

Washing of PEDOT: PSS for Perovskite Solar Cells Performance Optimization

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Abstract

Poly (3,4-ethylenedioxythiophene): poly (styrene sulfonate) (PEDOT:PSS) is one of the most commonly used Hole Transporting Materials (HTMs) in a Perovskite Solar Cell (PSC). However, its low conductivity and local charge transport are issues that restrict the device's performance. Moreover, PEDOT:PSS films have many pinholes and are inhomogeneous, leading to increased charge recombination. Thus, these significant drawbacks require an urgent address to enable PSCs to reach their desired efficiency values. In this paper, we have experimented by incorporating a washing technique using Isopropyl Alcohol (IPA). It was observed that the modified device improved the device efficiency to above 11%. The increase in device efficiency can be attributed to better conductivity, smoother charge transfer, superior film quality and improved material compatibility of different PSC components.

Keywords: Perovskite Solar Cells, Solvent Washing, Solar Cell Materials, Third-Generation Photovoltaics, Solar Cells

1.0 Introduction

Perovskite Solar Cell is a third-generation photovoltaic device which has seen massive improvements in device efficiency in the past decade. Before studying the progress of PSCs, it is essential to understand the various components of the device and how the device functions.

A PSC has three distinct material components: perovskite active layer, charge transport materials and the electrodes. The charge transport materials are again of two types: the hole transport material (HTM) and electron transport material (ETM). PSCs are being heavily researched for the following reasons: tuneable bandgap, which allows the device to absorb from different regions of light, majority of the materials are solution-processable, cheap and abundant raw materials and sourcing, flexible and lighter than conventional silicon-based

photovoltaics^{1,2}. However, in order to commercialize PSCs, there are plenty of issues that need to be tackled. One of the significant issues lies in material compatibility. The parameter determining material compatibility is the energy band alignment and interfacial charge transfer properties.

PEDOT:PSS has been one of the actively used HTMs in PSC applications. The good film-forming ability, high mechanical flexibility, high transmittance to visible light, and excellent stability make this material an ideal candidate for an inverted PSC configuration^{3,4}.

However, a pure film of PEDOT:PSS has a conductivity lower than 1 S cm^{-1} , which does not allow it to be used commercially and on a large scale. Researchers have shown a few methods to improve its performance: doping, treating or compositionally engineering the molecule⁵.

Xia et al., showed that treating PEDOT:PSS with Sulphuric acid or oxalic acid can significantly improve its conductivity from 0.3 to 2400 and 200S cm^{-1} , respectively⁶. Zhao et al.,

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used a method to improve PEDOT:PSS conductivity by treating it with 3-Hydroxy-1-Propane Sulfonic Acid⁷. There have also been methods reported where PEDOT:PSS in acidic solutions can be used to obtain films with high conductivity and more homogeneity. Hamed et al., reported PEDOT:PSS doped with Zinc, consequently improving device fill factor (FF) by reducing trap density and thus, increasing device efficiency⁸. It is important to note that majority of the studies available today focus on either doping PEDOT:PSS or dissolving it in a suitable acid-based solvent. This eventually makes the processing conditions, fabrication process and sourcing of materials very difficult.

In this paper, we have experimented with improving device performance significantly using a straightforward approach of solvent washing. The advantages of this method are plenty: common solvent materials like Isopropyl Alcohol (IPA) are used, the fabrication process remains fundamentally the same, and there is no variation in processing conditions. This work observed that implementing the solvent washing method on the PEDOT:PSS film improves film quality, conductivity, smoother charge transfer and superior material compatibility. These qualities are further reflected through the device performance, wherein the modified devices show greater efficiency than the reference devices.

2.0 Materials

The chemical requirements for this experiment are, Indium-doped Tin Oxide glasses (ITO), PEDOT:PSS (P VP Al 4083), IPA, Lead Iodide (PbI_2), Methylammonium iodide ($\text{CH}_3\text{NH}_3\text{I}$ (MAI)), ([6, 6]-phenyl-C61-butyric acid methyl ester, Sigma 99%), (2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) and Silver (Ag). The perovskite precursor solution was prepared in a 1.4:1.4 molar ratio of MAI and PbI_2 , dissolved in Gamma-Butyrolactone (GBL). The solution was stirred continuously overnight at 65°C, and then filtered using a 0.45 μm PVDF syringe filter before depositing on the substrate. To prepare the ETM solution, PCBM was dissolved in a suitable concentration (20 mg/ml) of Chlorobenzene with two hours of continuous stirring at 45°C. BCP solution was prepared in absolute ethanol with a 0.5 mg/ml concentration.

3.0 Device Fabrication

In this section, we shall delve into how the device is fabricated. It is important to note that the entirety of the device fabrication was done in ambient conditions (Relative humidity 50-60%, Temperature 20°C). Initially, the ITO substrate is cleaned using Toluene, Ethanol, Acetone and Ultrasonic-assisted cleaning with Acetone and IPA. Upon drying the devices, the HTM is spin-coated at 2000 rpm for

50 s, with thermal annealing at 135°C for 10 mins. Before depositing the perovskite layer, IPA is deposited on the substrate to wet the entire surface at the same spin-coating settings. The substrate is thermally annealed at 135°C for 10 mins. The perovskite layer is then deposited and thermally annealed at 100°C for 20 mins. For perovskite deposition, precursor and substrates are kept at 95°C then hot cast at 2000 rpm for 10 seconds and 4000 rpm for 20 seconds. Anti-solvent washing was done by 100 μl toluene after the twenty-eighth second while spinning reached 4000 rpm.

Perovskite growth was completed in ambient air with heat treatment on a hot plate at 100°C for 20 minutes. PCBM is spin-coated at 1500 rpm for 15s and 2000 rpm for 20s. The substrate is then thermally annealed at 90°C for 90s. A buffer layer of Bathocuproine (BCP) is spin-coated at 4000 rpm for 40s. Lastly, the Ag electrodes are deposited using the thermal evaporation method.

4.0 Characterization

The atomic force microscope (AFM) measurements were taken using a Parks Systems XE-100 model device in a non-contact mode. UV-vis spectra were recorded with a PG Instruments T80 spectrophotometer. The current density-voltage curves are measured in a glovebox with a Keithley 2400 model measurement unit. The scan rates are fixed at 0.025 V/s with a 0.03 delay time. These measurements were analyzed under a solar simulator of the intensity of 100 mW/cm² (AM 1.5 G).

5.0 Results and Discussion

The photovoltaic performance of the modified PEDOT:PSS device was calculated along with a reference PEDOT:PSS device. Table 1 shows a tabular representation of the various photovoltaic parameters.

From Table 1 it is very evident that the modified device shows an improved device performance, with an open-circuit voltage (V_{oc})=0.97 V, short circuit current density (J_{sc})=18.1 mA/cm², FF=63 and PCE=11.10%. The reasons for an improved J_{sc} value can be attributed to the high charge carrier lifetime. There is smoother charge transfer observed in the modified devices. Moreover, the improved film quality allows

Table 1: Photovoltaic parameters of the fabricated devices

Samples	V_{oc} (mV)	J_{sc} (mA.cm ⁻²)	FF(%)	PCE(%)
PEDOT:PSS	968	16.2	54	8.37
Modified-PEDOT:PSS	974	18.1	63	11.10

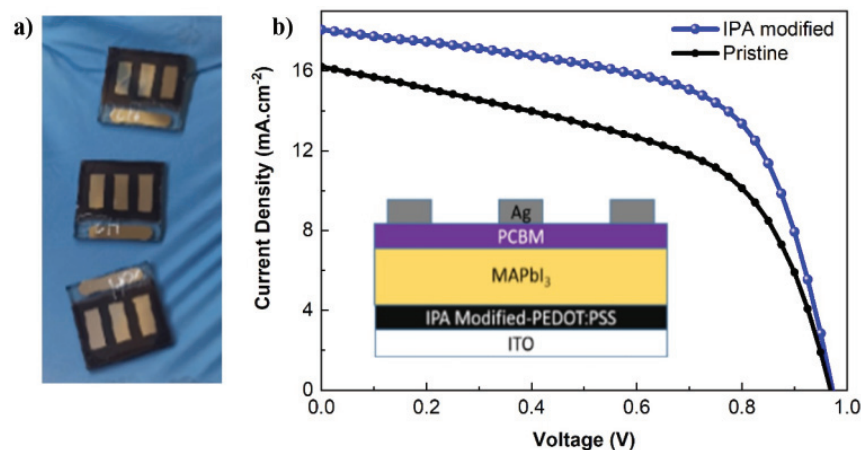


Figure 1(a) Fabricated PSC, (b) J-V curve of the champion PSCs with a schematic representation of device structure

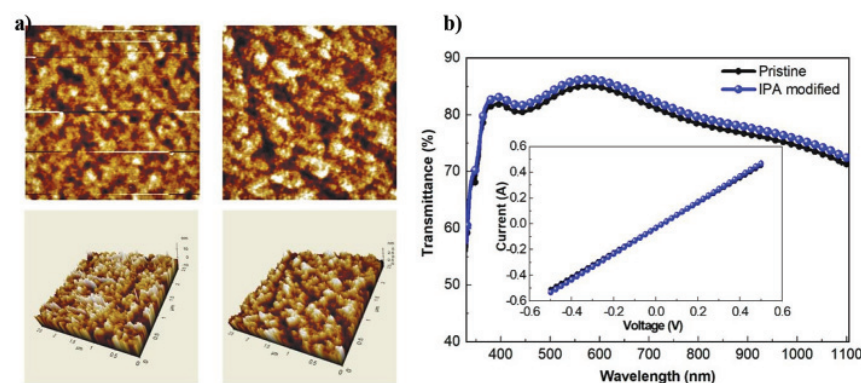


Figure 2(a) AFM images (b) UV-visible transmittance and I-V spectra of the PEDOT:PSS layers

effective charge transfer, especially through interfaces. On treating PEDOT:PSS with IPA, lesser pinholes are observed, which directly reduces the number of recombination sites and trap density states. Figure 1(a) shows a clear representation of the fabricated devices and a schematic diagram depicting the device structure. Figure 1(b) shows the J-V curves for pristine and modified-PEDOT:PSS-based devices. This representation shows that there is a clear increase in J_{sc} and V_{oc} values. The increased V_{oc} values can be attributed to the more suitable material alignment and film thickness due to solvent washing. Figure 1(c) shows the J-V curves for pristine and modified-PEDOT:PSS-based devices.

Figure 2(a) shows a clear representation of the AFM analysis. On observing the modified PEDOT:PSS film closely, the result is highly uniform, has a smoother coverage and forms a stable adhesive with the substrate and subsequent device components. This very advantage even makes this technique highly reproducible. From this observation, we can conclude that equal and uniform surface coverage is essential for consistent and impactful photovoltaic performance.

Moreover, the smoother film surface and reduced surface roughness enable effective charge transfer, especially across interfaces, and reduce charge leakage. The IPA-washing of PEDOT:PSS provides a suitable mechanism for the HTM for charge extraction, transportation and passivation. After the modification, another significant impact has been added is the reduction in interfacial recombination. This can be validated through the improvement in FF values. Typically, this reduction in interfacial recombination will be at the perovskite/PEDOT:PSS interface. Figure 2(b) shows the UV-Vis absorption analysis results of modified and pristine PEDOT:PSS films. From the analysis, it is clear that although the modified film has a similar absorption range to the pristine film, the transmittance is higher. This indicates that there are lesser optical losses in the device and reduced current leakage or auxiliary losses, thus improving device performance. Figure 2(b) also highlights the conductivity results of pristine and modified PEDOT:PSS films. The figure shows that the modified film shows an apparent increase in conductivity. This further facilitates quicker hole charge transport, a visible increase in the J_{sc} value and a notable increase in device performance.

6.0 Conclusion

We have reported a simple IPA-solvent washing technique to entirely modify the performance of PEDOT:PSS and the fabricated device. A reasonable and improved FF, PCE, V_{oc} and J_{sc} value is obtained through this modification. One can conclude that the improvement in photovoltaic performance can be related to better film quality⁹. The higher value for J_{sc} can be attributed to the smoother charge collection, extraction and transfer, which enables the charge carriers to travel effectively, especially across device interfaces. The AFM analysis shows the evened-out surface, resulting in better photovoltaic performance. The IPA-washing technique may also have passivated the traps and defects on the PEDOT:PSS surface, which can be seen through the superior FF value. As a result of all these added advantages, the PCE obtained is 11.1%. Notably, 11.1% PCE is not the highest recorded value for the same materials. If the same device is fabricated in a glovebox or more commercially available materials are used, the efficiency values will be higher.

Moreover, suppose the ITO substrate is replaced with Fluorine-doped Tin Oxide (FTO) glass substrate. In that case, the efficiency values will further increase¹⁰.

7.0 Conflict of Interests

The authors declare no possible conflict of interests.

8.0 References

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