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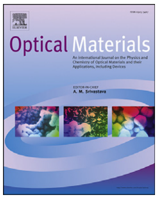

Growth of $MAPbBr_3$ perovskite crystals and its interfacial properties with Al and Ag contacts for perovskite solar cells

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ABSTRACT

In this work, the $MAPbBr_3$ perovskite crystals were grown and the interfacial properties of the poly-crystalline $MAPbBr_3$ with Aluminum (Al) and Silver (Ag) contacts has been investigated. $MAPbBr_3$ crystals are turned into the poly-crystalline pellets (PCP) using compaction technique and the Al/PCP, Al/interface layer/PCP, Ag/PCP, and Ag/interface layer/PCP contacts were investigated. Scanning Electron Microscopic (SEM), Energy-dispersive X-ray spectroscopy (EDX) and current-voltage (I-V) characteristic technique were used to have an insight of the degradation mechanism happening at the Metal/perovskite interface. The Ag/PCP contact appears to be stable, whereas Al is found to be highly reactive with the $MAPbBr_3$ perovskite crystals due to the infiltration setback of Al in to the perovskite crystals. The interface layer showed a slight effect on the penetration of Al in to the perovskite crystals however it does not seem to be an appropriate solution. It is noteworthy that the stability of the underlying metal/perovskite contact is very crucial towards the perovskite solar cells with extended device lifetime.

1. Introduction

Since the invention of the use of perovskite materials for solar cells by Miyasaka and co-workers in 2009 [1], a significant advancement has been observed in this field of solar cells which led to improving efficiency from 3.8% to a qualified value of 22.1% on laboratory-scale cells in 2016. Perovskite solar cells consist of organic-inorganic hybrid system with general formula ABX_3, where A is an organic/inorganic cation (methylammonium (CH_3NH_3)), B is the metal cation (Pb), and X is a halide anion (I, Br, Cl). However due to number of factors, the perovskite solar cells are facing the issue of low stability. The high affinity of $CH_3NH_3PbX_3$ for moisture leads to degradation of the material to PbX_2 and thus the biggest hindrance on the way towards commercialization of perovskite solar cells is lack of environmental stability [2,3]. The reaction of the metal contact with the perovskite films is another important reason behind the instability of the perovskite solar cells. To overcome these discrepancies, and to obtain stable perovskite solar cells, diffusion barrier layers are inserted between metal electrodes and the perovskite layer to keep away from each other [4,5]. However, the shielding lifetime of buffer layers is very short and cannot be used to resolve the problem in the long run. Due to the repetitive illumination or thermal cycles, the electrode materials tend to diffuse through the buffer layers after several months or years of operation [6,7]. To limit the diffusion effect, various approaches including device hardening by introducing alternative perovskite active layer chemistries [8], integrating buffer layers [6,9] or by initiating more robust or hydrophobic hole-transporting layers (HTLs) and top electrodes [10] have been studied.

A careful and proper selection of a top electrode and a corresponding hole-transport layers on top of the perovskite layer, might help to safeguard the active layer [11,12]. Preceding studies have revealed stability issues with Ag [13,14] or Al [15] electrodes in perovskite devices. Kato et al. [16], have reported that $MAPbI_3$ iodizes Ag electrodes. Back et al. [17], employed a chemical inhibition layer to counteract corrosion of Ag and Al electrodes. 2,2′,7,7′-tetrakis (*N*,*N*-di-*p*-methoxyphenylamine)- 9,9′-spirobifluorene (spiro-OMeTAD) is the most extensively used HTL in high performance perovskite solar cells [18]. When utilized in perovskite solar cells as an HTL, the spiro-center aggravates crystallization while

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safeguarding appropriate penetration of the material into the mesoporous layer [19]. Nevertheless, the glass-forming affinity of spiro-OMeTAD layer remains as a major holdup in the progression of perovskite solar cell efficiencies. Recently, a molecularly engineered novel dopant-free hole transporting materials (HTMs) for perovskite solar cells (PSCs) have been reported [20]. These materials are more stable and efficient as well. For instance, the PSC based FA-CN showed exceptional stability up to 500 h. These reports open a new avenue for efficient and stable PSCs, exploring new materials in an alternative to Spiro-OMeTAD.

Au is well known as a top electrode material for perovskite solar cells, but it is expensive, so there is need to investigate the alternative metals which should be economical. In this work, the interfacial properties of $MAPbBr_3$ perovskite crystals were investigated with Al and Ag metal contacts. The aim of this work is to have an insight on the interfacial properties of the Metal (Al, Ag)/ perovskite ($MAPbBr_3$) interfaces and visualize their interface properties. The contacts were characterized using Scanning Electron Microscopic (SEM), Energy-dispersive X-ray spectroscopy (EDX) and current-voltage (I-V) techniques.

2. Experimental

2.1. Materials

Lead (II) bromide ($PbBr_2$)(for perovskite precursor) is purchased from TCI chemicals, Japan, Dimethylformamide DMF (anhydrous, 99.8%), Chlorobenzene, Acetonitrile, Bis(trifluoromethane)sulfonimide lithium salt (LiTFSI) and 2,2′,7,7′-tetrakis (*N*,*N*-di-*p*-methoxyphenylamine)- 9,9′-spirobifluorene (spiro-OMeTAD) were purchased from Sigma Aldrich. MABr were purchased from Dyesol Limited (Australia). We used the chemicals as received without further purification.

2.2. Preparation of $CH_3NH_3PbBr_3$ poly-crystals

The perovskite crystals were grown by the inverse temperature crystallization method. In brief, 1 M solution of the Lead (II) bromide ($PbBr_2$) and Methylammonium bromide (MABr) in 5 ml DMF solution is mixed at room temperature. Then the solutions were filtered using 0.2-mm pore size PTFE filters. The filtered solution is transferred to glass vials and kept in oil bath at 80 °C for 30 min. Then 60 μl of Formic acid (FA) was added to the solution and the crystallization was started within 5 min as shown in Fig. 1.

2.3. Preparation of $CH_3NH_3PbBr_3$ pellets

The small crystals (size ~0.5 mm) grown using inverse temperature crystallization method were then filtered without altering the temperature using filter paper. The dried crystals were then pressed under a pressure of 20 MPa using a hydraulic press which resulted in the formation of perovskite crystals pellet of diameter 1 cm and thickness 4 mm.

2.4. Deposition of spiro-MeOTAD HTL layer

Spiro-MeOTAD solution was prepared following previously reported method by Nam-Gyu Park et al. [21]. Briefly 72.3 mg of spiro-MeOTAD is dissolved in 1 ml of chlorobenzene, to which 28.8 μl of 4-*tert*-butyl pyridine and 17.5 μl of lithium bis(trifluoromethanesulfonyl)imide (Li-TFSI) solution (520 mg LI-TSFI in 1 ml acetonitrile) were added. After overnight stirring, the solution is spin coated over the crystal pellets using a one-step spin coating process at 2000 rpm for 30 s.

2.5. Characterization

X-ray diffractometer (PAN analytical EMPYREAN, Netherland)

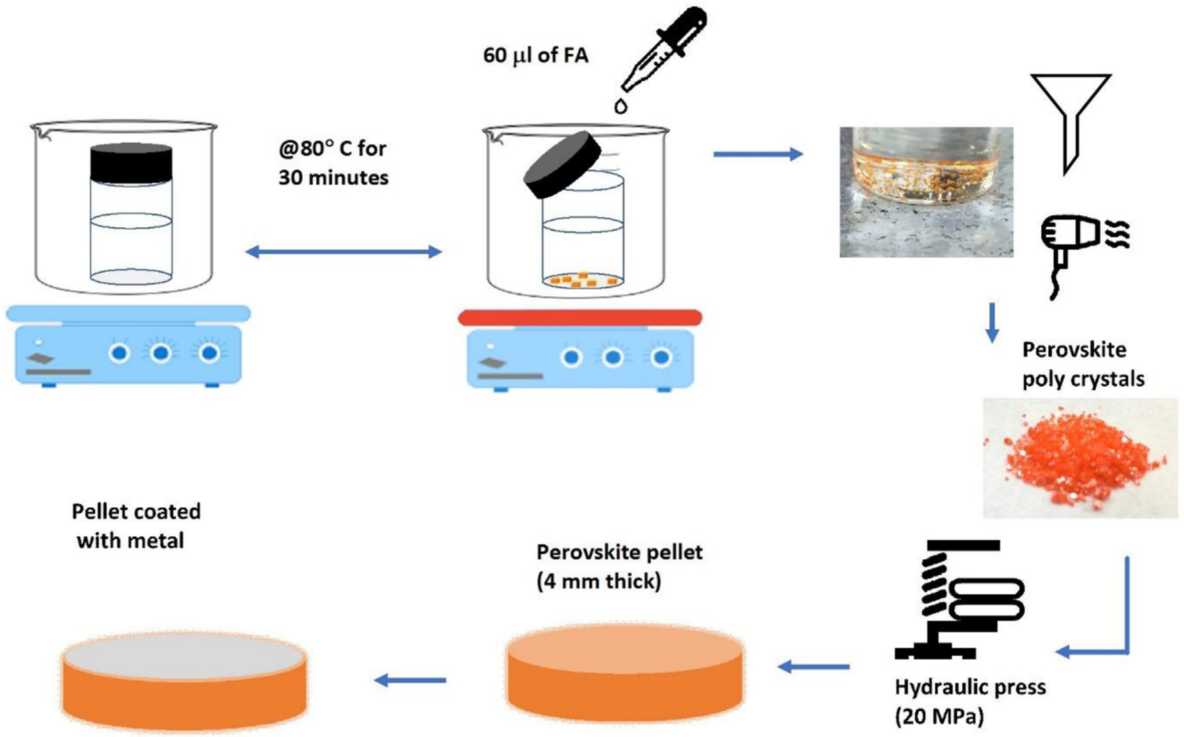

Fig. 1. Schematic diagram showing the perovskite crystal growth process and the pellet formation. The grown perovskite poly-crystals.