

Conference Paper

Effect of Methylammonium Iodide ($\text{CH}_3\text{NH}_3\text{PbI}_3$) Perovskite Concentration on the Performance of Perovskite Solar Cell

AltafYahya AL-she'irey, Akrajas Ali Umar, Muhamad Mat Salleh, and MohdYusriAbd Rahman

Institute Micro Engineering and Nanoelectronics, UniversitiKebangsaan Malaysia, 43600, BangiSelangor, Malaysia

Abstract The effect of Methylammonium Iodide ($\text{CH}_3\text{NH}_3\text{PbI}_3$) perovskites (MIP) concentration on the performance of perovskites sensitized solar cell (PSC) was studied. Three MIP concentrations, namely 0.2, 0.4, and 1.0 M were prepared. In this study PSC with a sandwich structure of ITO/TiO₂/ MIP /electrolyte/ Pt film was fabricated for this purpose. It was found that power conversion efficiency (PCE) increased with the increasing the concentration of MIP, from 0.01 to 0.21% as the concentration increase from 0.2 to 1.0 M. Photoluminescence (PL) study showed that the increase of the MIP concentration decrease therecombination of carrier in the device. Electrochemical impedance spectroscopy (EIS) analysis also shows that with the increased of MIP concentration results in the decreased of the R_{ct} due to the improvement of the carrier transport in the devices.

Keywords: Perovskite Solar Cell, $\text{CH}_3\text{NH}_3\text{PbI}_3$, TiO₂ microtablet

Corresponding Author:
AltafYahya AL-she'irey;
email:
aaltaf_yahyazoo6@yahoo.
com

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1. Introduction

Perovskite material such as ($\text{CH}_3\text{NH}_3\text{PbI}_3$) is one of the most important candidates for high absorber materials in thin film photovoltaic (PV) applications. This material demonstrates many interesting properties including high absorption coefficient, direct bandgap, high stability, and high carrier mobility. Perovskite can be fabricated by mixing ($\text{CH}_3\text{NH}_3\text{I}$) with PbI_2 indimethylformamide. It absorbs light very strongly from visible region to near infrared region. A perovskite solar cell has so far generated power conversion efficiency as high as 15% [1, 2]. Owing to its excellent optical properties, perovskite could be used as sensitizer in the dyesensitized solar cell (DSSC) system. In this work, we study the effect of MIP concentration on the power conversion of the DSSC with structure of ITO/TiO₂/ MIP /electrolyte/ Pt. It was found out that the efficiency increased with increasing of the MIP concentration. The device fabrication and its performance relationship with the concentration will be discussed.

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2. Experiment

2.1. Preparation of TiO_2 microtablet

TiO_2 microtablet on ITO substrate was prepared using a liquid phase deposition method at room temperature by immersing a cleaned ITO substrate into a growth solution that contains 5 mL of 0.5 M ammonium hexafluorotitanate ($(\text{NH}_4)_2 \text{TiF}_6$) purchased from Sigma - Aldrich), and 5 mL of 1.0 M Boric Acid, (H_3BO_3 , RM Chemicals). The substrate was positioned vertically in the solution by hanging the substrate with an adhesive tape. The reaction was kept undisturbed for 4h. After that, the substrate was taken out from the solution and washed with a copious amount of pure water. Then, the sample was annealed at 400 °C for 1 h to obtain an anatase phase.

2.2. Preparation of $\text{CH}_3\text{NH}_3\text{PbI}_3$

The $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite was prepared using the following step. Firstly, 30 mL of hydroiodic acid and 27.8 ml methylamine were mixed together and stirred in ice bath for 2 h. The resulting solution was then evaporated by using a rotatory evaporator operated at 50°C for 2h. The powder of $\text{CH}_3\text{NH}_3\text{I}$ was then obtained using this approach. The powder was washed three times in diethyl ether, and then dried under vacuum for 1 h. To prepare the MIP with different concentration, 0.1 g of $\text{CH}_3\text{NH}_3\text{I}$ was mixed with different concentration of PbI_2 , namely 0.2, 0.4, and 0.1M, in 20 mL dimethylformamide where the reaction was carried out for 15 h at 60 °C.

2.3. PSC device

PSC with structure of ITO / TiO_2 / perovskite / electrolyte / Pt was fabricated. Prior to the reaction, preferably the perovskite was deposited onto the TiO_2 microtablet by immersing the TiO_2 modified ITO substrate into the perovskite solution for 1h at room temperature. By taking the sample out and dried on a hotplate at 50°C for 15 min, thin film of perovskite was successfully grown on TiO_2 microtablet. The PSC device was then prepared by clamping the substrate together with a Pt nanostructure electrode grown on ITO substrate (thickness approximately 150nm). An electrolyte containing iodide/triiodide redox couple was then injected into the space between the platinum counter electrode and the photoanode. The active area of the cell was 0.25cm².

3. Result and Discussion

We have successfully grown the TiO_2 microtablet on the ITO surface using the LPD method (see Fig.1A). From the Fig. 1A it can be seen that the TiO_2 microstructure actually composed of uniquely quasi square shape of a tablet-like structure that grow

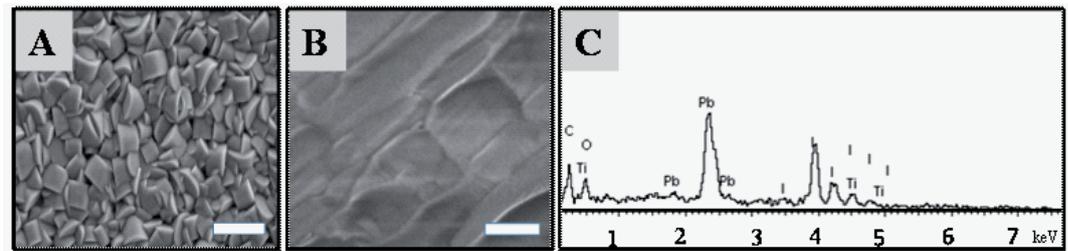


Figure 1: FESEM image of TiO₂ microtablet (A) and TiO₂-covered MIP (B). (C) EDX result of TiO₂ covered MIP. Scales bar is 2 μm in A and B, respectively.

CH ₃ NH ₃ PbI ₃ concentration (M)	η(%)	J _{sc} (mA/cm ²)	V _{oc} (V)	FF	R _s (Ωcm ⁻²)	R _{ct} (Ωcm ⁻²)
0.2	0.01	1.59	0.38	0.01	114	1891
0.4	0.05	1.06	0.54	0.09	104	1820
1.0	0.21	0.3	0.46	1.52	109	1228

TABLE 1: Photovoltaic parameters of CH₃NH₃PbI₃ with three different concentrations as sensitizer for PSC device.

in high density on the substrate surface. These compactly grown microtablets are randomly arranged on the substrate surface without a particular orientation growth. As also can be seen in the Fig.1A the TiO₂ microtablet has covered the ITO surface without the presence of any crack or open surface. The average diameters and thickness of TiO₂ microtablet was found to be ca. 2.78 and ca. 4.35 μm, respectively. The thickness obtained for TiO₂ microtablet in this study is suitable for DSSC application [1,5]. Thus, we expect promising performance can be achieved utilizing TiO₂ microtablet as photovoltaic material for DSSCs application. Fig.1 B shows the FESEM images of the MIP layer grown on the TiO₂ microstructure thin film. As can be seen from image, the MIP layer homogeneously covers the TiO₂ microtablet, leaving no exposed TiO₂ structure. This condition should be useful for DSSC application [9].

The energy dispersive X-ray (EDX) spectrometry has been used to verify the formation of MIP TiO₂ microtablet (Fig.1C). As the Fig.1C reveals, there are three main peaks which are related to the elements exists in the perovskite structure of CH₃CH₂PbI₂ namely C, Pb and I. Other than these three main elements, the peaks represent Ti and O elements were also observed in the EDX spectrum. From the spectrum, we can conclude that the sample prepared is in high quality as no peaks related to the impurities were observed.

TiO₂ microtablet was then used as photoanode in the study of the effect of MIP concentration on the performance of the PSC. Figure 2 shows typical PSC, utilizing three different concentration of MIP response under a simulated AM 1.5G sunlight irradiation. The photovoltaic parameters of PSC are recorded in table 1. As can be seen from Figure 2, the PCE increased with the increasing of MIP concentration. The highest performance was obtained using MIP with concentration of 1M with PCE recorded for this device is 0.21 %. The increase of the performance can be associated with the decreasing of

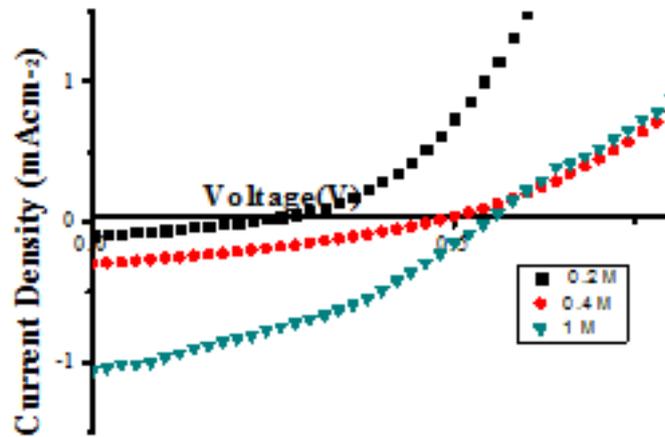


Figure 2: ($J-V$) characteristics of $\text{CH}_3\text{NH}_3\text{PbI}_3$ with different concentrations as sensitizer for PSC device.

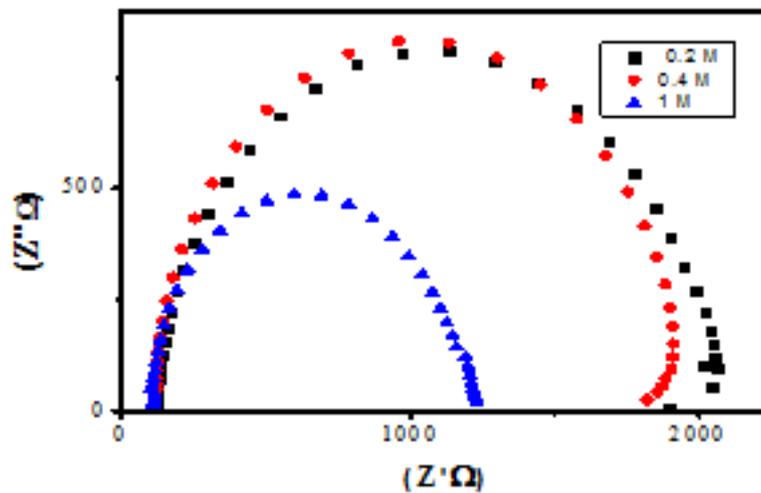


Figure 3: EIS spectra of $\text{CH}_3\text{NH}_3\text{PbI}_3$ with different concentrations as sensitizer for PSC device.

device R_{ct} resistance (discussed later in EIS section). However, the performance of the device dramatically drops when MIP with concentration higher than 1M was used. At this condition, the quality of the thin film is low, causing a high recombination rate and the series resistance [3,6] contributing the decrease in PCE of the device.

EIS measurement was carried out to confirm these phenomena. Figure 3 shows a typical Nyquist plot for impedance spectra of TiO_2 microtablet based PSC device with different concentration of MIP. The frequency use is from 0.1 to 10^6 Hz with alternating current (ac) amplitude of 400 mV. Here, we will denote that the charge transfer resistance (R_{ct}) is attributed to the charge transfer process at the interface between TiO_2 /perovskite layer/ electrolyte. From the spectra we can see that the R_{ct} decreased

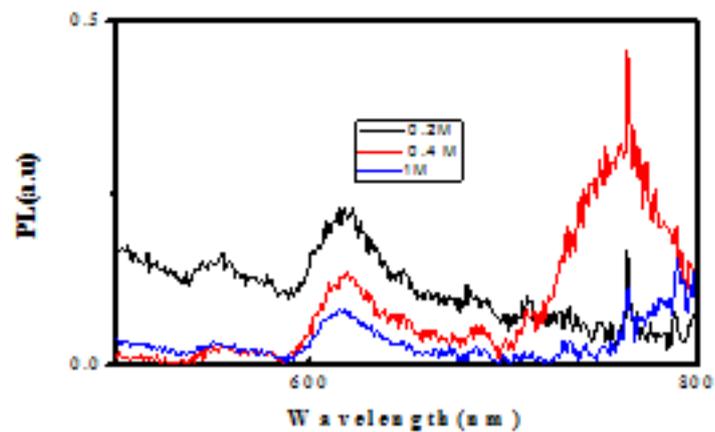


Figure 4: PL spectra of $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite with various MIP concentrations as sensitizer in PSC device.

with the increasing of the concentration of MIP. The low R_{ct} for the device utilizing 1.0 M MIP indicates lower recombination phenomena that will contribute to a higher lifetime of exciton in the device [7].

To further verify the recombination phenomena of MIP as a sensitizer on TiO_2 microtablet PSC device, we carried out the photoluminescence (PL) characterization. The characterization was carried out using an excitation wavelength of 300 nm. The PL spectra of PSC device with different concentration of MIP are shown Fig. 4. PL analysis confirmed that the recombination is decrease with the increase of MIP concentration. This can be seen by the decreasing of the PL intensity upon the increase of MIP concentration, indicated by the decreasing of red emission as concentration of MIP layer increased. The decreasing of the red emission also indicates the MIP layer of higher concentration has a good crystal quality [8]. Besides, PL curves quenching behavior when MIP layer with higher concentration used shows a good charge transport process, leading to a higher performance of PSC device [4, 5].

4. Conclusion

We found that the performance of PSC increases with the increasing of the MIP concentration. EIS and PL analysis shows that by effectively incorporating MIP into PSC results in high exciton life time and lowers the recombination. Thus, improved performance of the PSC device is achieved.

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