Solar Energy Materials & Solar Cells 149 (2016) 154–161

Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Reverse gravure coating for roll-to-roll production of organic photovoltaics

Doojin Vak [a,*], Hasitha Weerasinghe [a], Jyothi Ramamurthy [a], Jegadesan Subbiah [b], Michael Brown [a,b], David J. Jones [b]

[a] Commonwealth Scientific and Industrial Research Organisation (CSIRO), Manufacturing Flagship, Bayview Avenue, Clayton, VIC 3169, Australia
[b] Bio21 Institute, University of Melbourne, 30 Flemington Road, Parkville, VIC 3010, Australia

ARTICLE INFO

Article history:
Received 24 December 2015
Received in revised form
12 January 2016
Accepted 17 January 2016

Keywords:
Organic solar cells
Reverse gravure coating
Gravure printing
Printed electronics
Flexible solar cells
Roll-to-roll process

ABSTRACT

Reverse gravure (RG) coating is reported here as an alternate film deposition method for potential large scale roll-to-roll production of organic photovoltaic devices (OPVs). The basic working principles of RG coating are shown and compared to the more well-known gravure printing. Gravure printing is similar to RG coating from a process point of view, but the films produced using each method are very different to each other. An optical thickness measurement system was developed and used to monitor film thickness variation of RG coated photo-active layers with various coating parameters *in situ* in the roll-to-roll process. Partially and fully printed OPV modules were fabricated using, primarily, the roll-to-roll RG coating process and devices showed 2.1% and 1.5% power conversion efficiencies, respectively.

1. Introduction

Thin film, solution-processed organic photovoltaic devices (OPVs) are a promising future energy source [1–4]. OPVs can be fabricated using industrial roll-to-roll printing/coating techniques, facilitating low cost, high volume production. OPVs can be flexible and/or semi-transparent, enabling easy integration into building materials, as well as other non-conventional applications [5,6]. Although OPVs have many unique advantages over other solar technologies, the power conversion efficiencies of these devices have been lower than other commercialized photovoltaic technologies. However, intensive research on OPVs has resulted in rapidly increasing efficiencies from laboratory devices and over 10% power conversion efficiency (PCE) has been achieved from single junction OPVs [7,8].

Most laboratory investigations into OPV technologies to date have been conducted using spin coating as the material deposition method [9]. While spin coating is useful to study fundamentals, it is not suited to large scale, roll-to-roll production. Other scalable processes must be used for the fabrication of large area OPVs, resulting in discrepancies between record efficiencies of laboratory devices and that of large area printed devices [10,11]. Well-optimized fabrication schemes employing spin coating are typically not transferable to other application processes and many device configurations used in research devices are not compatible with printing. Accordingly, it is very important to consider scalability and compatibility of processes for the commercialization of large scale OPVs. If small-scale but scalable processes are used for research and development, the laboratory process can be translated to a roll-to-roll process without repeating the optimization procedure [12].

Large area OPVs have been produced using various scalable processes such as blade coating [13], spray coating [14], screen printing [15], gravure printing [16,17], inkjet printing [18] and slot die coating [19,20]. Among these methods, slot die coating has been most widely used to prepare photo-active layers of OPVs. A slot die coating head consists of several components assembled in such a way that the ink in the enclosed head is pressurized. Slots in the coating head determine the pattern produced and the ink is forced through these slots to form a uniform film. Slot die coating is a pre-metered process; all of the supplied material is deposited on the substrate and no material is wasted [21]. As a result, wet film thickness can be controlled by the predetermined flow rate, coating width and web speed. Slot die coating is a one dimensional coating technique, *i.e.* by moving the substrate past the coating head it is possible to produce the striped patterns that are required for the production of series-connected modules. Due to these

* Corresponding author: Tel.: +61 3 9545 7827.
E-mail address: Doojin.Vak@csiro.au (D. Vak).

http://dx.doi.org/10.1016/j.solmat.2016.01.015
0927-0248/

practical advantages, slot die coating has been the most successful method for the large scale OPV production.

Here we report reverse gravure (RG) coating as an alternate method for large scale roll-to-roll production of OPVs. Although the RG coating process has been used for fabrication of other organic electronics [22], discussed in review articles as an alternative OPV production method [21] and presented as a fabrication method of small flexible OPVs [23], we found no peer-reviewed journal article on RG coating for the fabrication of OPVs. Therefore, basic working principles of RG coating are discussed in comparison to the better known gravure printing process. Fully printed OPV modules are fabricated using, primarily, the roll-to-roll RG coating process.

Gravure printing is a high-throughput industrial printing method that produces high quality images. It is commonly used to produce magazines and packaging materials, and has been considered a promising alternative technique for commercial-scale production of printed OPVs [24]. Gravure printed OPVs have been demonstrated using a lab-scale benchtop printer [16], a roll-to-roll printer [25], and an industrial scale proofer [17]. Although the process has been successfully used for large scale printed OPVs by many research groups, including us, we found that the method is not ideal for the production of OPVs due to some inherent characteristics of the process.

In its simplest form, gravure printing consists of a two-part system; a gravure cylinder and an impression roll. The gravure cylinder is engraved with discrete cells in the pattern to be printed and is partially immersed in a bath of ink. Any excess ink from the engraved cells is removed from the cylinder using a doctor blade immediately before the roll is brought into contact with the web. The gravure cylinder and the web run in the same direction. Ink in the engraved cells on the coating roll is transferred to the substrate in the form of dots in same pattern as the engraved cells. In typical printing conditions used for image production, each dot is separated from the surrounding dots so that high contrast images can be produced. OPVs require continuous, pinhole free, uniform thin layers; discrete dots must be merged and any thickness variation of the wet film should be minimized through a levelling process. Although this can be realized by optimization of the ink formulation and printing conditions, it is very challenging to make defect-free large area coatings in this manner. In particular, high speed roll-to-roll gravure printing of OPVs is difficult as any levelling process requires time for the printed solution to spread out. Ideally, rather than deposition of an uneven wet film that requires a subsequent levelling process, a uniform wet film should be deposited initially.

RG coating, also called as microgravure coating or kiss coating depending roll system, is similar to gravure printing with regard to components. However, the process works very differently. As reflected in the name, *i.e.* coating rather than printing, the method is designed to produce a uniform layer and unable to produce two dimensional patterns. A uniform wet film is deposited, so no levelling process is required. The coating process can be seen in Fig. 1a. Similar to gravure printing, a coating roll is partially immersed in a bath of ink, with excess ink being removed from the roll using a doctor blade as shown in the inset. The coating roll can be thought of as equivalent to the printing roll in gravure printing. The major difference between RG coating and gravure printing is the rotational direction of the coating roll. As shown in the figure, the coating roll rotates in the opposite direction to the web during RG coating, resulting in the coating being applied to the web in a shearing manner. Two web guide rolls are used in RG coating, rather than the impression roll employed in gravure printing. The coating roll is typically touching the web with minimal pressure, this pressure originating from the tension of the web. Therefore the substrate is not trapped, or nipped, between the coating roll and the guide rolls. The absence of the nip eliminates many problems that are frequently encountered in printing and coating, such as web creases or breaks, along with other mechanical stresses that may affect coating quality.

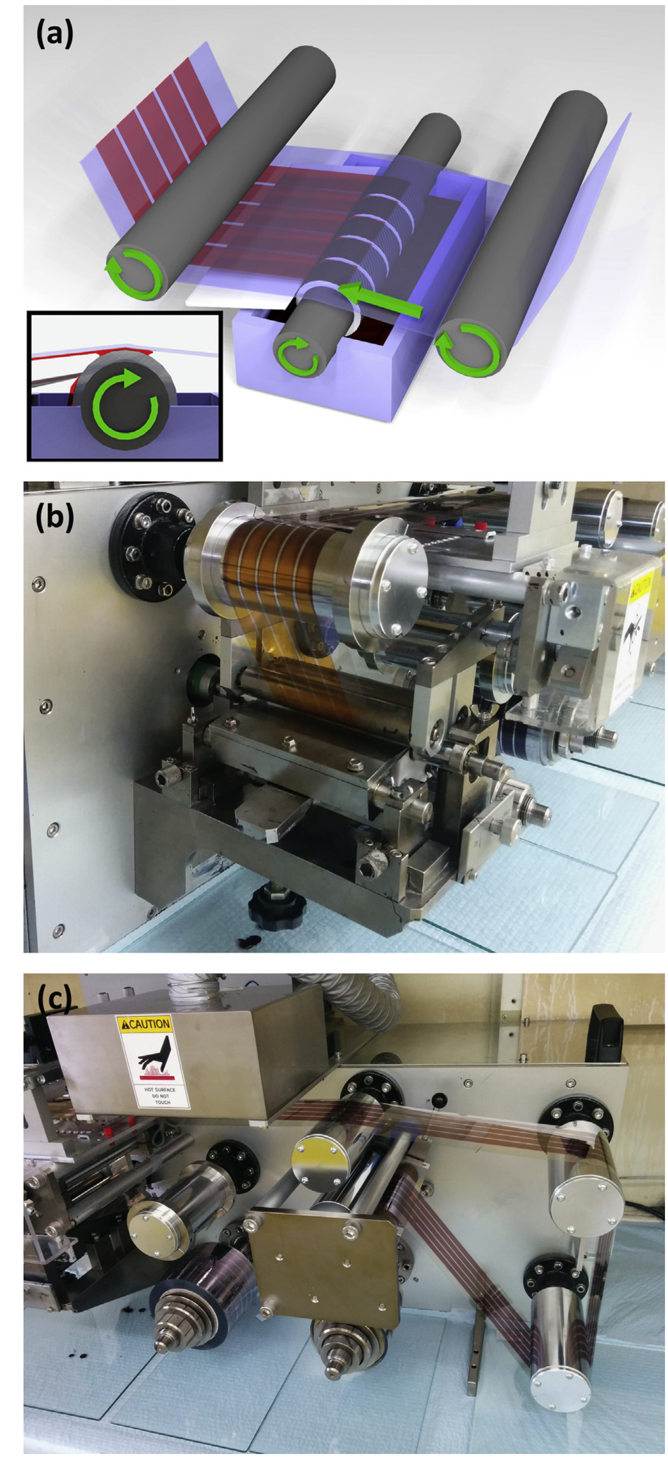

Fig. 1. (a) Schematic illustration of reverse gravure coating process. (b) Head unit of a reverse gravure coater and P3HT:PCBM wet film as prepared by the coater. (c) Roll processer of the reverse gravure coater and dried P3HT:PCBM film.

2. Experimental

2.1. Materials and formulations

ZnO nanoparticle solution (20 mg/ml in ethanol) was prepared by previously reported procedure [26]. For the photoactive layer, poly(3-hexylthiophene) (P3HT, SP001, Merck) and phenyl-C61-butyric acid methyl ester (PCBM, Tech Grade, Solenne) blend was prepared by dissolving 0.7 g of P3HT in 10 ml of co-solvent