### Printed Interlayers for Organic Solar Cells



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## Abstract

Organic photovoltaics (OPVs) have emerged as a promising technology for renewable energy generation due to their potential for flexible form factors and scalable fabrication. Interlayers play a crucial role in enhancing charge transport within OPVs, as well as other emerging photovoltaic technologies such as perovskites. Emerging photovoltaic research tends to prioritize efficiency over scalability, focusing on spin-coating small area devices. Here, we investigate the transition from spin-coating to a more scalable method printing—of interlayers in OPV devices. We demonstrate that printed interlayers achieve comparable performance to spun interlayers in OPVs, with our best printed-interlayer devices exceeding the power conversion efficiency of spun devices. These results indicate the potential for scalable fabrication techniques in emerging photovoltaic technology without sacrificing performance. Overall, this work advances our understanding of scalable manufacturing processes in renewable energy, paving the way for broader adoption and integration into practical applications.

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## Chapter 1

## Introduction

Organic photovoltaics (OPVs) are a rapidly growing field of research due to the potential of scalable, roll-to-roll fabrication [1, 2]. Recent developments in OPV materials have reached efficiencies have reached 19% for laboratoryscale, spin-coated films [3]. Due to these high efficiencies from new material developments, coupled with scalable fabrication and flexible form factors make this a promising technology to source renewable energy from niches silicon solar cells cannot fill, such as power from indoor lighting [4], or power for conformable medical devices [5].

There tends to be a disconnect in research between the labs chasing efficiency and those chasing scalability and throughput. Printing layers of the solar cell is essential for roll-to-roll compatibility. The aim of this work is to facilitate the transition from spin-coated films to printed films, testing if we, with the same solutions, can achieve printed films without sacrificing performance.

### 1.1 OPV Working Principle

The device stack is depicted in Fig. 1.1. OPVs consist of five layers of materials in order of deposition: 1) the transparent conductive electrode, Indium-Tin-Oxide (ITO), 2) the Electron Transport Layer (ETL), Polyethylinimeneethoxylated (PEIE) 3) the active layer, PCDTBT and PC<sub>70</sub>BM, 4) the Hole Transport Layer (HTL), Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), and 5) the silver top electrode. Light shines through the glass substrate, through the transparent conducting electrode and the ETL, where it gets absorbed in the active layer. The absorption of the light in the active layer generates charges, electrons and holes, which then travel to the electrodes. This work will focus on the interlayers 2) and 4), the electron and hole transport layers.

In order to understand the interlayer function, we must first understand the light to charge conversion process in the active layer. We use on widelystudied workhorse OPV active materials PCDTBT and PC<sub>70</sub>BM [6–8]. These active materials are organic semiconductors, conjugated polymers with  $sp^2 + p_z$  hybridization. With this hybridization, these semiconductors have a highest occupied molecular orbital (HOMO) and a lowest unoccupied molecular orbital (LUMO) which depend on the molecular structure, shown in Fig. 1.2. The donor, PCDTBT, has HOMO and LUMO energy levels closer to vacuum level than the acceptor, PC<sub>70</sub>BM.

In the active layer, the donor and the acceptor are blended together in one film called a bulk heterojunction depicted in Fig. 1.2. When a photon is absorbed by the active layer, it generates an exciton–an electron and hole



Figure 1.1: OPV stack of materials used with pictures of the interlayer solutions.

bound together by the Coulomb force. Organic semiconductors have a strong exciton binding energy [9], so in order to split the charges into electrons and holes, there needs to be a place with enough energy to split the exciton. The heterojunction interface between two different semiconductors, the donor and the acceptor, has a step in energy level that provides the energy to split the charges [9]. Mixing the donor and acceptor into a bulk heterojunction increases the area of interfaces where charges can separate [10]. Once the charges are separated at the interface, they need to be collected at the electrodes.



Figure 1.2: Bulk Heterojunction Structure. Domains of organic donor and acceptor, PCDTBT and  $PC_{70}BM$ , are in blue and red with the HOMO and LUMO energy levels of each shown as well.

### **1.2** Importance of OPV Interlayers

Interlayers are utilized in organic solar cells to facilitate charge transport to the electrodes. They can do this by tuning the energy level at the active-electrode interfaces, as well as improving selectivity of electrons and holes [11]. The role of the electron transport layer is to facilitate the transfer of electrons to the ITO contact. Conversely, the role of the hole transport layer is to facilitate the transfer of facilitate the transfer of holes to the silver contact. The use of interlayers expands beyond organic solar cells. Perovskite solar cells are another emerging solar cell technology, and often use similar interlayer materials [12, 13].

The band diagram of this material system is shown in Figure 1.3 with and without interlayers. In the diagram the active layer bulk-heterojunction of



Figure 1.3: OPV Band diagram with no interlayers and with interlayers. The bright pink demonstrates the impact of the addition of interlayers.

organic molecules PCDTBT and PC<sub>70</sub>BM is depicted as two overlayed boxes in red and blue. The charge carriers and their paths through the device are shown in green. Without interlayers, it is more difficult to draw net current out of a solar cell. The addition of interlayers pushes holes to the silver electrode and electrons to the ITO electrode.

The addition of PEIE as the electron transport layer shifts the work function of ITO. By lowering the work function of ITO such that the energy level is closer to the electron energy level in the active layer, it facilitates electron transport to the ITO electrode. PEIE as an ETL is explained further in Chapter 2.

PEDOT:PSS as the hole transport layer works by blocking electron transport with a high energy barrier. This HTL also adds an intermediate energy step that helps holes move from the active layer to the silver contact. The role of PEDOT:PSS as an HTL is elaborated in Chapter 3.

### **1.3** Comparison of Spin-coating and Printing

For deposition of the interlayers, there are many methods such as spin-coating, slot-die coating, dip-coating, spray-coating, ink-jet printing, and blade-coating. The focus of this work transitioning from spin-coating to blade-coating.

Conventionally, the solution-processed layers are spin-coated, a lab-scale repeatable method to get a thin and uniform films. Spin coating, shown in Fig. 1.4, is a coating method that is achieved by deposition of the solution and controlled rotation of the substrate. Changing the spin speed to modifies the film properties. For this work, spin-coated films are the standard for comparison. Spin coating is a reliable coating process, but it comes at the expense of solution being wasted. To coat the entire substrate, more solution needs to be deposited, and the excess will fly off due to the centrifugal force.

Fig 1.5 depicts blade-coating, or printing.<sup>1</sup> The blade, hundreds of microns from the substrate, drags the ink over the surface. The remaining film is a fraction of the spacing between the blade and the substrate, depending on the adhesion properties of the solution. We then can control the speed of the blade, which controls the resulting film. Optimization of the print speed is an important parameter that changes the properties of printed films.

The pros and cons of spin-coating and printing are outlined in Table 1.1.

<sup>&</sup>lt;sup>1</sup>Though printing can mean a variety of different methods, in this work, we use "printing" as shorthand for "blade-coating," "knife-coating," or "doctor-blading."



Figure 1.4: Spin coating diagram. Solution is deposited in the spin coater, which spins the substrate at a specific rotational speed.



Figure 1.5: Blade coating diagram. Solution is deposited and a blade is dragged over, pulling the solution to coat the substrate.

Spin-coating is used on an industrial-scale in cleanrooms to coat photoresist on Si wafers. However, spin-coating is a challenge roll-to-roll compatibility and large-area substrates. On the other hand, blade-coating is a roll-to-roll (R2R) and large area compatible fabrication method. Another benefit of bladecoating is that it mitigates the issue of wasted solution. We use approximately 10 times less solution per device with blade-coating than spin-coating.

	Spin-Coating	Blade-Coating
R2R Compatibility	No	Yes
Large Area Compatibility	No	Yes
Solution Wasted	High	Low
Film Uniformity	High	Medium
Drying Time	Low	High

Table 1.1: Comparison of coating method properties.

Our previously-optimized fabrication process process is: 1) clean and surface treat ITO-patterned glass substrates, 2) spin coat electron transport layer, 3) spin coat active layer, 4) spin coat hole transport layer, and 5) thermally evaporate the top electrode. We aim to identify and remedy the issues that arise from transitioning from spin-coating to blade-coating interlayers (2) and (4) in the OPV stack.

### 1.4 Outline

The remainder of this report is structured as follows. Chapter 2 describes the electron transport layer, PEIE, and it's printing optimization from films to devices. Chapter 3 provides similar information about the hole transport layer, PEDOT:PSS. Chapter 4 highlights the integration of both printed interlayers, as well as conclusions and implications of this research.

## Chapter 2

## **Electron Transport Layer**

The electron transport layer (ETL) is the first layer deposited on our ITO-glass substrates. In this chapter, we will describe the material, film characterization, and device optimization of the ETL of our inverted OPV.

### **2.1 PEIE**

Polyethylenimine ethoxylated (PEIE) is a polymer used as the electron transport layer in our inverted OPVs. PEIE lowers the work function of ITO, creating an electron-selective contact for the organic semiconductors in the active layer [14]. As seen in Fig. 2.1, the PEIE contains aliphatic amine groups, which are the nitrogen-containing groups with only single bonds. These aliphatic amine groups produce surface dipoles, which decrease the work function of the ITO [15]. The non-conjugated amine groups indicate little charge mobility, so the work function modification comes at the expense of conductivity, so if the PEIE layer is too thick, it can limit charge transport across the ETL, limiting current out of the device [16]. We chose PEIE because of its work function lowering ability as well as its solution processability and low-temperature processing [17]. However, because we want to find the optimal tradeoff between the work function modification and the insulating nature of PEIE, we will focus on characterizing the thickness of ETL layer.



Figure 2.1: PEIE molecular structure. The x, y, and z sections of the polymer repeat in no particular order to form the structure. The x structure contains aliphatic amine groups.

Because of PEIE's non-conductive nature, film thickness is important as it must be thin enough to allow charge to tunnel through the film [18]. The solution of PEIE was diluted to 0.048% wt. in ethanol, so the solution was very dilute, and had very similar solution properties as pure ethanol. The PEIE solution was optimized for spin-coating [4], and had such low wt.% PEIE because the ideal PEIE film is very thin. In this chapter, we demonstrate similar film and device performance between spun and printed PEIE ETLs.

### 2.2 PEIE Film Characterization

We investigated printed PEIE films compared to spun PEIE films. To understand the tradeoffs in switching from spin-coating to blade coating, we must characterize and understand the changes in PEIE films.

#### 2.2.1 PEIE Thickness

Because printing parameters influence the resulting thickness of a film, we investigate the PEIE print speeds impact on film thickness (Fig. 2.2). In general, because the solution is so dilute, we did not see an appreciable dependence of print speed on thickness outside of margin of error. All blade heights for printed PEIE were  $75\mu$ m above the surface. The spun PEIE control was spun at 5000 rpm.

We measure the film thickness with spectroscopic reflectometry, on a Filmetrics F20 setup, which interpolates film thickness utilizing the index of refraction. We use reflectometry because our films are very thin, on the order of nanometers, but one drawback of this tactic is inability to measure on transparent substrates, so the printing of these PEIE films is on silicon substrates instead of ITO-glass substrates.

For blade coated films, the thickness of the PEIE film varied greatly across the substrate, much more than the spun film which was relatively constant. However, the spread of thickness measurements varied, indicating variations in quality of film. An important note is that while blade-coating is compatible with large scale printing and is a scalable method of fabricating thin films,



Figure 2.2: PEIE Print Speed and Measured Thickness. Note how the spun film has much smaller variation. However, all of the print speeds are within margin of error of the spun film.

one downside is that on small substrates, the ink can spread back and pool around the edges of the substrates. This means there will be a larger variation of thickness on smaller substrates, but this will be mitigated at larger scales when the ink doesn't flood the substrate.

#### 2.2.2 PEIE Work Function

To investigate the impact of printing parameters on work function modification, we measured the work function of ITO-glass substrates with spun and printed PEIE layers. Recall that the function of the electron transport layer is to lower the work function to align closer to the LUMO energy level of the organic acceptor, around 4eV for  $PC_{70}BM$  [19], facilitating electron transport to the ITO electrode.

We measure the work function with a kelvin probe from KP Technologies. Utilizing Kelvin Probe Force Microscopy (KPFM), the kelvin probe measures changes in work function by changes in the contact potential difference as the probe tip oscillates. This apparatus inherently measures relative values, but is calibrated against a gold reference sample to get absolute units.

In Fig. 2.3, we can see the impact of PEIE on the work function of ITO. Bare ITO has a work function around 4.8 eV, and spun PEIE and the lower print speeds of PEIE decreased the work function by 0.6-0.8 eV. The higher print speed of 24 mm/s showed little modification, indicating that the PEIE film quality is not ideal. 18 mm/s was the closest speed that resulted in a work function closest to spun PEIE.



Figure 2.3: Impact of printing parameters on ITO Work Function. PEIE layers lowered the work function of bare ITO, but print speeds modified to varying degrees.

### 2.3 **PEIE Device Optimization**

To fully understand the tradeoffs between the thickness, work function modification, and the inherent insulating nature of PEIE, we must make complete devices to see the impact on efficiency. We fabricated devices with printed PEIE layers while the rest of the device layers are conventionally fabricated with spin-coating and evaporation. In doing so, we were able to optimize the PEIE layer print parameters to compare device performance.

#### 2.3.1 PEIE External Quantum Efficiency



Figure 2.4: Printed PEIE External Quantum Efficiency. Spun PEIE had the highest EQE across wavelengths, followed closely by print speeds of 18mm/s and 24 mm/s, with 12 mm/s was the lowest. Each trace represents an average of n measurements.

In Fig. 2.4, we look at External Quantum Efficiency (EQE) across devices

with varied PEIE print speed. EQE is a measurement of the percentage of electrons that are measured as current from the device over the number of incident photons at a given energy that are shined onto the device. We see a similar shape among spun and printed PEIE films, but the printed PEIE films were slightly lower, with 12 mm/s print speed the lowest. This indicates that for a given visible photon, we are losing the analogous current measured from the device. It could be due to the PEIE layers being too thick to tunnel through in certain regions due to the non-uniformity. Or, it could be that the work function modification of 12 mm/s was too far, and no longer facilitates, but blocks some electrons from passing through to the ITO electrode.

#### 2.3.2 PEIE Current-Voltage Characteristics

To analyze the current-voltage characteristics of spun and printed PEIE OPVs, we ran a sweep measuring the current across voltages for an OPV in the dark and under simulated sunlight. The measurements were made with an Ossila Solar Cell I-V Test System, with the AM1.5G light provided using an Oriel Sol1A solar simulator.

Looking at the JV data in Figure 2.5, we can see that we achieved similar efficiencies with printed 18mm/s to spun PEIE. Looking at the dark current, the spun devices have the lowest reverse-bias dark current, which can be explained by the consistent PEIE film quality and thickness of spun films. The dark current determines the base current that the photocurrent must overcome; more consistent films could decrease the dark current further. For the light current, the spun device has the higher open-circuit voltage, but some of



Figure 2.5: Printed PEIE Current-Voltage Characteristics. Each trace highlights the best performing pixel, with PCE's showing comparable performance from printed PEIE to spun, especially for 18mm/s.

the printed films have a higher magnitude of short-circuit current. If the work function is not matched ideally to the active layer, the open circuit voltage  $V_{oc}$  will be impacted. More optimization of consistency of printed PEIE films could further improve performance with attaining the same  $V_{oc}$  and minimizing dark current.

The performance across multiple devices is highlighted in Table 2.1, which shows key performance parameters of short-circuit current,  $J_{sc}$ , open circuit voltage,  $V_{oc}$ , fill-factor, FF, power conversion efficiency, PCE, and series resistance  $R_s$ . This information highlights the variation of device-to-device performance, and demonstrates the consistency of films.

The results show that 12mm/s is too slow a speed for printing PEIE to achieve ideal OPV performance. We see that though the  $V_{oc}$  is closest to the

	$J_{sc}(mA/cm^2)$	$V_{oc}(V)$	FF(%)	PCE(%)	$R_s(\Omega \cdot cm^2)$
Spun	$8.2 \pm 0.3$	$0.80 \pm 0.02$	$44 \pm 2$	$2.9 \pm 0.2$	$32 \pm 7$
Print: 12 mm/s	$6.6 \pm 1.1$	$0.69 \pm 0.11$	$37 \pm 5$	$1.7 \pm 0.5$	$60 \pm 29$
Print: 18 mm/s	$8.5\pm0.2$	$0.62 \pm 0.11$	$40 \pm 3$	$2.4 \pm 0.5$	$34 \pm 7$
Print: 24 mm/s	$8.0 \pm 0.6$	$0.68 \pm 0.05$	$42 \pm 3$	$2.3 \pm 0.4$	$40 \pm 9$

Table 2.1: Performance parameters of spun and printed PEIE devices.

spun control, the  $J_{sc}$  falls short, as well as fill factor and series resistance, resulting in a lower power conversion efficiency.

For OPVs with printed PEIE at 18mm/s, we achieved comparable performance to spun PEIE. The  $J_{sc}$  of 18mm/s and 24mm/s devices were at or above the  $J_{sc}$  of spun devices, but the  $V_{oc}$  of printed devices were lower. This tradeoff of  $J_{sc}$  and  $V_{oc}$  from spun to printed films can be further tuned, and more study of PEIE printed film consistency can help with device-to-device reproducibility. These results demonstrate printed electron transport layers in OPVs without sacrificing performance.

## Chapter 3

## Hole Transport Layer

The Hole Transport Layer (HTL) is deposited on top of the active layer. The role of the HTL is to facilitate hole transfer to the silver electrode. In this chapter, we will describe the material, characterization, and printing optimization of the HTL of the OPV.

### 3.1 PEDOT:PSS

Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate), or PEDOT:PSS, is a conductive polymer blend shown in Figure 3.1. PEDOT:PSS, is used ubiquitously throughout printed electronics, and is one of the most popular hole transport layer materials [2, 3, 11, 14, 20–23]. PEDOT is a conjugated polymer, with alternating single and double bonds leading to  $sp_2$  hybridization and a conductive extended pi system. PSS is a negatively charged counterion to balance the oxidized PEDOT in an aqueous dispersion. When deposited, it forms a conductive film held together with the pi-pi stacking of the PEDOT and the hydrogen bonding between the PSS groups [20]. PEDOT:PSS is ideal as an HTL because it's energy bands block electrons from the active layer to the electrode, and facilitates hole transport.



Figure 3.1: PEDOT:PSS molecular structure, with PEDOT on the left and PSS on the right. PEDOT is a conjugated polymer, and PSS is the charged counterion.

In order to be able to deposit films, either with spinning or printing, the solution must wet to the surface underneath. PEDOT:PSS typically is sold as an aqueous solution, like AI4083 sold by Clevios<sup>TM</sup>. HTL Solar PEDOT:PSS is another commercial formulation sold by the same company, optimized with surfactants for wettability to the active layer [24]. Figure 3.2 shows the contact angles between the PEDOT:PSS formulations on the active layer material, showing that the HTL formulation wets better to the active layer surface.

We use the PEDOT:PSS HTL formulation inverted OPV devices. Inverted solar architectures, wherein the HTL is deposited on top of the active layer, improve the lifetime by preventing degradation of the ITO reacting with acidic PEDOT:PSS. [11] Though there has been previous work with printed PE-DOT:PSS, researchers use their own formulation. [21, 23] To our knowledge,



Contact Angle: 100°

Contact Angle: 15°

Figure 3.2: PEDOT:PSS contact angle comparison of AI4083 and HTL formulations on active layer film. The HTL formulation has a much lower contact angle and better wettability to the active layer.

the HTL Solar commercial formulation has not been fully characterized and for use in printed OPVs.

The HTL Solar formulation of PEDOT:PSS is a 1:2.5 w/w blend of PE-DOT:PSS with a weight concentration of 1.0-1.3% in water. [24] The HTL Solar formulation includes surfactants to increase the wettability to the active layer. More technical properties are included in Table 3.1. In this chapter, we demonstrate comparable film and device performance between spun and printed PEDOT:PSS HTLs with a commercial PEDOT:PSS formulation.

Conductivity (S/cm)	Resistivity ( $\Omega \cdot cm$ )	Viscosity (mPa·s)	рН
0.1 - 1.0	1 - 10	8 - 30	2

Table 3.1: PEDOT: PSS HTL Technical Data Specifications from [24]

### 3.2 PEDOT:PSS Film Characterization

We investigated printed PEDOT:PSS films compared to spun PEDOT:PSS films, characterizing microscopy, thickness, and sheet resistance. All blade heights for printed PEDOT:PSS were  $100\mu$ m above the surface. After deposition with spin-coating or printing, the films were annealed at 70°C for 10 minutes.

#### 3.2.1 PEDOT:PSS Microscopy

We wanted to investigate if the deposition method would have an impact on the quality of the film, which we analyzed at two different length scales using both optical and atomic force microscopy. The spun PEDOT:PSS film was spun at 4500 rpm and the printed film was printed at 18 mm/s.



Figure 3.3: Optical Microscope Images of Spun and Printed PEDOT:PSS Films. Scale bar is  $100\mu$ m. Note the small dots on both films.

Figure 3.3 shows optical microscope images of spun and printed PEDOT:PSS. In the images, we can see dots on both films, but the dots are larger on the printed film. The measurements were conducted on a Nikon microscope, with 20x magnification.

In order to get a closer look at the film topography, we look at a spun and printed PEDOT:PSS films with an atomic force microscope (AFM), shown in Fig. 3.4. Both had topographical deviations around 10 nm, and have very similar profiles. Measurements were taken with Bruker AFM in tapping mode, with TESPA-V2 tip.



Figure 3.4: Atomic Force Microscope Images of Spun and Printed PEDOT:PSS Films. On this length scale, the topography of the films look similar whether they are spun or printed.

Looking closely at the films, they appear very similar, with only small differences in the spun and printed films. To understand the relevance of these differences, we want to investigate the electrical properties to see if these morphological differences impact device performance.

#### 3.2.2 PEDOT:PSS Sheet Resistance

The function of PEDOT:PSS as the hole transport layer relies on its ability to conduct charge. After holes are generated in the active layer, they need to travel through the PEDOT:PSS layer before arriving at the silver electrode. Thus, understanding the conductivity of the PEDOT:PSS layer is imperative to understanding the impact on device performance.



Figure 3.5: Sheet Resistance of Spun and Printed PEDOT:PSS Films. Note how slow print speeds result in much higher sheet resistance than higher speeds, with 12 mm/s as the lowest sheet resistance, similar to spun films.

We measured sheet resistance of spun and printed PEDOT:PSS films, shown in Figure 3.5. Measurements were conducted with a Keithley 2400 sourcemeter with the four point probe method. Our ideal HTL has low sheet resistance, and is highly conductive. We can see that for the low print speeds, we see high values for sheet resistance. This could be due to the film quality, or an issue with contacting to the film with the probes. Meanwhile, for print speeds 12 mm/s to 24 mm/s, we have much lower sheet resistance values, with 12 mm/s being the lowest. Since sheet resistance is a measurement that depends on the thickness of the film, and perhaps the contact issue may be due to the film being too thin, we investigated the thickness of the films to fully understand the film properties.

#### 3.2.3 PEDOT:PSS Thickness

In literature, the transition from spinning to blade-coating typically aims for print speeds that match similar thickness of spun films. [21, 25] If there are any other film properties that change from spinning to printing, the switch to the scalable process may impact device performance. We aim to find the optimal printed film, not necessarily match the film thickness. Regardless, characterizing the film thickness is imperative to understanding the impact of this layer on performance.

Since the thickness of PEDOT:PSS films varies based on the print speed, we measured the film thickness as a function of print speed and compared to spun films (Fig. 3.6). We measured PEDOT:PSS thickness with a Dektak profilometer. To do this, we scratched the PEDOT:PSS films, and measured across the the scratch to analyze the step height.

For the ink properties of PEDOT:PSS HTL, as the print speed increases, the thickness of the film increases. As the speed increases, there is less time for the solution to form a meniscus, resulting in a thicker film. The 6 mm/s film was of a similar thickness to spun films, but also had a high sheet resistance measurement.



Figure 3.6: PEDOT:PSS Print Speed and Measured Thickness and Blade-Coating Ink Diagram. This shows that as the print speed increases, the thickness increases.

#### 3.2.4 PEDOT:PSS Resistivity

To get a normalized measurement of PEDOT:PSS HTLs conductivity, we calculated the resistivity from the measured values in previous sections, shown in Figure 3.7. Resistivity was calculated by multiplying thickness and sheet resistance measurements.

The spun film and the 12 mm/s printed film had resistivity values within error bounds of each other, around 40  $\Omega$ ·cm. The other printed films had higher resistivity, with the slow speeds because of the high sheet resistance, and the high speeds because of the thickness.

We take the slower print speed values with a grain of salt, since the films are much thinner, if there was an issue getting contact to the film, it would impact the sheet resistance measurement. However, these higher values could indicate a difference of film quality or uniformity, which would impact the



Figure 3.7: PEDOT:PSS Resistivity for Spun and Printed Films. Printed PEDOT:PSS at 12 mm/s had the lowest resistivity.

device performance.

These resistivity values are lower than the specified value from the ink supplier was 1 - 10  $\Omega$ ·cm. This can be explained because the resistivity for PEDOT:PSS HTL from the datasheet is specified for an annealing temperature of 120°C. For our fabrication of inverted solar cells, any heating to the PEDOT:PSS HTL layer also heats up all of the layers underneath it, and for our materials, 70°C is not damaging to the active layer morphology.

Understanding the interplay with print speed and film properties such as sheet resistance, thickness, and resistivity gives us some insight on the PE-DOT:PSS HTL films, but we also want to see how these film properties translate into device performance.

### 3.3 **PEDOT:PSS** Device Optimization

We fabricated devices with printed PEDOT:PSS HTL layers while the rest of the device layers are conventionally fabricated with spin-coating and evaporation. For devices with printed PEDOT:PSS HTLs, we measured external quantum efficiency and current-voltage characteristics. We achieved similar device performance with printed PEDOT:PSS HTLs as with the spun control. For our best performing devices, we achieved higher efficiencies with a printed PEDOT:PSS HTL than spun.

#### 3.3.1 PEDOT:PSS External Quantum Efficiency



Figure 3.8: Printed PEDOT:PSS External Quantum Efficiency. Each trace represents an average of n measurements. Note how as print speed increases, and the PEDOT:PSS film gets thicker, the EQE decreases.

In Fig. 3.8, we look at External Quantum Efficiency (EQE) across devices with spun and varying print speeds of PEDOT:PSS HTLs. As the print speed increases, the PEDOT:PSS film gets thicker, and there is more resistance, and more distance for holes to travel through the HTL before being measured at the contact. This means that there is a higher chance of recombination, and less current out of the device per photon. The printed 3 mm/s film had the highest EQE, even higher than spun PEDOT:PSS devices.

The EQE measurement is performed on an unbiased solar cell, and when integrated over the wavelength, will be proportional to the short circuit current,  $J_{sc}$ . While this is an important value, it is not the only important factor in determining power conversion efficiency.

### 3.3.2 PEDOT:PSS Current-Voltage Characteristics

To find the power conversion efficiency, we measured the current-voltage characteristics of OPVs made with spun and printed PEDOT:PSS HTLs, shown in Figure 3.9. We measured the device in the dark and under simulated sunlight.

This highlights an interesting tradeoff as print speed increases, and PE-DOT:PSS films get thicker, our  $J_{sc}$  values decrease, but our  $V_{oc}$  increases. Since power conversion efficiency depends on both  $J_{sc}$  and  $V_{oc}$ , the ideal performance peak is at the inflection point of both. The  $V_{oc}$  depends on a variety of factors including the work function of the electrodes, depends on achieving an ohmic contact to the active layer through the PEDOT:PSS. [26] If there are deviations in the PEDOT:PSS film quality or coverage when printed on the active layer, it may impact the  $V_{oc}$ . Perhaps the inability to get sheet resistance measurements on the thin, slow-printed PEDOT:PSS films is indicative of a failure of film coverage or uniformity. Either way, the PEDOT:PSS HTL



Figure 3.9: Printed PEDOT:PSS Current-Voltage Characteristics. Each trace highlights the best performing pixel, with PCE's of slower printed PE-DOT:PSS films showing better performance than the spun devices.

print speed for optimal power conversion efficiency is 12 mm/s.

We found similar trends and tradeoffs across multiple OPVs, shown in in Table 3.2, which shows performance parameters of short-circuit current,  $J_{sc}$ , open circuit voltage,  $V_{oc}$ , fill-factor, FF, power conversion efficiency, PCE, and series resistance  $R_s$ . This information highlights the variation of deviceto-device performance, and demonstrates the consistency of films. The  $J_{sc}$ decreases as the PEDOT:PSS film gets thicker. The  $V_{oc}$  appears to increase as the PEDOT:PSS film gets thicker, but still within the margin of error. The spread of  $V_{oc}$  decreases as the print speed increases, indicating higher repeatability. Our FF is better with 12 mm/s and higher print speeds.

The PCE of these devices show that printed PEDOT:PSS films could match performance, or even outperform the spun films. The optimal PE-

	$J_{sc}(mA/cm^2)$	$V_{oc}(V)$	FF(%)	PCE(%)	$R_s(\Omega \cdot cm^2)$
Spun	$8.4 \pm 0.3$	$0.67 \pm 0.11$	$43 \pm 4$	$2.4 \pm 0.6$	$37 \pm 7$
Print: 3 mm/s	$9.3 \pm 0.6$	$0.62 \pm 0.11$	$43 \pm 5$	$2.5\pm0.8$	$29 \pm 7$
Print: 6 mm/s	$8.8 \pm 0.2$	$0.70 \pm 0.10$	$43 \pm 4$	$2.7 \pm 0.6$	$37 \pm 10$
Print: 9 mm/s	$8.1 \pm 0.5$	$0.70 \pm 0.09$	$42 \pm 4$	$2.4\pm0.5$	$38 \pm 11$
Print: 12 mm/s	$7.0 \pm 0.5$	$0.8 \pm 0.04$	$49 \pm 3$	$2.8 \pm 0.5$	$32 \pm 8$
Print: 18 mm/s	$5.4 \pm 0.5$	$0.79 \pm 0.06$	$50 \pm 2$	$2.2 \pm 0.4$	$37 \pm 7$
Print: 24 mm/s	$5.2 \pm 0.4$	$0.80 \pm 0.02$	$51 \pm 2$	$2.1\pm0.2$	$35 \pm 5$

Table 3.2: Performance parameters of spun and printed PEDOT:PSS devices.

DOT:PSS HTL print speed was 12 mm/s, which yielded the lowest resistivity film; this speed achieved a PCE of 3.74%. The thickness-matched PE-DOT:PSS film, 6mm/s, and the highest EQE print speed, 3 mm/s both also performed very well.

## Chapter 4

## Conclusion

After analyzing and optimizing both the hole and electron transport layers individually, we were able to achieve comparable performance from spun to printed devices. This chapter shows the results of devices with both printed interlayers, achieving comparable performance, with our best printed devices exceeding spun performance. We also discuss the implications of these results and future work.

### 4.1 Integration of Printed Interlayers

We combined the two best performing print parameters into a solar cell with both interlayers printed. For a PEIE ETL, the best performing devices had a print speed of 18 mm/s, and for PEDOT:PSS HTL the best performing devices had a print speed of 12 mm/s.

We measured the external quantum efficiency of the devices with printed interlayers (Fig. 4.1), and we achieved comparable EQE between devices with spun and printed interlayers. There is a discrepancy between Chapter 2 and Chapter 3's spun control performance because of a process change in the evaporation of the top electrode due to equipment availability, and we wanted to ensure apples-to-apples comparisons. However, we compare printed-interlayers performance to the best-performing spun devices, and we see that the devices with printed interlayers match performance to the spun devices.



Figure 4.1: Printed Interlayers External Quantum Efficiency. Each trace represents an average of n measurements. The printed and spun show similar shape and EQE values.

We also analyzed the current-voltage characteristics of spun and printed interlayer OPVs, shown in Figure 4.2. The printed interlayer device has a similar short circuit current and open circuit voltage, but the fill factor is better than our best spun device, resulting in a higher power conversion efficiency. The best printed-interlayer device where our best PCE of 3.65%, surpassing the best spun device performance of 3.31%.



Figure 4.2: Current-voltage characteristics of devices with spun and printed interlayers. Each trace shows the best performing OPVs with spun and printed interlayers, with the power conversion efficiency of printed interlayer devices exceeding the spun devices.

We also analyzed the OPV performance variability from spun and printed interlayer devices in Table 4.1. The performance parameters of short-circuit current,  $J_{sc}$ , open circuit voltage,  $V_{oc}$ , fill-factor, FF, power conversion efficiency, PCE, and series resistance  $R_s$ , were all within margin of error of each other from spun to printed devices.

	$J_{sc}(mA/cm^2)$	$V_{oc}(V)$	FF(%)	PCE(%)	$R_s(\Omega \cdot cm^2)$
Spun	$8.2 \pm 0.3$	$0.80 \pm 0.02$	$44 \pm 2$	$2.9 \pm 0.2$	$32 \pm 7$
Printed	$8.1 \pm 0.5$	$0.71 \pm 0.14$	$47 \pm 6$	$2.9\pm0.5$	$29 \pm 7$

Table 4.1: Performance parameters of spun and printed interlayer devices.

This shows that we successfully transitioned from spinning to printing both interlayers of the OPV without sacrificing performance.

### 4.2 Implications and Future Work

In this work, we were able to show a successful transition from spin-coating to blade-coating of interlayers of OPVs. Since blade-coating is a technique much more scalable to large area devices and roll-to-roll manufacturing, as well as wastes much less solution, this work opens the door for scaling OPV fabrication. With the same inks and substrates, our work highlights a potential simple transition towards scalable processing of OPVs, that could be implemented at a laboratory scale where researchers typically process with spin-coating. Additionally, since these transport layer materials are utilized in multiple active material systems, such as perovskites, this work is generalizable to other emerging photovoltaic technologies.

Our future research directions include: 1) achieving fully-printed OPVs 2) scaling to larger area devices.

To achieve fully printed OPVs, we also need to print the active layer and the silver electrode. This will require optimization of printing mechanisms and processing of films, similar to this work. However, printing the active layer is no small feat. Since it is a bulk heterojunction blend, the morphology of this layer depends greatly on the drying dynamics of the solvent, which vary greatly from spin-coating to printing, impacting the performance of the device. [27] Our preliminary optimization experiments with the active layer did not match performance with spun devices with the same ink formulations, as seen in Figure 4.3. Differences in film morphology from spun to printed active layers result in lower short circuit current, and lower PCE. Further ink optimization is required to match achieve comparable performance with spun and printed



Figure 4.3: Comparison of best-performing current-voltage characteristics of spun devices, printed interlayer devices, and printed active layer devices. Each trace shows the best performing OPV, along with the power conversion efficiency. The printed active devices have a much lower short circuit current, and as a result, a lower PCE.

active films. Additionally, with the printed active devices, our yield was much lower, with few devices showing OPV current voltage characteristics. This nonuniformity may be helped by switching to substrates with an area more suitable for blade-coating.

For scaling to larger area devices, this would help bridge the gap between the research-scale and industrial-scale towards large-area processing. This work provides an essential step of transitioning to a scalable deposition method, but kept the same small substrate and device size. Some of the printed film variability may be due to the substrates being too small, letting the ink back flow, so transitioning to larger area devices will be more suitable for blade-coating, perhaps leading to more uniform performance while scaling up. In the big picture, this work provides a step towards large-area scalable fabrication of flexible solar cells. Our hope is that understanding the tradeoffs between spin-coating and blade-coating from films to device performance will encourage more to implement scalable and low-waste techniques to fabricate solar cells.

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