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Improved performance and thermal stability of perovskite solar cells prepared via a modified sequential deposition process

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ABSTRACT

In conventional sequential deposition, PbI_2-free $CH_3NH_3PbI_3$ perovskite films can be achieved when the PbI_2 substrate reacts with a relatively low-concentration CH_3NH_3I solution (<8 mg of CH_3NH_3I per 1 mL of IPA) but the film that is formed has only sparse and uneven coverage. On the other hand, a dense $CH_3NH_3PbI_3$ perovskite film can be formed on a mesoporous TiO_2 substrate when the PbI_2 substrate reacts with a relatively high-concentration MAI solution (>8 mg of CH_3NH_3I per 1 mL of IPA), but unreacted PbI_2 is still present in $CH_3NH_3PbI_3$ layer in such cases. Here, we developed organic-inorganic perovskite solar cells with high efficiency by preparing dense and PbI_2-free films using a modified sequential deposition process. By utilizing the reaction behavior between PbI_2 and MAI, solar cells with an average power-conversion efficiency of 15.1% and high reproducibility and stability were achieved.

1. Introduction

Organic-inorganic hybrid perovskite materials have emerged as light absorbers in solar cells because of their high absorption coefficients, band gap tunability, long diffusion lengths and mobility [1–3], with organic-inorganic hybrid perovskite solar cells first being introduced in 2009 [4]. However, their less than adequate stability must be addressed before widespread use can be realized. To improve cell efficiency and stability, Park et al. reported stable perovskite solar cells using spiro-OMeTAD instead of a liquid electrolyte, optimizing cell fabrication to achieve 9.7% efficiency [5]. Grätzel et al. reported a sequential deposition process for obtaining a uniform perovskite layer [6], a step-by-step process by which PbI_2 was coated onto the substrate and then reacted with CH_3NH_3I (MAI) solutions; they obtained over 15% efficiency for their perovskite solar cells. In 2014, Seok et al. reported 17.9% efficiency for mixed perovskite solar cells [7], by mixing $HC(NH_2)_2PbI_3$ and $CH_3NH_3PbBr_3$ perovskite materials to achieve both stability and high efficiency. This same group recently reported the materials development and fabrication process for perovskite solar cells with 22.1% efficiency [8].

There are several methods available for fabricating perovskite films, such as single-step deposition, sequential deposition, and vapor-assisted solution processing [5,6,9,10]. In sequential deposition, the size of the perovskite crystal and the conversion rate can be altered by controlling several factors such as solution and substrate temperature, and PbI_2 and MAI concentration [11–14]. Sequential deposition is advantageous in that more uniform and dense perovskite films can be formed compared to conventional one-step deposition methods [6]. However, the resulting films have unreacted PbI_2 because reacting the inner area of the PbI_2 layer with MAI is difficult due to the initially formed compact $MAPbI_3$ layer on the PbI_2 surface [15]. There are several methods available for complete conversion of PbI_2 layers into perovskite layers. Huang et al. reported a solvent annealing method where a $MAPbI_3$ film was annealed in the presence of dimethylformamide (DMF) solvent [16]. DMF assists in controlling the lattice parameter in perovskite layers for easy intercalation of organic iodides. Thus, MAI molecules can move to proper sites in the lattice during solvent annealing.

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