC was demonstrated by Kim *et al.* with a PCE of 9.7% [7]. After these pioneer studies, enormous attentions from scientists worldwide have been attracted to the perovskite materials, triggering intensive research on its PV properties and applications. Today, only a few years from its first report, PSCs exhibit skyrocketing enhancements in efficiencies and already reached a certified PCE of 22.7% [8]. The rapid enhancement in the efficiency achieved by PSCs is attributed to the physical properties of perovskite materials such as their broadly tunable bandgaps [9], high absorption coefficients over a wide range of visible light spectrum [10], long carrier diffusion lengths [11,12], good crystallinity [13,14] and high carrier mobilities [14–16]. The desirable properties of the perovskite and the promising results reveal that PSCs have huge potential to compete with the state-of-the-art PV technologies such as copper indium gallium diselenide (CIGS) solar cells and crystalline silicon solar cells (c-Si). It is noteworthy that c-Si solar cells have been dominating the PV market for several decades with relatively marginal enhancement in performance over the past years. The world record PCE for Si-based solar cells is currently at 26.6% [8] achieved by the amorphous silicon/crystalline silicon heterojunction structure, which is approaching the theoretical limit of 29.4% [17]. Considering the fact that Si solar cells have strong PV response at long wavelengths, combining Si solar cells with other types of solar cells with strong PV response at

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short wavelengths is an effective strategy to overcome the efficiency bottleneck. High efficiency tandem solar cells have been developed by stacking wide-bandgap compound semiconductor such as gallium arsenide [18] and indium gallium phosphide [19] on top of Si. However, these classes of solar cells are prohibitively costly for commercial terrestrial applications. Perovskite materials with tunable bandgap energies and low fabrication cost are promising candidates for the top cell in tandem with the small bandgap c-Si solar cell. The perovskite/c-Si tandem structure offers an effective approach to broaden the PV response to the solar spectrum and to minimize loss due to thermalization by adopting light absorbers with complementary absorption spectra. Recently, several groups have successfully demonstrated perovskite/Si tandem solar cells in different structures with promising PCEs. Chen *et al.* [20] have reported that using an efficient copper (Cu)/ gold (Au) semitransparent electrode yields a PCE of 16.5% for individual perovskite top solar cell and a PCE of 23.0% for perovskite/Si tandem solar cells. Bailie et al. [21]. utilized a transparent silver (Ag) nanowire electrode on PSC to achieve a PCE of 12.7% for individual perovskite top cell and a PCE of 17.0% for perovskite/Si tandem solar cells (18.6% for perovskite/CIGS tandem solar cells). Albrecht *et al.* [22] and Werner *et al.* [23] have demonstrated perovskite/Si tandem solar cell in monolithic structure with the PCE of 18% and 21.2% respectively. The high PCE values obtained from different groups clearly indicate the huge potential of using the tandem configuration.

A tandem solar cell is generally designed in three different configurations: monolithic two-terminal tandems with a tunnel junction (recombination layer) [24,25]; monolithic three-terminal tandems with the middle electrode being shared between the two sub cells [26,27] or mechanically stacked four-terminal tandems [28,29]. Several review papers have summarized and compared different types of perovskite based tandem devices [30–32]. In a monolithic two-terminal tandem system, the top cell is directly fabricated on the bottom cell, which allows simplified manufacturing steps and minimizes parasitic absorption due to the transparent conductive oxide (TCO) electrodes such as indium tin oxide (ITO) and fluorine doped tin oxide (FTO). However, the total current of a monolithic two-terminal tandem cell is determined by the lowest of the currents generated in either the top cell or the bottom cell [33]. Therefore, precise management of current generation in each individual sub cell is required to avoid current mismatch between the top cell and the bottom cell in monolithic two-terminal tandem structures. The compatibility of the fabrication processes for the top and bottom cells have to be considered as well. Monolithic three-terminal tandem systems can be also regarded as a parallel tandem, for which a mutual interlayer electrode with high electrical conductivity and good optical transparency is used to connect the sub cells with complementary absorption in parallel. This configuration allows the optimization of the current of each sub cell independently. However, compatibility of the fabrication processes during the monolithic growth is still needed. Mechanically stacked four-terminal tandem devices offer larger flexibility for different combinations of the top and bottom cells as the sub cells are mechanically stacked with independent connections between the top and bottom cells. Therefore, the issues of the current matching and the fabrication compatibility between the sub cells are not key concerns. Additional TCO electrodes involved in the four-terminal tandem devices will cause additional parasitic absorption, leading to current loss in the devices and thus the costs for materials, transportation and installation will increase due to the extra materials needed and the complexity of electrical connections. In the optical aspect, on the other hand, optical engineering such as different light trapping strategies can be employed to minimize the reflection losses at the various interfaces. Nevertheless, the four-terminal tandem architecture provides much more freedom for engineers to pair up the high-performance sub cells having complementary absorption spectra, facilitating the optimization of the module transmissivity of each sub cell. For the case of perovskite/c-Si tandem devices, perovskite solar cells can be used as an add-on device placed on top of the commercial Si-based solar cells to further enhance the efficiency. It is noted that the semitransparent perovskite solar cell exhibits a PCE difference when it is illuminated from different sides of the electrodes due to the different transmittance of the electrodes and the parasitic absorption of the carrier transport layers (e.g. Spiro-MeOTAD). In the case of 4 terminal tandem devices, the side with larger transmission can be chosen as the illumination side. With proper integration of external circuit connections, four-terminal tandem devices can be effectively applied in practical applications such as the building-integrated photovoltaic (BIPV) system.

Previously, our group has successfully fabricated high performance single junction $CH_3NH_3PbI_3$ devices with PCE over 15% [34] for solution process and 17.6% for hybrid chemical vapor deposition process (HCVD) via careful controlling of the trap density in the bulk of the perovskite and material interfaces through post-deposition oxygen annealing treatment [34,35]. The next step to further boost the solar cell efficiency to a higher value desired for practical application is to combine the PSCs with c-Si solar cells in a tandem configuration. In this work, we report the development of high performance perovskite/c-Si four-terminal tandem devices accomplished by detailed material optimization and optical engineering of the devices. The strategy focuses on three aspects: (i) Enhancement of the transmissivity of the transparent rear electrode of the top cell; (ii) Passivation of defects in $CH_3NH_3PbI_3$ devices by post-deposition oxygen annealing; and (iii) Maximization of the light harvesting power of the tandem devices.

A number of techniques for preparing high transparency electrodes for perovskite top cells in tandem structures have been reported such as synthesis of silver nanowires [21], deposition of TCO by sputtering process [28,29,36], application of conducting multilayer [37], graphene [38] and poly(3,4,-ethylenedioxythiophene) (PEDOT: PSS) [39]. Among different techniques, the transmission spectra of multilayer electrodes can be easily tailored in accordance with the absorption spectra of the absorbers used by adjusting the individual thickness of the constituent layers to increase the transparency of the electrode at certain wavelengths. A comprehensive investigation of optical and electrical properties of semi-transparent transition metal oxide/metal/ transition metal oxide multilayer electrode has been conducted in this work. The transparency of the transition metal oxide/metal/ transition metal oxide multilayer is then optimized based on the theoretical calculations specifically for the application in perovskite/c-Si four-terminal tandem solar cells.

Secondly, post-deposition oxygen treatment has been applied for achieving high performance MAM based perovskite devices. Both theoretical and experimental results show that oxygen is an essential element to reduce the localized states in the bulk of perovskite as well as at the material interfaces of the devices, yielding improved PV performance [34,35,40]. Furthermore, a light trapping strategy for increasing the effective optical paths passing through the absorber layer and thus enhancing the light harvesting power is incorporated into our perovskite/c-Si four-terminal tandem solar cells. It is demonstrated that a biomimicking elastomeric petal (BEP) replicated from natural rose petals can be used as versatile substrates for stretchable thin-film electronics. This is because the unique three-dimensional microscale crater-like topographies of the BEP prohibit the propagation of microcracks in the conducting films that are deposited on the elastomeric petal substrates during large-strain deformation [41]. It is recently discovered that such unique biological surface texture also behaves as an effective light scattering layer [42], exhibiting ultrahigh transmission haze of 75% and high diffusion transmittance of 97%. Therefore, the BEP can also be placed on the surface of the perovskite top cell to scatter the incident solar light to enhance the light harvesting power of the tandem structure. With the optimized optical properties for the device structure and careful defect engineering of the perovskite layer by low-temperature oxygen annealing, we have accomplished MAM-PSC with an average PCE of 16.6%. The device is utilized to form a tandem structure with a PCE 19.1% of c-Si cell and the tandem structure demonstrates an