Spin-Coating Perovskite Solar Cells on Plastic

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Abstract

Perovskite solar cells (PSCs) have received great attention in the photovoltaics community due to multiple recent breakthroughs. The goal of this research is to develop a spin-coating process of each PSC layer onto a plastic substrate instead of on the widely used glass substrate. The current method of spin-coating on plastic is the same used for glass. However, multiple issues arise when spinning on plastic due to its material properties, such as non-uniformities found around the substrates' edges, warping of the substrate during annealing, and the uniformity of the electron and hole interface layers. In order to solve these issues an etching process, a Teflon holder, and a slower spin speed for the electron interface layer were added to the original spin-coating process for plastic substrates. To assess the efficacy of this process, several optical and electrical measurements were performed via Olympus profilometer, atomic force microscope, and solar simulator.

I. INTRODUCTION

Photovoltaics have become increasingly prevalent over the past 40 years. Currently, commercial solar cells are made of crystalline silicon. However, the high production cost of these cells has left researchers searching for a new low-cost alternative with power conversion efficiencies (PCEs) matching or exceeding those of silicon solar cells. [2] Perovskites solar cells (PSCs) have gained much attention for demonstrating these qualities. Since 2009, the effectiveness of perovskite solar cells has been under study by members of the photovoltaics community and have exhibited PCEs of 3.8% from the first cell measured to the highest of 22.1% in 2017. [1] Methylammonium lead triiodide ($MAPbI_3$), for example, results in high PCEs and has been a widely researched perovskite solar cell; however, the cell begins to deteriorate at high temperatures. [3] Researchers tried remedying this problem by combining the $MAPbI_3$ solution with formamidinium lead triiodide ($FAPbI_3$), which produced extremely high PCEs of over 20%; however, the problem of deterioration of the cell due to heat stress remained. [3] Researchers later found that by replacing $MAPbI_3$ with $CsPbX_3$, the PSC achieved stability at room temperature and had high PCEs [3]; this discovery led to the solution's popularity amongst the photovoltaics community.

PSCs are made using a plethora of processes, however almost every process uses glass substrates. While glass substrates can withstand high temperatures, it is generally the most expensive part of the PSC and it is fragile. These characteristics mean that glass substrates are not the optimal substrate choice for low-cost manufacturing processes. Researchers have found that plastic substrates are the best choice for those processes, mainly because they are flexible and inexpensive. Although plastic cannot withstand temperatures above 170°C, there are processes for creating PSCs that do not require that high of temperatures. The goal of this research is to develop a spin-coating process for the $CsFAPbI_3$ perovskite onto a plastic substrate instead of on the widely used glass substrate. This perovskite formula is managed at low temperatures and consists of abundant materials, which are essential qualities necessary for low-cost manufacturing processes.

II. EXPERIMENT

A. *Creating $CsFAPbI_3$ and PCBM Solutions*

To form the perovskite solution 52 mg of Cesium Iodide (CsI), 137.6 mg of Formamadinium Iodide (FAI), and 461 mg of Lead Iodide (PBI_2) was mixed with 100 µL of Dimethyl Sulfoxide (DMSO) and 900 µL of Dimethylformamide (DMF) in a clear 8 mL vial. For the PCBM solution, 20 mg/mL of PCBM71 (Phenyl-C_{61}-butyric acid methyl ester) with Chlorobenzene (CBZ) were mixed in an 8 mL amber vial. After adding a stir bar in each solution, each mixture was stirred at room temperature at 400 rpm for 14-18 hours or until solution was homogeneous.

B. *Spin-Coating PEDOT:PSS*

In order to exclude the non-uniformities (missing areas of solutions and pinholes) created when spin-coating each PSC layer, an etching of the ITO layer was performed. After masking the desired ITO section with Kapton tape, the substrates were submerged in 1.0 molar hydrochloric acid for 15 mins. Each substrate was then cleaned by means of sonication for 5 minutes at room temperature using 2% Hellmanex mixed in distilled water, Acetone, and 2-Propanol. They were then placed in an oxygen plasma treatment to increase the wetting of the substrate. After the plasma treatment, the substrate was placed on a Teflon holder, which was then placed in the spin-coater. I designed the holder with an array of vacuum holes matching the substrates size in order to counter any warping of the substrate. Filtered (PVDF 0.45 µm), room temperature PEDOT:PSS (poly[3,4-ethylenedioxythiophene] polystyrene sulfonate) was then deposited on the substrate and immediately spun in a spin-coater at 3000 rpm speed/ 3000 rpm acceleration for 30 seconds after the oxygen plasma treatment. After the spin-coating process was completed, the substrate was immediately annealed on a hotplate at 105°C for 30 min.

C. *Spin-Coating the $CsFAPbI_3$ Perovskite*

In order to prevent contamination from humidity and other pollutants found in ambient air, the remaining spin-coating processes were done in an inert environment. A Teflon holder was used for this spin-coating process due to the warping of the substrate during annealing. A two-stage spin-cycle was used for the spinning of the perovskite solution. During the first cycle, 100 µL of perovskite solution was deposited and spun at 1000 rpm speed and 500 rpm acceleration for 10 seconds. Using a syringe pump, 110 µL of chlorobenzene was dripped 8 sec. into the 20 sec. second stage while the substrate spun at 6000 rpm speed and 3000 rpm acceleration to crystalize the perovskite. The substrate was then placed onto a 100°C hotplate for 10 min. immediately after spin-coating the perovskite.

D. *Spin-Coating the PCBM*

Before spinning the PCBM layer, the PCBM solution was filtered through a 0.45 µm PTFE syringe filter into new amber vial. 70 µL of the filtered solution was then deposited onto the center of the substrate and spun for 45 sec. at 2000 rpm speed and 1000 rpm acceleration for a thin PCBM layer and 1000 rpm speed and 500 rpm acceleration for a thick layer. The substrate was annealed after spinning on a 100°C hotplate for 60 min. After the annealing process is done, thermal evaporate aluminum onto the substrate was done for the aluminum contacts, which were used for measuring of the device.

III. RESULTS

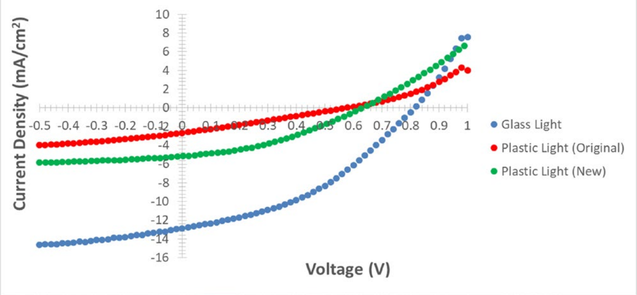

Fig. 1 IV curves of champion cells from glass device, plastic device made with original process, and plastic device made with optimized process. IV curves measured while device was under illumination from AM 1.5 solar simulator.

	Original	New	Glass
Voc (V)	0.573	0.634	0.814
Jsc (mA/cm^2)	-2.65	-5.19	-12.84
FF (%)	26.5	34.8	39.7
PCE (%)	**0.402**	**1.14**	**4.15**

Fig. 2 Comparison of detailed measurements from Fig. 1.

A. *PEDOT:PSS Uniformity*

After further investigation into the loss in efficiency, it was discovered by an atomic force microscope (AFM) that tall spikes found in the ITO layer of plastic substrates were protruding through the PEDOT:PSS layer into the perovskite layer. This results in recombination sites in the solar cell, which leads to a loss of carriers-- i.e. a loss of current, i.e. a loss in overall efficiency of the device cell. In order to cover these spikes, a thicker PEDOT:PSS layer was spun by decreasing the spin speed and acceleration. Spin-coating multiple layers of PEDOT:PSS was also attempted. Neither solution had the desired effect of covering up most of these ITO spikes. The spin-coating of multiple PEDOT:PSS layers failed due to the lack of ink spreading across the substrate. This was due to the hydrophobic nature of dried PSS. In a separate experiment, the stripping of dried PSS before the spin-coating of the next PEDOT:PSS layer was attempted by soaking the dried PEDOT:PSS covered substrate in methanol for 10 minutes. This still did not effectively cover the majority of ITO spikes. Future work includes sanding down the ITO spikes using Texwipes, sandpaper, or a plasma etching process and comparing the amount of spikes found on other brands of ITO covered plastic.

IV. CONCLUSION

In conclusion, this optimized spin-coating process has increased the efficiency of the original process three-fold. The etching of the ITO layer resulted in more accurate measurements of the device cells. After completely vacuuming down the plastic substrates for each spin-coating process I was able to easily deposit my solutions and achieved uniform layers. The optimization of the PCBM layer can be applied in a single batch of devices, where the amount of thin and thick PCBM layered devices would be split. This would allow for a better representation of the devices performance. Unfortunately, devices made on plastic substrates do not reach the high power conversion efficiencies that devices made on glass substrates meet. However, I believe the plastic device efficiency will be able to match or exceed the glass device efficiency once the issue of the uniformity of the PEDOT:PSS layer is solved.

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