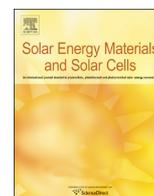




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Structural, optical and electrical properties of planar mixed perovskite halides/Al-doped Zinc oxide solar cells

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ABSTRACT

Mixed halide perovskites $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ thin films were successfully fabricated by spin coating using a precursor solution obtained by reacting a (1:1) molar ratio of methyl-ammonium iodide ($\text{CH}_3\text{NH}_3\text{I}$) and lead bromide (PbBr_2). The films obtained by this one step process were used to fabricate a solar cell using an aluminum-doped Zinc oxide (Al-ZnO) thin film as an electron selective layer. The Al-ZnO films were fabricated by RF sputtering on indium-tin-oxide (ITO) covered glass substrates. The structural and optical properties of the films were analyzed by X-ray diffraction and ultraviolet–visible spectroscopy. The mixed halide perovskite films were polycrystalline with an optical band gap of 1.85 eV. Al-ZnO/perovskite solar cells were fabricated without a hole-selective layer by directly deposition silver (Ag) contacts on the perovskite films. The electrical properties of the cells were investigated by current–voltage (*I*–*V*) measurements. The devices showed an open circuit voltage of 0.12 V, a fill factor of 0.29 and conversion efficiency of 1.6% under an illumination of 480 mW cm^{-2} . The film and cells are stable under ambient atmosphere as revealed by the absence of any changes in their color.

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1. Introduction

The term halide perovskites encompasses a class of compounds with a general chemical formula ABX_3 where A is an organic ammonium cation (in general CH_3NH_3^-), B is a metal (Pb or Sn) and X is a halide anion. These compounds exhibit peculiar properties that made them very attractive in the fabrication of thin film solar cells: excellent optical properties that are tunable by changing the chemical composition [1], ambipolar charge transport [2], and very long electron and hole diffusion lengths [2]. In addition, these perovskites are notably easy to prepare and to deposit by simple solution processing [3], including one-step spin-coating method, a two step sequential method [4] and physical vapor deposition under high vacuum [5].

Solar cells based on these halide perovskite compounds were found to exhibit excellent properties in mesoscopic [5,6] or planar heterojunction configurations [7,8]. In particular, methyl ammonium lead iodide, $\text{CH}_3\text{NH}_3\text{PbI}_3$ referred to as (MAPbI₃) with a band gap of 1.5–1.6 eV and a strong light absorption extending up to a wavelength of 800 nm, has been extensively used as light harvester in solar cells [9]. For example, when MAPbI₃ was loaded on a mesoporous TiO₂ electrode by sequential deposition of PbI₂ and methyl ammonium iodide (MAI), a 15% power conversion efficiency

was achieved under 1 sun illumination [10]. An efficiency of 15.4% with an open-circuit voltage of 1.07 V and a short circuit current density of 21.5 mA cm^{-2} were achieved in planar hetero-structure cells with a uniform mixed halide perovskite $\text{MAPbI}_{3-x}\text{Cl}_x$ deposited by vacuum thermal evaporation [7]. More recently, 19.3% efficiency was reported in planar device architecture [11], which is a record for solution processed cells. However, MAPbI₃ suffers from degradation under moisture, oxygen and ultraviolet radiation [12]. As a consequence, MAPbI₃-based cells were found to be unstable under atmospheric conditions, thus limiting their commercial use. The search of more stable perovskite phases is therefore required.

Mesostructured TiO₂ or Al₂O₃ planar electrodes have been widely used in these cells as a hole blocking electrodes. ZnO, on the other hand, is known to have an electron mobility that is substantially higher than that of TiO₂ [13–16], which makes it an ideal choice for an electron selective contact in such cells. Dong et al. [17] reported a 10.7% efficiency for perovskite solar cell based on ZnO nanowires. On the other hand, Mahmood et al. [18] used a ZnO thin film doped with aluminum deposited by electro-spray as an electron selective layer and achieved an efficiency of 10.8%. These cells however use MAPbI₃ as a light harvester and are therefore instable under atmospheric conditions.

In addition to its suitable optoelectronic properties, the interest in ZnO stems from the fact that thin films of this material doped with various elements (Al, Co, Ga) can be deposited by different methods such as, spray pyrolysis, chemical vapor deposition, sol gel process, sputtering, chemical spray, plasma enhanced CVD, pulsed

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laser chemical spray, plasma enhanced CVD and pulsed laser deposition [14–16].

In this work, we report on the fabrication of $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ perovskite/Al–ZnO planar heterojunction solar cells. The perovskite acts as an absorber and at the same time as a hole conductor. The $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ perovskite was used because the substitution of I with Br in MAPbI_3 greatly improved the stability of the cell in ambient atmosphere. The structure, microstructure and optical properties of the as-prepared films were investigated. In addition, perovskite/Al–ZnO heterojunctions solar cells were fabricated and their preliminary parameters were measured.

2. Experimental section

2.1. Synthesis of methyl ammonium iodide (MAI)

$\text{CH}_3\text{NH}_3\text{I}$ was synthesized by reacting methylamine and hydroiodic acid (HI, Sigma Aldrich) in a bottom flask at 0 °C for 2 h. The precipitate was collected by drying at 70 °C for 1 h and washed three times with diethyl ether and then dried at 100 °C for 24 h under vacuum [4].

2.2. Synthesis of perovskite $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$

The mixed halide perovskite precursor solution was obtained by dissolving methyl-ammonium iodide and lead bromide PbBr_2 at room temperature in a (1:1) molar ratio in dimethylformamide for 2 h. The films were then fabricated by spin-coating at a speed of 3500 rpm and solvent-dried on a hot plate at 100 °C for 30 min.

2.3. Synthesis of Al doped ZnO layer

Al-doped ZnO (Al–ZnO) thin films were prepared by reactive RF-sputtering in an ALCATEL SC451 deposition system equipped with an ALCATEL ARF 601 RF generator operating at 13.56 MHz, as described in previous works [19]. These films were deposited using a high purity Zn metal target with a diameter of 10 cm, partially covered by a high purity Al sheet in an argon/oxygen gas mixture.

The substrates were mounted at a distance of 10 cm above the Zn/Al target. The vacuum chamber was evacuated to a base pressure of 1.5×10^{-6} mbar. The target was pre-sputtered for 10 min and the substrates were not intentionally heated during the deposition. The reactive sputtering was carried out in an Ar/O₂ gas mixture with 30% of O₂. The films were deposited using an RF power of 200 W for 30 min.

2.4. Solar cell fabrication

Solar cells were fabricated on pre-cleaned ITO-coated glass substrates. First, Al–ZnO the perovskite thin films were sequentially deposited on the ITO-coated glass as described above. The perovskite film was dried at 100 °C for 30 min. The cell was completed by depositing silver dots on the surface of the perovskite film by thermal evaporation at a base pressure of 10^{-5} mbar. No hole-transporting layer was used in our case. A schematic diagram of the fabricated ITO/Al–ZnO/perovskite/Ag cell is presented in Fig. 1

2.5. Characterization techniques

The structural properties of the samples were investigated by X-ray diffraction with an X-pert MPD diffractometer (Panalytical) using the $\text{CuK}\alpha$ radiation. Optical transmittance measurements were performed using a Shimadzu UV-PC spectrophotometer in the 200–2000 nm range.

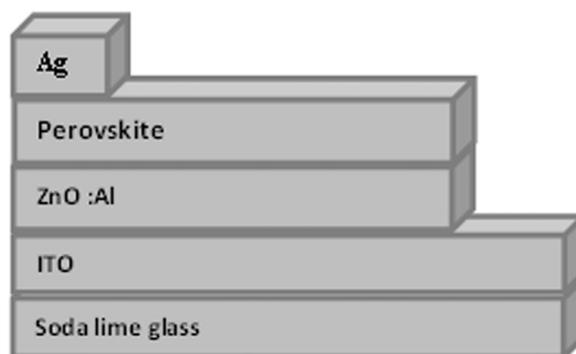


Fig. 1. Schematic representation of the Al–ZnO/ $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ solar cell.

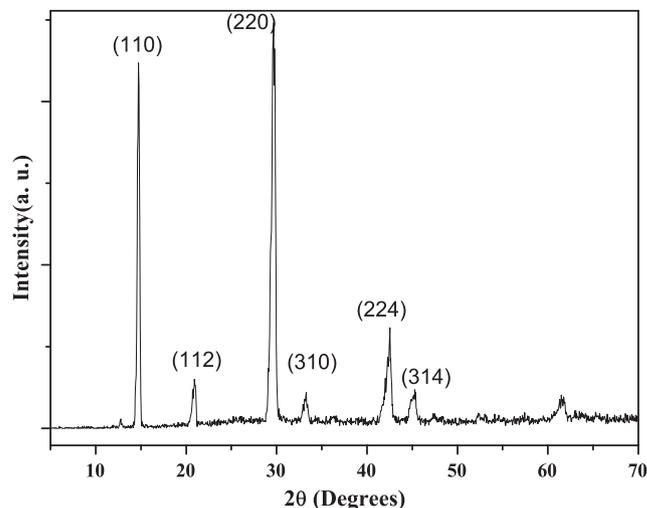


Fig. 2. XRD patterns of a mixed halide perovskite $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ film deposited on a glass substrate by spin coating.

The current–voltage (I – V) and impedance measurements were performed on the glass/ITO/Al–ZnO/perovskite/Ag devices at room temperature. The measurements were carried out using a Keithley 410 programmable pico-ammeter and a 610 C programmable micro-voltmeter. All the instruments are controlled by a computer via a GPIB card.

3. Results and discussion

The structure of the resulting $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ mixed halide perovskite films was investigated by X-ray diffraction (XRD) as shown in Fig. 2. The XRD patterns of the as-deposited films revealed the presence of peaks at the following 2θ values: 14.73°, 20.9°, 29.67°, 33.27°, 42.52°, 45.27° and 61.47°. These peaks correspond respectively to the reflections from the (110), (112), (220), (310), (224) and (314) lattice planes that are characteristic of the perovskite phase. When compared to $\text{CH}_3\text{NH}_3\text{PbI}_3$ [9], the diffraction peaks of $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ are shifted to higher Bragg angles indicating an effective substitution of I ions by Br ions in the lattice of MAPbI_3 (Fig. 2).

The crystallite size g of the films was estimated from the Full Width at Half-Maximum (FWHM) $\Delta(2\theta)$ of the (220) peak using Scherrer's formula:

$$g = \frac{0.94 \lambda}{\Delta(2\theta) \cos \theta}$$

where θ is the Bragg diffraction angle of the peak and λ is the X-ray wavelength. For the as-deposited film, this value is found to be on the order of 24 nm.

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