Partial coverage methylammonium lead bromide films for solar cell application

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Abstract

Hybrid perovskite compounds as methylammonium lead halides are currently been investigated for different optoelectronic applications. $\text{CH}_3\text{NH}_3\text{PbBr}_3$ is a promising member because of its electronic structure and large bandgap that gives rise to a high open circuit voltage. Although higher performances can result by using sophisticated processing, one-step methods are particularly interesting because of their easy implementation. Solar cells based on $\text{CH}_3\text{NH}_3\text{PbBr}_3$ are fabricated using one-step spin coating deposition. Remarkable effect on the morphological, structural, optical and electrical properties is observed by changing the molar ratio of the precursors. Dual film morphology, with micrometer-size $\text{CH}_3\text{NH}_3\text{PbBr}_3$ crystals embedded in a mesoporous TiO_2 background, explains minimal shunting paths and open-circuit voltage reduction. Full perovskite coverage of TiO_2 background is not necessary for proper operation.

Keywords: perovskite; film deposition; optical properties; photovoltaics; methylammonium lead bromide.

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1. Introduction

The successful introduction of hybrid perovskite compounds as methylammonium lead halides, acting as light absorber materials [1, 2], have fostered an unprecedented development of the so called perovskite solar cells (PSCs) reaching photovoltaic efficiencies as high as 24.2 % [3]. The use of these compounds has been extended into other optoelectronic [4-6], and even lasing [7, 8] applications. These devices basically consist of a layered architecture where the absorber perovskite is sandwiched between an electron transport material, regularly TiO_2 , and a hole transport material (HTM), usually made of the organic compound 2,2',7,7'-tetrakis(N,N-di-p-methoxyphenylamine)-9,9-spirobifluorene (*spiro*-OMeTAD) [9, 10]. The most commonly used absorbing materials are $\text{CH}_3\text{NH}_3\text{PbI}_3$ [11, 12] and $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ [13, 14], although $\text{CH}_3\text{NH}_3\text{PbBr}_3$ (MAPbBr₃) has also received particular attention [15, 16] because of its wide bandgap. These materials are used in different types of cells. Firstly, they were implemented as sensitizers for titania electrodes in a liquid junction solar cell giving efficiencies of 3.8 % with $\text{CH}_3\text{NH}_3\text{PbI}_3$ and 3.1 % with $\text{CH}_3\text{NH}_3\text{PbBr}_3$ [17]. Then, hybrid perovskites were used in all-solid-state solar cells to achieve higher efficiency. $\text{CH}_3\text{NH}_3\text{PbBr}_3$ exhibits a higher open-circuit voltage (V_{OC}) compared to $\text{CH}_3\text{NH}_3\text{PbI}_3$ because of its wider bandgap. V_{OC} values have risen from 0.96 V in 2009 to 1.3 V by Edri et al. in 2013 [18], up to 1.45 V in 2015 by Sheng et al. [19]. Also in these studies, an increase in the values of J_{sc} and FF has been noticed.

Furthermore, the fabrication techniques are known to influence the quality of the optoelectronic films. Four different deposition methods were reported in different works: one-step spin coating [20], two-step spin coating [21], dual source vapor assisted [22] and vapor assisted solution process [23]. For MAPbBr₃ based solar cells, the highest efficiency of 10.4 % has been reported by Heo et al. [24] using crystallization controlled spin-coating process. The values of short-circuit current density (J_{sc}) and fill-factor (FF) for their study were 8.4 mA/cm^2 and 82 %, respectively. Although higher performances can result by using sophisticated processing, one-step methods are particularly interesting because they can be easily implemented [15, 16]. For instance, Cai et al. realized MAPbBr₃-based solar cells using the one-step spin coating method and the Poly[N-90-heptadecanyl-2,7carbazole-alt-3,6-bis(thiophen-5-yl)-2,5-dioctyl-2,5-dihydropyrrolo[3,4]pyrrole-1,4-dione] (PCBDPP) polymer as HTM [25]. This

photovoltaic device based on MAPbBr₃ displays a high V_{oc} ~1.16 V, FF~0.59 and J_{sc} ~4.47 mA/cm². More efforts should be done to deeply explore the suitability of one-step deposition methods for optoelectronic and photovoltaic applications.

In this paper, solar cells based on MAPbBr₃ with the structure FTO/TiO₂/MAPbBr₃/Spiro-OMeTAD/Au was fabricated. The MAPbBr₃ layer was deposited by one-step spin coating of the precursor solution and the effect of the variation of the MaBr molar ratio in the solution was investigated. Remarkable effect on the morphological, structural, optical and electrical properties was demonstrated. The J_{sc} and FF obtained in this report are in good agreement with those obtained previously for one-step methods [25]. Dual film morphology, with MAPbBr₃ micrometer-size crystals embedded in a small mesoporous TiO₂ background, explains the achieved low V_{oc} values because of the detrimental charge recombination occurring at grain boundaries. It is noticed that full perovskite coverage of TiO₂ background is not necessary for proper operation.

2. Experimental Section.

2.1. Synthesis and device fabrication.

The substrate used was an F-doped SnO₂ (FTO) conducting glass (25*25 mm, 15 Ω/sq resistance). These substrates were patterned with HCl and zinc powder, then they were cleaned in Hallmanex detergent and rinsed with deionized water and ethanol, acetone and isopropanol. A blocking layer of TiO₂ was deposited onto the FTO glass by spray pyrolysis at 450 °C for 35 min, using a solution of titanium diisopropoxide bis(acetylacetonate) (75 % in isopropanol, Sigma Aldrich) in ethanol. The mesoporous TiO₂ layer was deposited on top of the blocking layer by spin coating (2000 rpm) with an acceleration of 2000 rpm, during 10 s) using 100 μL of a solution of a TiO₂ paste (Dyesol 30-NRD, 30 nm average particle size) diluted in ethanol absolute. The obtained films were then dried at 100 °C for 10 min and annealed at 500 °C for 30 min. The MAPbBr₃ solution was prepared by reacting N,N-dimethylformamide solutions (50 wt %) containing PbBr₂, MABr, and Dimethylsulfoxide (DMSO) with various molar ratio of MABr (1:0.5:1, 1:0.75:1, 1:1:1, 1:1.25:1, 1:1.5:1) respectively. The solution was deposited using one step spin-coating at 4000 rpm for 50 s. After deposition, the

substrate was heated up at 65 °C for 1 min, sufficient temperature to remove entirely DMSO, and then annealing at 100 °C for 2 min. To complete the solar devices, the perovskite film was covered with the hole-transporting material (HTM) by dynamically spin coating at 4000 rpm for 30 s, using 50 µl of *spiro*-OMeTAD. The *spiro*-OMeTAD solution was prepared by dissolving; 72.3 mg of (2,2',7,7'-tetrakis(N,N'-di-p-methoxyphenylamine)-9-9'-spirobifluorene), 28,8 µl of 4-tert-butylpyridine, and 17.5 µl of a stock solution of 520 mg/ml of lithium bis-(trifluoromethylsulfonyl)imide in acetonitrile in 1 ml of chlorobenzene. The samples were left overnight in dry air before 100 nm gold contacts were thermally evaporated on top of the device to form the electrode contact (Fig. 1).

2.2. Characterization methods.

The structural properties MAPbBr₃ was studied using X-ray diffraction (XRD) using Bruker AXS-D4Endeavor Advance X-ray diffractometer using Cu K α , wavelength $\lambda=1.5406$ Å, **working with the Bragg-Brentano geometry**. The surface morphologies of MAPbBr₃ films were analyzed by scanning electron microscope using a JEOL JEM-3100 F field emission scanning electron microscope (FEG-SEM) **with 15 kV acceleration voltage**. The optical properties of the perovskite films were obtained by UV-vis spectroscopy using a Varian Cary 300 Bio spectrophotometer and the photoluminescence PL spectra were obtained by using an fluorimeter Horiba FL-1000 **with an excitation wavelength of 430 nm**. The current density-voltage (J-V) curves were recorded under AM 1.5 G (100 mW.cm⁻²) by a solar simulator (ABET Technologies Sun 2000) with a Keithley 2400.

3. Results and discussion.

3.1. Structural properties.

Controlling the ratio of the composite is also a crucial factor to achieve high performance. Given in Fig. (2) are the XRD patterns of MAPbBr₃ prepared with different molar ratio of MABr as mentioned in the experimental section and deposited on FTO/TiO₂ substrates. This figure shows a set of sharp reflections peaks at the following 2θ values: 14.9°, 21.5°, 25.6°, 26.7°, 30.1°, 33.9°, 37.9°, 43.4° and 46°. We defined M as the proportional coefficient of the molar ratio of MABr in PbBr₂: MABr: DMSO. For all samples, strong peak are located at 14.9° and 30.1°, corresponding to the (1 0 0) and (2 0 0) planes, which confirms the formation of a cubic perovskite structure

with a high crystallinity. The intensity of these diffraction peak increase when M increases from 0.5 to 0.75, then it decrease when $M=1$ and it increases again with increasing M to 1.25 and 1.5. The XRD patterns show clearly that the change of molar ratio of MABr effect on the crystallinity of the MAPbBr₃ films. The concentration of the precursor has remarkable effect on the morphology of organic-inorganic halide perovskites, which influences the photovoltaic performance [26-27].

3.2. Morphological properties.

The variation in the XRD result can be an indicative of a change in the morphology of MAPbBr₃ deposited on mesoporous TiO₂ layer. Therefore, we have examined the perovskite with different ratio of MABr sing scanning electron microscope (SEM). Figure (3) shows SEM images of MAPbBr₃ films deposited on m-TiO₂/FTO with different ratio of MABr. A, B, C, D and E images has similar morphology: low magnification SEM micrograph shows highly dense and uniform crystals. However, with higher magnification, the SEM image for all ratio of MABr shows an isolated large crystals which are micrometer-sized cubes in agreement with their cubic symmetry [28]. These large crystals are embedded in a backgorund formed by much small crystals. The increase of MABr content in the precursor of solution promotes the growth of large MAPbBr₃ crystals. The number of large crystallites increase when M increase from 0.5 to 0.75, then it decrease when $M=1$, with the increase of the size of large crystals, and it increases again for $M= 1.25$ and $M=1.5$. These results are similar with that of MAPbI₃ [29] and MAPbBr₃ [30]. The SEM images reveal the formation of a dual morphology including large MAPbBr₃ micrometer-sized crystals on a nano-sized background. The background comprises the TiO₂ mesoporous layer. Although this structure does not assure the whole perovskite coverage, it is beneficial for the performance of devices as the mesoporous TiO₂ avoids electrical shorts.

3.3. Optoelectronic properties.

The change of the molar ratio of MABr has also an effect on the optical properties of the MAPbBr₃ films. To study the variation of optical properties of MAPbBr₃ films, UV-Vis absorption and photoluminescence measurement were studied. The absorbances of MAPbBr₃ films deposited on top of m-TiO₂/FTO were measured by an UV-vis spectroscopy using Varian cary 300 Bio spectroscopy. The Ultraviolet- visible (UV-vis) absorption spectra of the perovskite material deposited on mesoporous TiO₂ with

different ratio of MABr are presented in Fig. 4. The samples shows the typical absorption spectrum of the perovskite $\text{CH}_3\text{NH}_3\text{PbBr}_3$ as previously reported [31- 32]. Increasing the molar ratio of MABr from $M=0.5$ to $M= 1.25$ shows a slight increase in the absorbance then it decrease with $M=1.5$.

The variation of the absorbance intensity is usually consistent with that of the emission wavelength. Therefore, the photoluminescence (PL) emission spectra of $\text{CH}_3\text{NH}_3\text{PbBr}_3$ with different ratio of MABr were measured with excitation wavelength at 430 nm. Figure. 5 shows the PL emission spectra of MAPbBr_3 with different ratio of MABr between 450 nm and 650 nm. This figure displays the relationship between the molar ratio of MABr and the PL intensity of MAPbBr_3 . The photoluminescence peak position for all the substrates of MAPbBr_3 were centred at 540 nm as previously reported [33- 19]. While, the emission intensity change with changing the molar ratio of the MABr in the solution precursor. The MAPbBr_3 films fabricating from ($\text{PbBr}_2\text{:MABr:DMSO}=1\text{:}0.75\text{:}1$) exhibit the strongest PL emission intensity. Then, the intensity decrease with the increase of the ratio of MABr ($M=1$) which is consistent with the reduction of grain in SEM images. The reduction of the number of large crystals induce the decrease of the photoluminescence intensity [30]. Further increasing the MABr concentration produces the decreases of PL intensity of MAPbBr_3 for $M= 1.25$ and 1.5 . As shown in SEM studies of MAPbBr_3 thin films (Fig. 3), when the number of large crystals decrease, the size of crystals increase and the spacing between individual crystals becomes bigger. For $M>1$ the surface shows smaller spacing between crystals. The decrease of spacing between crystals leads to the decrease of PL intensity [30].

3.4. Photovoltaic performance.

Additionally to the materials properties, the composition employed for the preparation of hybrid perovskites can improve the performance of the solar cells. To further understand, current-voltage measurements (Fig. 6) were carried out on $\text{CH}_3\text{NH}_3\text{PbBr}_3$ -based devices with different concentration of MABr under standard air mass 1.5 global (AM 1.5 G) illumination. Table 1 summarizes the photovoltaic parameters J_{sc} , V_{oc} and fill factor (FF).

Interestingly, devices containing the perovskite with the composition (1:1:1) in (PbBr₂: MABr: DMSO) revealed the best photovoltaic performance. This device exhibits a short circuit current density (**J_{sc}**) of 5.16 mA/cm², an open circuit voltage (**V_{oc}**) of 0.888 V and fill factor (FF) of 73.5, resulting in an overall power conversion efficiency (η) of 3.36 % (Figure 6). The increase of the values of V_{oc} , FF and η has been shown to be an indicator of film quality. Also these values for M=1 indicate minimal shunting paths between TiO₂ and spiro-OMeTAD. As can be seen, the efficiency of the device is found to decline slightly for M=0.5 (with efficiency η =3.1 %) and for M=0.75, the efficiency was decreased to 1.8 %. Also, when the concentration of MABr increase for M=1.25 and M=1.5, the efficiency of MABr decrease respectively to 1.4 % and 2.02 %. Remarkably, the higher efficiency, **J_{sc}**, and FF are obtained for M=1. The film morphology for that MABr molar ratio is observed to include the largest crystals. There is therefore a direct relationship between the perovskite crystal size and the device performance. It is also notice that the achieved (V_{oc} ~0.9 V) is below the expected values corresponding to this wide bandgap semiconductor (E_g ~2.3 eV). The small size of the background crystals are responsible for a high amount of grain boundaries in which defects concentrate. High defect density fosters charge recombination reducing, as a consequence, the achivable open-circuit voltage.

5. Conclusion

In conclusion, we report on one-step spin coating method for fabricating methylammonium lead bromide perovskite film for optoelectronic application. Our result show that the change of molar ratio of MABr in the precursor solution has significant effect on the morphological, optical and electrical properties of MAPbBr₃-based devices. The films present large cubic crystals in agreement with their cubic symmetry. The XRD and SEM data show dual crystallinity of the layers with large MAPbBr₃ crystals ontop of mesoporous TiO₂ backgraound minimizing shunting paths, and open-circuit voltage reduction. It is remarked that full perovskite coverage of TiO₂ background is not necessary for proper operation. The best MAPbBr₃ perovskite solar cell achieved a conversion efficiency of 3.46 %, V_{oc} of 0.888 V, J_{sc} of 5.16 mA/cm² and fill factor of 73.5.

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Figure captions:

Fig. 1: Structure of methylammonium lead bromide perovskite solar cell.

Fig. 2: XRD patterns of $\text{CH}_3\text{NH}_3\text{PbBr}_3$ films obtained with different ratio of MABr

Fig. 3: SEM images of MAPbBr_3 films with low and high magnification micrographs for different ratio of MABr; A(M=0.5), B(M=0.75), C(M=1), D(M=1.25), E(M=1.5).

Fig. 4: UV-vis absorption spectra of MAPbBr_3 with different ratio of MABr.

Fig. 5: Photoluminescence spectra of MAPbBr_3 with different ratio of MABr.

Fig. 6: Photovoltaic performance of perovskite devices: Current voltage characteristics of devices with different ratio of MABr.

Table captions:

Table 1: Photovoltaic performance of perovskite solar cells with different ratio of MABr.

PbBr ₂ :MABr:DMSO	Voc (V)	Jsc (mA/cm ²)	FF	Efficiency (%)
(1:0.5:1)	0.861	4.58	71.04	3.10
(1:0.75:1)	0.862	3.06	68.44	1.80
(1:1:1)	0.888	5.16	73.50	3.36
(1:1.25:1)	0.856	2.67	61.47	1.40
(1:1.5:1)	0.935	3.63	56.75	2.02

Table 1

Figure 1

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Fig.1

Figure 2

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Fig.2

Figure 3
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Fig.4

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