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Inverted Planar Perovskite Solar Cells Fabricated by All Vapor Phase Process

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Abstract

A simple vapor-based fabrication process of perovskite solar cells is developed. In this process, PbI_2 film deposited by vacuum evaporation is followed by converting it into $CH_3NH_3PbI_3$ ($MAPbI_3$) perovskite phase by annealing under methylammonium iodide (MAI) vapor. Using the converted $MAPbI_3$ films, we fabricate inverted planar perovskite solar cells by all vapor phase-based process. A power conversion efficiency of 16.1% is achieved by all vapor-based process, the device structure of which is glass/TCO/CuPc/$MAPbI_3$/C60/BCP/Ag.

1. Introduction

Hybrid organic-inorganic perovskite $MAPbI_3$ solar cells have widely been studied by a large number of research groups all over the world since a first report by Miyasaka group [1]. In many studies, solution process was adopted to deposit perovskite films because of the easy fabrication process [2, 3]. On the other hand, vapor-based process has also been reported as an alternative fabrication process of perovskite films [4-10]. The vapor-based process has several advantages compared to the solution methods. It is suitable to fabricate planar heterojunction perovskite solar cells rather than solution process because of precise control of thickness, free from using organic solvent such as DMF and DMSO, easy for scale-up fabrication, etc.

In planar perovskite solar cells, there are two type of heterostructure, n-i-p and p-i-n heterostructure [10]. The latter is called inverted heterostructure. In p-i-n heterostructure, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) is often used as a hole transport layer (HTL) [5]. However, PEDOT:PSS is acidic and a form of aqueous solution when deposited by spin-coating. As a result, PEDOT:PSS layer causes degradation of perovskite solar cells. Therefore, more chemically stable HTL is expected to replace PEDOT:PSS layer. One of the candidates is copper phthalocyanine (CuPc) [11,12]. CuPc is chemically stable and inexpensive. In addition, high purity CuPc (99.95%) is readily available.

In this study, we fabricate p-i-n $MAPbI_3$ perovskite solar cells using CuPc as a hole transport layer by all vapor-based process. We also compare their device performances with those using PEDOT:PSS.

2. Experimental

In our vapor-based process, $MAPbI_3$ perovskite layer as a light absorber is fabricated by a conversion from PbI_2 film by annealing under a methylammonium iodide (MAI) vapor in a tube furnace. The anneal temperature was changed from 120 to 160 °C to make the converted perovskite film high quality. The device architecture we adopt is an inverted planar structure with glass/TCO/HTL/$MAPbI_3$/C60/BCP/Ag, where TCO is transparent conductive oxide. CuPc and PEDOT:PSS are used for HTL. CuPc, C60, BCP and Ag films were deposited by a conventional vacuum evaporation. PEDOT:PSS film was deposited onto TCO coated glass substrate by spin coating. The thickness of each CuPc, PbI_2, C60, and BCP layer is monitored by a quartz microbalance sensor.

The deposited films were evaluated x-ray diffraction (Bruker, D8 Discover) and AFM (Bruker, Innova) measurements. The current density-voltage (*J-V*) characteristics was evaluated by a source meter (Keithley, 2612A) under a standard light, AM1.5G (100 mW/cm^2).

3. Results and Discussion

A typical x-ray diffraction (XRD) pattern of $MAPbI_3$ perovskite film deposited on CuPc are shown in Fig. 1. As can be seen in the figure, the prominent XRD peaks at 2θ=14.1° and 28.4° come from (110) and (220) perovskite planes. The weak XRD peaks from (211) and (310) planes can also be seen. In addition, no XRD peaks from PbI_2 phase can be detected in Fig. 1. This result shows that the complete conversion into perovskite phase from PbI_2 is accomplished by the MAI annealing.

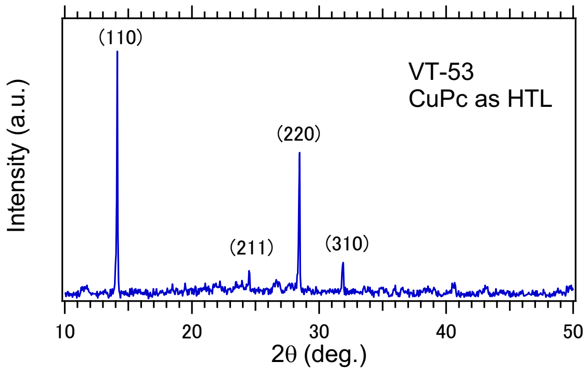

Fig. 1 X-ray diffraction pattern of $MAPbI_3$ perovskite film on CuPc.

Figure 2 shows typical *J-V* curves for p-i-n planar perovskite solar cells using CuPc and PEDOT:PSS as a hole transport layer. The so-called hysteresis associated with scan direction of voltage can be seen in both of the *J-V* curves.

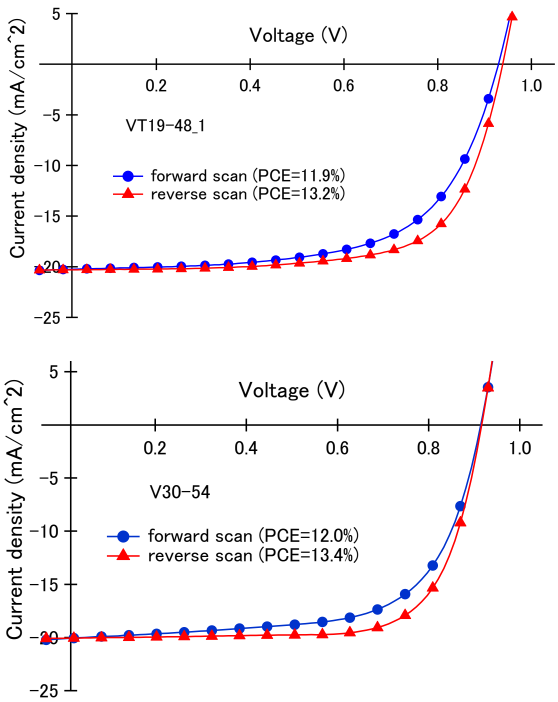

Fig. 2. Typical *J-V* curves of inverted planar $MAPbI_3$ perovskite solar cells using CuPc (upper) and PEDOT:PSS (lower) as a HTL.

The hysteresis has often been observed in planar solar cells fabricated by solution process as well as vapor-based process [9,13]. In some of the solar cells fabricated, nearly hysteresis-free planar perovskite solar cells have been obtained. Figure 3 shows an example of such a hysteresis-free perovskite solar cell, in which CuPc is used as HTL. However, we have still not grasped conditions to fabricate reproducibly hysteresis-free perovskite solar cells. Because, the solar cells are exposed several times in ambient air with humidity during our vapor-based process, and these exposures affect the interfaces in the planar solar cells. Therefore, we believe that a fully vapor-based process without any exposures in ambient air is required to be established for reproducible fabrication of hysteresis-free perovskite solar cells.

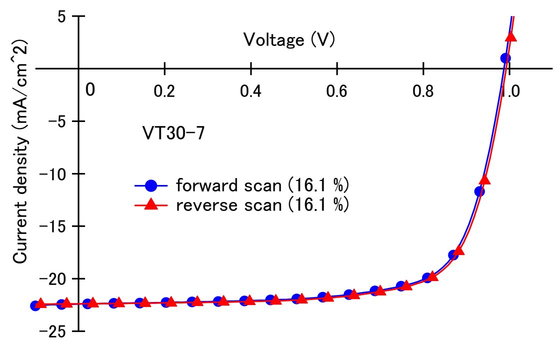

Fig. 3. A best *J-V* curve of inverted planar $MAPbI_3$ perovskite solar cell using CuPc as a HTL.

A best performance of the inverted planar perovskite solar cell with a device structure of glass/TCO/CuPc/$MAPbI_3$/C60/BCP/Ag is shown in Fig. 3. The power conversion efficiencies for forward and reverse scans are 16.1 %. A notably small hysteresis can be seen on the *J-V* curve in Fig. 3. On the other hand, such hysteresis-free solar cells using PEDOT:PSS cannot be obtained. In the best solar cell, short current density J_{SC}, open voltage V_{OC} and fill factor *FF* are 22.4 mA/cm^2, 0.987 V and 0.727, respectively. This high performance was achieved on the solar cell fabricated by a fully vapor-based process and under ambient air exposures with humidity.

3. Conclusions

We developed a simple fabrication process based on vapor phase for inverted planar perovskite solar cells with a device structure of glass/TCO/CuPc/$MAPbI_3$/C60/BCP/Ag. By the all vapor phase process, we succeeded in making planar perovskite solar cell exceeding PCE of 16% with remarkably small hysteresis on *J-V* characteristics.

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