CsPbI₃ based Planar Heterojunction Perovskite Solar Cells Using Vacuum Deposition Method

Kyosuke Yonezawa¹, Kohei Yamamoto¹, Md. Shahiduzzaman¹, Yosikazu Furumoto¹, Teresa S. Ripolles², Makoto Karakawa^{1, 3, 4}, Takayuki Kuwabara^{1, 3}, Kohshin Takahashi^{1, 3}, Shuji Hayase², Tetsuya Taima^{1, 3, 4}

¹Graduate School of Natural Science and Technology, Kanazawa Univ., ²Graduate School of Life Science and Systems Engineering, Kyushu Institute of Technology, 2-4 Hibikino, Wakamatsu campus, Kitakyushu, Fukuoka, 808-0135, Japan ³Research Center for Sustainable Energy and Technology (RSET), ⁴Institute for Frontier Science Initiative (InFiniti), Kanazawa Univ., 1C510, Kakuma, Kanazawa, Ishikawa, 920-1192 Japan.

TEL:+81-76-264-6279, e-mail: taima@se.kanazawa-u.ac.jp

Abstract

Cesium iodide (CsI) is attracting attention as a substitute material of organic material such as CH_3NH_3I . In this contribution, we fabricated sequential vacuum deposited planar heterojunction (PHJ) cesium lead iodide (CsPbI₃) perovskite solar cells (PSCs) with enhanced efficiencies by varying annealing time of 0.5, 1, 5, and 10 min. The effect of performance enhancement was investigated as a function of varying annealing time at 350 °C by employing on a hot plate. The best-performing device was obtained by using annealing time of 1 min, delivered photocurrent density of 12.06 mA/cm², voltage of 0.71 V, and fill factor 0.67, leading to a power conversion efficiency (PCE) of 5.71 % at standard AM 1.5G solar illumination.

1. Introduction

Hybrid organometal halide perovskites have been regarded as a potential candidate for thin film photovoltaic because of their excellent cost- and energy-efficient light absorption. Kojima et al. reported hybrid PSCs with the power conversion efficiency (PCE) of 3.8% was fabricated for the first time in 2009 [1]. The encouraging performance discovery has been continued further to obtain a remarkable 22.1% efficiency until the date [2]. There are many researcher used the three-dimensional type perovskite crystal structure named as ABX₃, where A is organic cation, B is inorganic cation, and X is halogen anion. A site can be exchanged with inorganic cation (Cs⁺) instead of organic cations (CH₃NH₃⁺) in order to alter optical and electronic properties. All inorganic CsPbI₃ perovskite has been reported promising highly efficient optical and electrical properties. The PHJ structure have attracted more interest than mesoporous structure in PSCs. This is because the preparation of mesoporous structure film involves high temperature process, which limits their application in simpler architectures. In our previous work, we have reported the PHJ hybrid PSCs using sequential vacuum deposition method by introducing low-temperature compact-TiO_x as an electron collection layer, which enhanced performance of hybrid PSCs [3].

In this study, we apply the sequential vacuum deposition method and investigate the effect of the varying annealing time on the morphology and PCE of the resulting PHJ de-

vices.

2. Experiment

Devices adopted the PHJ characteristics of FTO / compact-TiO₂ / CsPbI₃ / P3HT / Ag. Bare FTO was treated with oxygen plasma for 20 min prior to use. A compact-TiO₂ layer was prepared by chemical bath deposition (CBD) method according to the procedure by Kuwabara et al. [4]. We have prepared compact-TiO₂ film by annealing temperature of 450 °C. The CsPbI₃ perovskite layer was formed by using sequential vacuum deposition of lead iodide (PbI₂) and cesium iodide (CsI), respectively. The thicknesses of the PbI₂ and CsI layers were 113 and 87 nm, respectively. As prepared films were annealed of 0.5, 1, 5, and 10 min at 350 °C and obtained 200 nm-thick CsPbI₃ films. Then, hole transport layer (P3HT) and Ag electrode were deposited in glove box and evaporation chamber without air expose, respectively. Measurement of solar cells characteristics was done in encapsulated container filled in nitrogen gas.

3. Results

The current density *versus* voltage (*J-V*) characteristics at scan speeds of 0.01 V/s is presented in Fig. 1. The parameters of forward scan measurements from the negative bias side to the opposing positive bias side under AM 1.5G simulated solar lights are listed in Table 1. The devices prepared with annealing time of 0.5 min showed a short-circuit current density (J_{sc}) of 9.27 mA/cm². An increase in the Jsc



Fig. 1 *J-V* characteristics of PSCs annealed at 350 $^{\circ}$ C for 0.5, 1, 5 and 10 min.

value to 10.54 mA/cm² was observed for the device fabricated by employing annealing time of 1 min, whereas the J_{sc} value are decreased to 9.25 and 9.14 mA/cm² annealing time of 5, and 10 min, respectively. The PCE also followed a similar trend as the J_{sc} values, viewing a higher PCE of 5.71% for the device fabricated with annealing time of 1 min. With the optimal annealing time of 1 min, we achieved CsPbI₃ perovskite having the most uniform morphology and, consequently, the maximum PCE. In order to ensure the reproducibility of our results, we fabricated on 10 separates devices for 0.5, 1, 5 and 10 min annealing time, respectively. The average solar cells devices performance are summarized in the Table 1.

Table 1 Solar cell parameters calculated from J-V curves.

	Jsc (mA/cm ²)	Voc (V)	FF	PCE (%)
0.5 min	9.27±2.58 (11.30)	0.71±0.05 (0.77)	0.45±0.17 (0.47)	3.14±1.55 (4.10)
1 min	10.54±1.28 (12.06)	0.68±0.05 (0.71)	0.55±0.13 (0.67)	4.02±1.20 (5.71)
5 min	9.25±1.11 (9.78)	0.70±0.04 (0.71)	0.56±0.14 (0.66)	3.75±1.17 (4.58)
10 min	9.14±0.55 (9.84)	0.57±0.26 (0.73)	0.56±0.19 (0.68)	3.43±1.96 (4.85)

Incident photon-to-current conversion efficiency (IPCE) curves of resulting solar cell devices with varying annealing times are showed in Fig. 2. The corresponding CsPbI₃ PSCs can be absorbed from 300 to 750 nm. The enhanced PCE of the devices annealing time of 1 min is consistent with the higher IPCE values of the devices than that of other annealed solar cells. The PHJ CsPbI₃ PSCs exhibited with absorption peak of intensity 50 to 90% at approximately 470 nm.



Fig. 2 IPCE spectrum of PSC annealed at 350 °C for 0.5, 1, 5 and 10 min.



Fig. 3. SEM images of $CsPbI_3$ surface annealed at 350 °C for 0.5, 1, 5 and 10 min.



Fig. 4 Schematic illustration of $CsPbI_3$ layer cross-section fabricated by 350 °C with time dependent.

To investigate the cause of the enhanced efficiency by using sequential vacuum deposition, we observed the morphology of CsPbI₃ perovskite layer by scanning electron microscopic (SEM). The corresponding morphology evaluation of CsPbI₃ films upon varying annealing is showed in Fig. 3. Annealing time plays an important role in the formation of the resulting morphologies. When the annealing time was increased up to 10 min, the resulting morphology exist less number of spherical bright contrast A as compare with others annealing time as shown in Fig. 3. We assume, spherical A can be considered as CsI. Schematic illustration of CsPbI₃ cross-section is shown in Fig. 4. A similar observation for self-induced passivation of PbI2-perovskite was reported by Chen et al [5]. and such concept can fit with our system. We expect that the optimized annealing time is 1 min, when the device coexists of CsI and perovskite in good condition as like as p-i-n structure. Hence, more charges accumulates, leading to enhanced performance of the resulting optimum condition of 1 min.

4. Conclusions

We succeeded to fabricate all inorganic CsPbI₃ PSCs by a sequential vacuum deposition method. The maximum PCE was obtained of 5.71% for the device with annealing time of 1 min.

Acknowledgements

This research work was partially supported by 2015 Basic Research Grant TEPCO Memorial Foundation, 35th Research Grand of Tonen General Sekiyu Foundation, and JSPS KAKENHI Grant 16K05882.

References

- [1] A. Kojima et al., J. Am. Chem. Soc. 131, 6050 (2009)
- [2] NREL Best Research-cell Efficiencies
- [http://www.nrel.gov/ncpv/images/efficiency_chart.jpg]
- [3] K. Yamamoto et al., Jpn. J. Appl. Phys. 54, 08KF02 (2015)
- [4] T. Kuwabara et al., J. Phys. Chem. C, 119, 5274 (2015)
- [5] Q. Chen et al., Nano Lett., 14, 4158 (2014)