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Manufacturing Processes of Solution-processed Organic Solar cells and Recent Advances

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Abstract

This paper studies Organic photovoltaic cells as an evolving photovoltaic cell technology that has the potential to become an economically viable source of power that will lead to a sustainable future. Thanks to its uncomplicated fabrication technique and therefore the ability to be produced from solution have rapidly reduced the manufacturing cost of organic solar cells. During this review, the main target is the variation of coating and printing techniques for the fabrication of organic solar cells. The features of the fabrication technique alongside their importance within the organic solar cell technology are discussed next. The review concludes with a discussion on the recent advancements in the sector of organic photovoltaic cells, a comparison between the cost of the traditional and organic solar cell technology, and future perspectives for this promising technology.

Keywords
Organic solar cell, Solution-processed solar cells, Manufacturing processes, Cost-Effective.

1. Introduction
The depletion of non-renewable energy resources, the drastic effect of climate change, and the ever-increasing energy demand call for a reduction in the dependency on fossil fuel. The abundance of solar energy makes it a promising alternative to fossil fuel (Sista et al., 2011). However, the wide approval of solar energy is hindered by the high manufacturing and material cost of inorganic solar cells (Li et al., 2012). A viable solution to this can be organic solar cells. An organic solar cell is a technology that uses organic semiconductors and the formation of excitons as an intermediate for the energy conversion process (Hoppe & Sariciftci, 2004). Although the efficiency of organic solar cells is still lagging behind that of silicon-based cells, the fast, simple, and low-cost processing with non-toxic solvent makes scope for commercialization (Sampaio et al., 2020). While the traditional solar cells are produced by vacuum processing, the organic solar cells can be fabricated using film-forming techniques, which is it's considered economic. However, as organic solar cells are composed of multiple layers, each layer needs different film-forming techniques. An ultimate processing method for all the layers will make the fabrication process simpler. Also, the comparatively low efficiency of the organic solar cells requires more research on its stability and efficient manufacturing process. The ideal process should involve solution processing of all layers with non-toxic materials, on flexible substrates by the combination of as few film-forming techniques as possible.

1.1 Objectives
The objective of this study is to review and compare the existing and suitable film-forming techniques (coating and printing) for the large-scale production of polymer solar cells, and to review the current state of the production cost. It is hoped that this study will help as a motivation for future research in this area.

2. Literature Review
2.1 Coating and printing techniques
The processing of organic solar cell requires a high-throughput, low cost, reproducible fabrication technology for large scale application. The fabrication techniques should also ensure the optimum morphology of the layers and have high speed. These requirements prevent many of the existing film-forming techniques to be considered for the manufacture of polymer solar cells (Krebs, 2009). The film-forming techniques that have been used so far are spin coating, slot-die-coating, doctor blading, spray coating, gravure printing, flexographic printing, screen printing, and inkjet printing

(Q. Wang et al., 2016). Although the spin coating is not R2R compatible, it's a useful method for lab-scale application (Petri, 2002).

The coating and printing techniques can be categorized by whether there is contact between stamp and substrate. The coating operations are usually non-contact techniques and printing can be classified as contact techniques, the only exception being inkjet printing which is a non-contact technique (Sampaio et al., 2020).

Usually, coating processes can only produce an even layer of solution over the surface, without any pattern, giving zero-dimensional control. Slot-die-coating enables striped pattern over the layer; thus, it gives one-dimensional control. Printing processes can produce two-dimensional patterns over the surface.

The manufacturing process of organic solar cells heavily depends on the nature of the ink. Ideally, the solvents should be environmentally friendly and non-toxic, with water being the cheapest and best case. The rheology (viscosity) of the ink or paste is important for the specific deposition process. Some printing or coating techniques may require higher viscous ink than others. The viscosity in gravure printing and flexo printing is higher. Slot-die coating has a wide range of viscosity (Sumaiya et al., 2017)

2.1.2 Spin Coating

Spin coating is regarded as the most used and most important fabrication technique for a solution-processed organic solar cell in the lab. It uses the characteristic of centrifugal force and the interaction between polymer chain-substrate and substrate-solvent. Depending on their interaction, the polymer chain may wet the substrate if the interaction between chain and substrate is stronger. The chain will be driven away from the substrate if the substrate and solvent have stronger interaction (Petri, 2002).

The spin coating process can be categorized into several steps. Primarily, the ink or solution is deposited over the steady substrate. The deposition may also take place in a rotating substrate. The evaporation of the ink takes place while the substrate or plate is rotating. The film reaches its desirable thickness after the ink gets completely evaporated (Sampaio et al., 2020). The thickness of the film is controlled by the centrifugal speed, acceleration, viscosity of the ink, and temperature of the ink (Albrecht et al., 2012). The steps of spin coating are depicted in Figure 1.

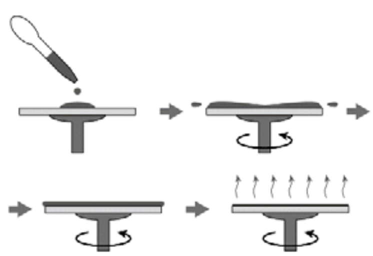

Figure 1: Principle of spin coating. (Top left) Initially, the liquid solution is applied to the substrate either during slow spinning or with the sample still. (Top right) The sample is rotated at a slow spin speed to distribute the solution. (Bottom left) The rotation speed is increased to give the desired thickness. (Bottom right) The rotation of the substrate is kept while the solvents evaporate (Sampaio et al., 2020).

Spin coating is highly reproducible and produces a homogenous film with the desired thickness. It's a zero-dimensional process as no patterns are created in the film. Although highly reproducible, the R2R incompatibility and high rate of wasted ink during rotation make spin-coating an unlikely method for large scale application. However, the wastefulness of spin coating is concluded as not critical since a very small volume of ink is required for the process (Krebs, 2009).

2.1.3 Slot die coating

Slot die coating is high throughput, one-dimensional coating process that has been used to coat the active layers in multiple organic solar cells (Berny et al., 2015; Krebs et al., 2010). Along with uniform thickness, it's suitable for coating multilayer cell with striped pattern. In literature, slot die coating is categorized as a lab-scale R2R method (Sampaio et al., 2020).

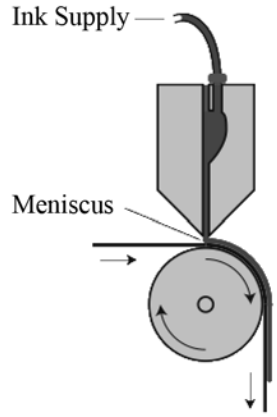

Figure 2: Principle of slot-die coating (Sampaio et al., 2020).

As depicted in Figure 2, the slot-die-coating process consists of a moving substrate where ink is being pumped through a coating head. The ink is continuously deposited over the moving substrate, which forms an even layer of thickness of the ink (Søndergaard et al., 2013). The thickness of the layer is dependent on the characteristics of the ink being used, the gap between coating head and substrate, and the speed and temperature of the plate (Pérez-Gutiérrez et al., 2017). Throughput speed can be improved and multilayer formation can be ensured by implementing the double slot-die-coating method (Larsen-Olsen et al., 2012). There is little or no waste of ink in the slot-die-coating process.

2.1.4 Doctor Blading or Knife coating

Doctor blading is a zero-dimensional coating technique that can ensure a specified thickness without any pattern. It's called knife coating if used in large-scale R2R applications (Sampaio et al., 2020). In knife coating, ink is supplied to a rigidly held knife, which is very close to a perpendicularly moving substrate. While the substrate is moving, ink can only go through the gap between knife and substrate. As a result, a uniformed and defined thickness is formed (Søndergaard et al., 2013). In doctor blading, the substrate remains rigid and the blade is dynamic. The blade moves over the substrate with a fixed speed and forms a uniform layer (G. Wang et al., 2019). The doctor blading process is shown on Figure 3.

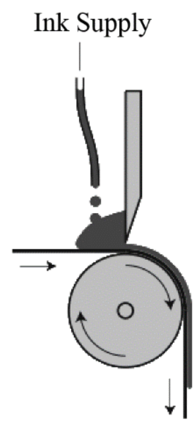

Figure 3: Principle of R2R knife coating or doctor blading (Sampaio et al., 2020).

The thickness of the film depends on the viscosity and surface tension of the ink. It may also vary due to the surface energy of the substrate. Despite the doctor blade being considered as a wasteless approach, the amount of ink lost initially while trying to implement the optimum condition is huge (Krebs, 2009). Solvents with high vapor pressure are not considered for doctor blading since the desired morphology may not be obtainable due to the fast evaporation rates (Pérez-Gutiérrez et al., 2017). Also being a slow process, the ink may crystallize. That's why spin coating is preferred for laboratory production (G. Wang et al., 2019). However, there is little or no waste of ink after initial application and high precision makes it suitable for large-scale applications.

2.1.5 Spray Coating

The spray coating technique is a widely used process in the automobile industry (Steirer et al., 2009). It also has a myriad of applications in the processing of organic solar cells. The step-by-step spray coating includes atomization of ink, evaporation of the solvent, impact on the substrate, and drying. As shown in Figure 4, the ink gets forced through a nozzle repeatedly. Outside of the nozzle, the ink disintegrates to lesser droplets by the presence of pneumatic flow. This is called atomization of the ink. The atomized droplets are directed to the substrate. Since the solvent has higher vapor pressure in the solution, evaporation may occur. Evaporation of the solvent will increase the concentration of the droplets which will hinder the even span of the ink on the substrate. The viscosity and boiling point of the solvent plays an important role in the evaporation of the solvent. Consequently, the feature of the used ink is a determinant of the efficiency of the spray coating. To avoid unnecessary shattering of ink on the substrate, the impact velocity of the droplets should be moderate or low (Reale et al., 2015).

The drying of the ink should be optimized to ensure even thickness (Sampaio et al., 2020). Therefore, it's clear that the productivity of spray coating is heavily dependent on the nature of the ink. Although spray coating is a zero-dimensional process, some patterning is possible on a millimetre-scale (Krebs, 2009). The process is R2R compatible and the thickness of the film can be efficiently controlled.

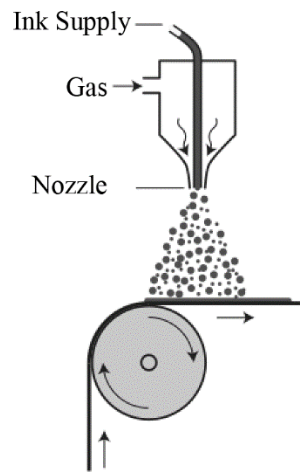

Figure 4: Simplified principle of spray coating (Sampaio et al., 2020).

2.1.6 Gravure and Flexographic Printing

Gravure printing is a complex, high-speed, and R2R compatible method that involves two rollers, an ink bath, and a moving substrate. It's a two-dimensional high-throughput process. Gravure printing has been described as a popular method for manufacturing printed electronics (Kapnopoulos et al., 2016). One of the two rollers has small cavities in it, this is the printing roller. The other roller is called the impression roller.

The printer roller stays partially immersed in the ink bath. While the printer roller is rotating, the small cavities are filled with ink. A doctor blade is used to trim excess ink from the printer roller. The remaining ink is then transferred to the substrate, which is guided by the impression roller (Sampaio et al., 2020). It's considered an expensive process since a printer roller can only engrave a unique pattern. To obtain a different pattern, a new roller has to be introduced (Krebs, 2009). Figure 5 consist the principle of gravure printing.

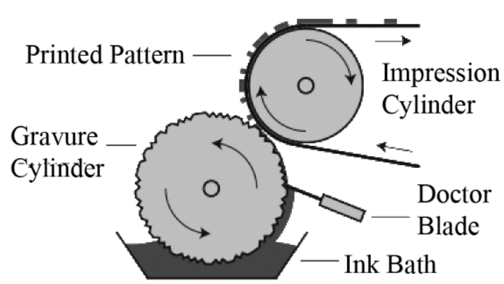

Figure 5: Principle of gravure printing (Sampaio et al., 2020).

A fairly recent printing technique used for the fabrication of organic solar cells is flexographic printing. It's an economical approach to gravure printing. Instead of two rollers, it consists of a fountain roller, an anilox roller, a printing cylinder, and an impression roller. The operation method remains similar to the gravure printing process. Flexographic printing is considered economical as the cost of the printing cylinders is less than the engraved cylinders (Søndergaard et al., 2013). Figure 6 depicts the principle of flexographic printing.

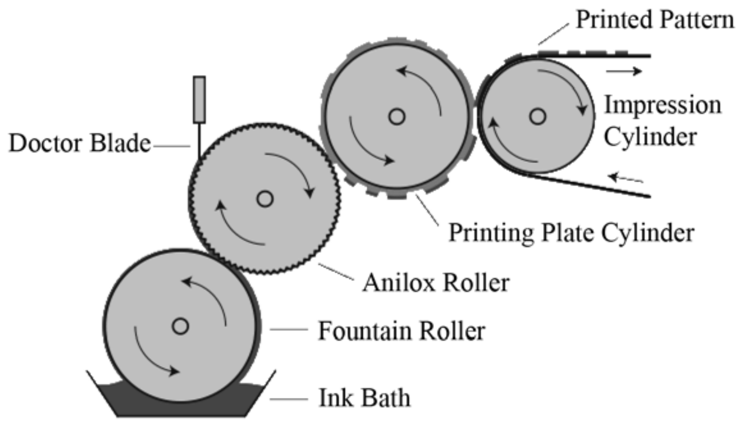

Figure 6: Principle of flexographic printing (Sampaio et al., 2020).

2.1.7 Screen Printing

Screen printing is a parsimonious, two-dimensional printing method. It's also an economical process considering that ink loss is kept at a minimum. Screen printing contains a screen with openings kept at a certain distance from the surface. The screen is filled with a coating solution and the printing area is kept open. A squeegee is introduced to the screen for which the screen and substrate are brought to contact. The linear movement of the squeegee forces the ink through the openings of the screen. Thus, patterns with uniform thickness are created (Krebs, 2009). The flat-bed screen printing process is depicted in Figure 7.

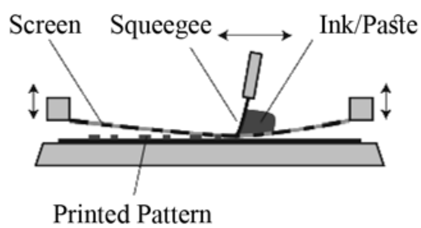

Figure 7: Principle of flat-bed screen printing (Sampaio et al., 2020).

Screen printing is viewed as economical as the waste of ink is minimum and the cost of the screen is low (Sampaio et al., 2020). However, as the screen remains open to the atmosphere, the ink may dry out in industrial applications. Also, there might be complications while screen printing active layers. Since a large wet thickness is achieved after screen printing, the desired morphology may not be obtained. Despite the complication, it has been possible to fabricate organic solar cells entirely with screen printing (Krebs et al., 2009).

2.1.8 Inkjet printing

Inkjet printing is a two-dimensional, low-cost printing process that has been developed from the typical inkjet printer used in home or office, only the ink is an organic solution here (G. Wang et al., 2019). The basic working principle of an inkjet printer is divided into droplet formation and ejection of the droplets. After that, the positioning of the droplets takes place over the surface. The ink droplets are either formed by the piezoelectric effect or heating (Krebs, 2009). The ink droplets are forced through a nozzle and pass through an electric field, where they get electrostatically charged. The charged droplets are then pushed to the substrate for deposition.

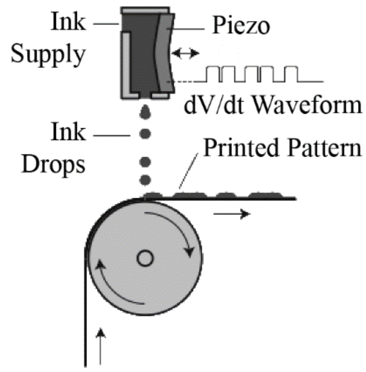

Figure 8: Simplified principle of inkjet printing for R2R processing (Sampaio et al., 2020).

After positioning, the evaporation of the solvent takes place and the solid film is formed. The operation type of an inkjet can be divided into very fast continuous inkjet printing and drop-on-demand inkjet printing. The continuous inkjet printing is very fast, but the printing area is limited since a single nozzle is used. The drop-on-demand inkjet printing consists of multiple nozzles and they are capable of high web speeds (Sumaiya et al., 2017). The formation of ink in inkjet printing is still not fully understood. Hence, more study is required before it's considered an important fabrication technique for the large-scale application of organic solar cells (G. Wang et al., 2019). The inkjet process is shown in Figure 8.

2.2 Roll-to-Roll technique

The fabrication processes described up to here are performed on a substrate which is a very long sheet. The folded roll of substrate, also called web material is uncoiled and moved through the coating or printing machine. Once done, the processed web material is coiled back to a roll again. Figure 9 contains the basic mechanism of the roll-to-roll process.

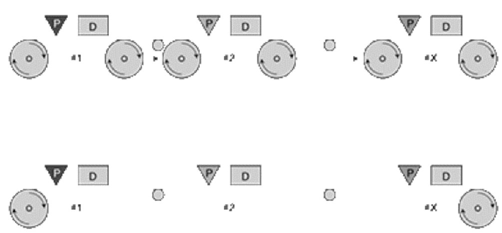

Figure 9: Discrete (Top) and Inline (Bottom) processing (Sampaio et al., 2020).

This is the basic operation of the desired roll-to-roll process. The integrated roll-to-roll technique expects that unprocessed web material going through the process will give a processed organic solar cell as an output. However, each coating and printing method requires a different time. Also, the drying of the film should is an important factor. Therefore, in literature, it has been advised to discrete roll-to-roll technique, where each layer will be coated or printed at a time (Krebs, 2009)

2.3 Organic Solar Cell Cost Analysis

The contribution of different elements associated with the manufacturing of OSC to total OSC cost is shown in Figure 10 (Mulligan et al., 2014). From the figure, it is evident that most of the costs originated from the materials, with operating costs also contributing significantly. The majority of this material cost comes from the PET substrate that contributes nearly half of the total cost. The use of OSC in this regard is quite significant as it offers a much lower substrate cost compared to the traditional solar cells.

The benefits of using OSC is that most layers are solution-processed, scalable, and vacuum free. The roll-to-roll process applies to most of the printing and coating techniques (Wang, 2016). This enables the transformation of OSC production from laboratory to Industrial scale. Kalowekamo et al. estimated that the manufacturing cost for OSC will range between \$48.8 and \$138.9/m^2 (Kalowekamo & Baker, 2009). Considering 5% module efficiency, 98% module yield, and 100 Wp/m^2 power output this results in a module cost between \$1.0 and \$2.83/Wp. This cost range is economically compatible with the traditional silicon-based PV technologies.

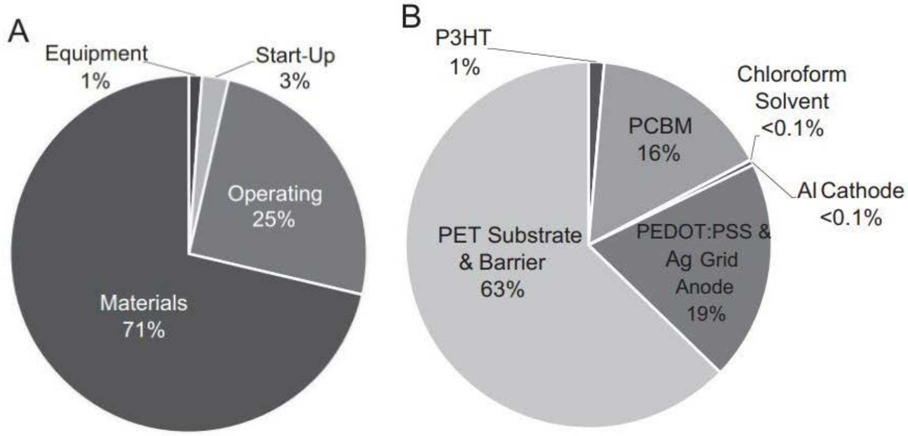

Figure 10: Contribution of different elements of the manufacturing costs to overall OSC cost. (B): Contribution of the separate materials components to the overall material cost (Mulligan et al., 2014).

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However, in terms of the general electricity market, the Levelized Energy Cost (LEC) for OSC ranges between \$0.49 and \$0.85/kWh when an assumption of 5% efficiency and 5 years lifetime is made, which is common for the currently available OSC module. Comparing the LEC cost for OSC with the general electricity market, it is observed to be far from being competitive. Although reports show that increasing the efficiency and lifetime of OSC can establish the feasibility of OSC significantly (Gambhir et al., 2016). With the higher efficiencies, this technology will attract its uses in broader fields and a great improvement on LEC will be apparent if the technology lasts more than 10 years; however, will experience less impact for increasing lifetime from 20 to 30 years (Shown in Figure 11).

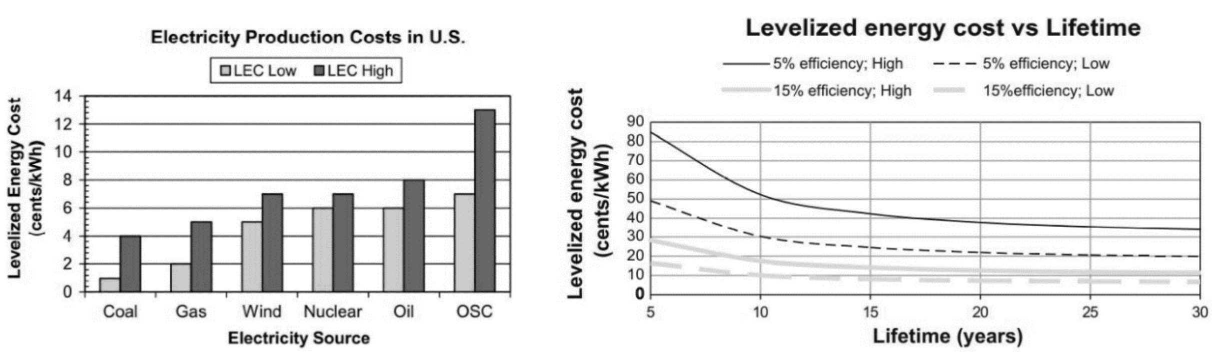

Figure 11: Electricity production cost in the U.S. based on 2002 averages and that of OSC (Organic Solar Cell) base. and LEC for OSC as a function of cell lifetime and efficiency based on the baseline low and high-end overall module manufacturing cost estimate (Kalowekamo & Baker, 2009).

It is estimated that LEC for OSC will range between \$0.07 and \$0.13/ kWh only if the industry can achieve an efficiency of 5% and a lifetime of 20 years. At this rate, the LEC of OSC is less than other PV technologies, whose average LEC is $0.25–$0.50/kWh. Compared to conventional electricity the cost is still higher than most conventional electricity but can be compatible at the economic level.

Replacing OSC technology for electricity generation in place of conventional electricity generation might not be a suitable option at the moment, but the use of OSC instead of traditional Si-based PV can be helpful in this regard that it offers a relatively simple manufacturing technique. Most importantly, with higher efficiency, OSC has lower energy payback time and overall cost compared to most of the traditional Si-based PV (García-Valverde et al., 2010) as shown in Figure 12.

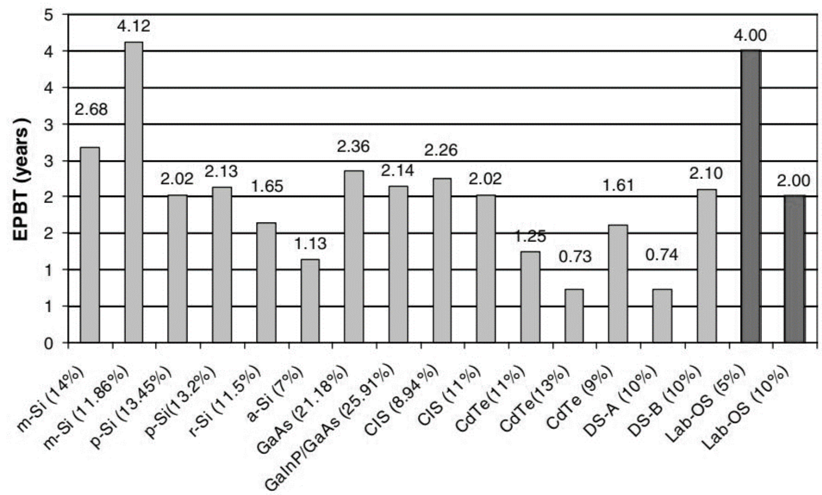

Figure 12: Energy payback time for PV modules technologies. Here DS indicates dye-sensitized modules and A and B stands for the low and high values of the energy range. Lab-OS means laboratory organic solar modules (García-Valverde et al., 2010).

3. Results and Discussions

The coating and printing processes have some differences regarding their R2R feasibility, dimension, ability to implement patterns and waste of ink. The differences are described in Table 1. An optimized combination of coating and printing methods to fabricate the multilayer structure and pattern formation of the organic solar cell is required for commercialization, which is yet to be found. The reason behind that is the varying operation speed, required properties of ink, and other operating methods of the fore-mentioned processes. The application of NFA-based devices has greatly increased the highest efficiency of organic solar cells over the past few years (Brabec et al., 2020). The stability of NFA based devices and FA based devices didn't show much difference. It's also been cited that NFA based devices will show greater stability as it has tunable properties.

NFA based devices can be tuned for operating on a range of illumination. Compared to silicon-based devices, greater working performance under low-light illumination has been shown by NFA based organic solar cell which was blade coated and screen printed (Luke et al., 2021). Since organic solar cell has a lower efficiency than the conventional solar cell, the proper utilization of properties like low-light applications, simple and low-cost processing, aesthetic application and flexibility may create chances for commercial success.

Table 1: Comparison of printing and Coating

Properties	Spin Coating	Doctor Blade	Spray Coating	Slot-Die Coating	Gravure Printing	Flexographic Printing	Screen printing	Inkjet printing
Dimension	0	0	0	1	2	2	2	2
Ink Waste	Significant	Little	Some	None	None	None	None	None
Pattern	No	No	No	Yes	Yes	Yes	Yes	Yes
R2R Compatible	No	Yes	Yes	Yes	Yes	Yes	Yes	Yes

4. Conclusion

The manufacturing technique for an organic solar cell is easy, simple, and exhibiting great potential on the laboratory scale, but to implement the technology on a larger scale a dedicated amount of work is required. While producing all layers of OSC using a single film-forming technique is much anticipated, it occurs rarely. In the production of OSC main focus need to be put on to reduce the steps, as it will save time and money. The paper discusses the characteristics of each of the recognized printing and coating techniques and gives direction towards the usage of each technique. To be industrially suitable the printing and coating techniques should be roll to roll compatible, ink wastage has to be minimal. Economic and environmental feasibility analysis is essential before applying the processes from a small to a bigger scale. OSC has a strong appeal in the theme of sustainability that intensive research is on this field will ensure its use on a commercial level to meet the prospect it's showing.

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