Organic Electronics 27 (2015) 12–17

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Simple fabrication of perovskite solar cells using lead acetate as lead source at low temperature

Jian Qing [a, b], Hrisheekesh-Thachoth Chandran [a, b], Hong-Tao Xue [a, b], Zhi-Qiang Guan [a, b], Tai-Li Liu [b], Sai-Wing Tsang [b], Ming-Fai Lo [a, b, c], Chun-Sing Lee [a, b, c, *]

[a] Center of Super-Diamond and Advanced Films (COSDAF), City University of Hong Kong, Hong Kong, PR China
[b] Department of Physics and Materials Science, City University of Hong Kong, Hong Kong, PR China
[c] City University of Hong Kong Shenzhen Research Institute, Shenzhen, PR China

ARTICLE INFO

Article history:
Received 2 June 2015
Received in revised form
21 August 2015
Accepted 22 August 2015
Available online xxx

Keywords:
Perovskite solar cells
Lead acetate
Solvent annealing

ABSTRACT

We reported a simple one-step, low-temperature solution-processed technique to fabricate perovskite solar cells using lead acetate as the lead source. Solvent annealing was applied for grain growth to obtain better morphology. Uniform perovskite films without pinholes can be obtained by solvent annealing for 5 min at 100 °C. Planar perovskite solar cells based on the high quality perovskite films deliver power conversion efficiency up to 12.71% with negligible hysteresis and good reproducibility. In addition, the substrate surfaces have little effect on the crystallization of perovskite when lead acetate was used, leading to uniform films on different substrates, which can provide a wide choice of substrates and interfacial materials.

1. Introduction

Methylammonium lead halide perovskites, such as $CH_3NH_3PbI_3$ and $CH_3NH_3PbI_xCl_{3-x}$ have emerged as promising photon absorbers for solar cell application because of their high optical absorbances, long charge diffusion lengths and small exciton binding energies [1–4]. Moreover, their solution processability and the earth abundance of their constituting materials endow them with high potential for the next generation solar cells [5–7]. High performance perovskite solar cells (PVSCs) have been reported with both mesoporous structures and planar heterojunction architectures with power conversion efficiency (PCE) over 15% [8–12]. Mesoporous structure devices usually require high temperature (over 450 °C) processing, which hinders their applications on typical glass or polymer substrates [8]. While the planar structure devices, which employ organic semiconductors as hole and electron transporting layers, can be fabricated at low temperature, offering a wide choice of substrates, electrodes and interfacial materials [6].

Various deposition techniques, including vacuum evaporation [13], one-step spin-coating [5,9], two-step sequential deposition [8,14] and vapour-assisted solution process [15], have been developed for preparing perovskite films. Both one-step spin-coating and two-step sequential deposition are all-solution-processes, which are compatible for large scale, cost-effective manufacturing. In the simple one-step spin-coating method, a metal halide (lead iodide (PbI_2) or lead chloride ($PbCl_2$)) is dissolved with methylammonium iodide (MAI) in a polar solvent (such as dimethylformamide (DMF)) as the precursor solution. This solution is then spin-casted onto the substrate followed by thermal annealing to form the perovskite layer. Nevertheless, discontinuous perovskite films with pinholes are usually obtained, leading to current leakage and limiting the device performance [7,16]. Besides, relatively long annealing time (typically 1–2 h) is required for the formation of perovskite [6,17]. In the two-step sequential method, PbI_2 solution is first spin-casted onto the substrate, followed by dipping into or spin-coating MAI solution to produce the perovskite layer. Although this two-step method provides better control over the film morphology, a porous scaffold is needed for the complete transformation of PbI_2 into perovskite layer. Incomplete conversion of PbI_2 usually exists in planar structure, which causes problem in devices reproducibility [18,19]. To achieve better performance, many different approaches have been developed to further improve the quality of the perovskite films, including solvent engineering

* Corresponding author. Center of Super-Diamond and Advanced Films (COSDAF), Department of Physics and Materials Science, City University of Hong Kong, Hong Kong, PR China.
E-mail address: apcslee@cityu.edu.hk (C.-S. Lee).

http://dx.doi.org/10.1016/j.orgel.2015.08.021
1566-1199/

[20], solvent annealing [21], fast deposition-crystallization [22], interface engineering [23–25] as well as adding various additives [26,27].

Recently, lead acetate ($Pb(Ac)_2$) was reported as a new lead source for efficient PVSCs [28,29]. Using $Pb(Ac)_2$ has the advantage that perovskite crystal growth can be accelerated because of the facile removal of CH_3NH_3Ac. As a result, ultrasmooth and almost pinhole-free perovskite film is obtained, which in turn leads to better device performance. However, the perovskite film consists of small grain size crystals owning to the faster rate of crystallization, which could increase the overall bulk defect density and trap states at grain boundaries [10,22]. On the other hand, Huang et al. showed that solvent annealing can be applied to perovskite films to effectively increase the crystallinity and grain size. However, the annealing time (100 °C for 1 h) for the two-step spin-coating is also long [21].

Based on the two works, here we report a simple one-step, low-temperature solution process to fabricate uniform $CH_3NH_3PbI_3$ layers without pinholes using $Pb(Ac)_2$ as the lead source. High quality perovskite films can be obtained by solvent annealing for only 5 min, which can deliver high device efficiency of 12.71%. We also found that this approach allows formation of uniform perovskite films on different substrates including Si, bare glass, ITO, FTO and PEDOT:PSS coated ITO glasses.

2. Experimental

2.1. Perovskite precursor solution preparation

$Pb(Ac)_2$ was first prepared by dehydration of $Pb(Ac)_2 \cdot 3H_2O$ (Sigma–Aldrich) at 80 °C under flowing N_2. To prepare the perovskite precursor solution, 3 mmol MAI (Lumtech.) and 1 mmol $Pb(Ac)_2$ were dissolved in 1 mL anhydrous DMF (RCI Labscan). The solution was then stirred at room temperature overnight.

2.2. Device fabrication

The devices have structure of ITO/PEDOT:PSS (40 nm)/$CH_3NH_3PbI_3$ (250 nm)/PCBM (60 nm)/BCP (10 nm)/Ag (70 nm). The indium tin oxide (ITO) coated glass substrates were cleaned as routine [30]. Poly(3,4-ethyl-enedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, Baytron P-VP AI4083) was spin-coated onto

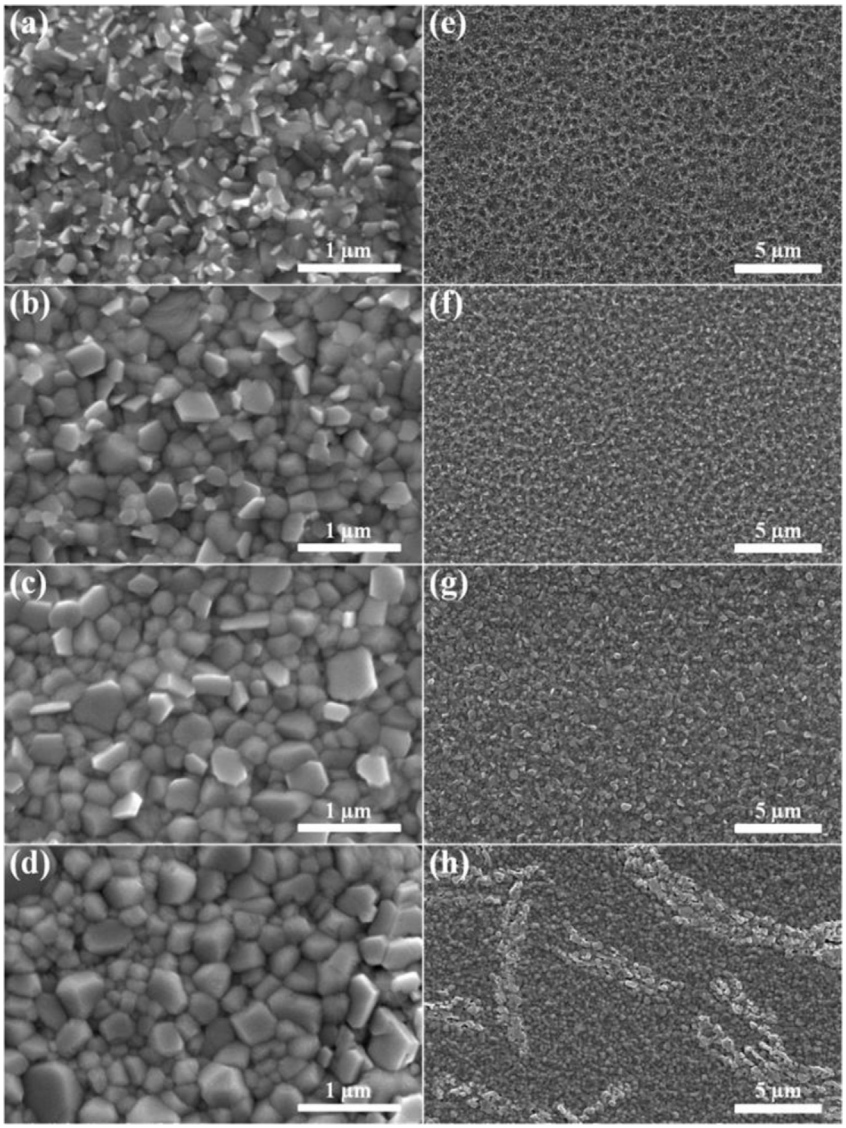

Fig. 1. Surface SEM images of perovskite films. (a, e) Thermal-annealed for 5 min and solvent-annealed for different times: (b, f) 5 min, (c, g) 10 min and (d, h) 20 min.