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Inkjet printing processing conditions for bulk-heterojunction solar cells using two high-performing conjugated polymer donors

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ABSTRACT

We report the processing conditions for inkjet-printed active layers of organic photovoltaic (OPV) devices comprising bulk-heterojunction blends of the low bandgap conjugated polymer poly[(4,4'-bis(2-ethylhexyl)dithieno[3,2-b:2',3'-d]silole)-2,6-diyl-alt-(4,7-bis(2-thienyl)-2,1,3-benzothiadiazole)-5,5'-diyl] (Si-PCPDTBT) as well as poly[N-9'-hepta-decanyl-2,7-carbazole-alt-5,5'-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)] (PCDTBT) used as electron donors with [6,6]-phenyl-C71-butyric acid methyl ester (PC₇₀BM) as electron acceptor. By controlling the inkjet printing processing conditions, we gain a significant improvement in device power conversion efficiency (PCE) to previously reported work using these materials. We obtain Si-PCPDTBT:fullerene and PCDTBT:fullerene-based inkjet-printed OPVs with power conversion efficiency of ~3% and ~4%, respectively.

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1. Introduction

In the last decade the interest in renewable energy sources has grown rapidly. The sun plays a crucial role as its incident light on Earth provides an abundant source of energy—the amount striking Earth in less than an hour is sufficient to satisfy the average yearly human consumption [1]. The research community has therefore been focused on harnessing this energy with the aim of providing a stable and renewable method of producing electricity. OPVs can contribute to this need for sustainable energy resources by providing a low-cost, solution-processable and environmentally friendly alternative.

Bulk-heterojunction (BHJ) devices are proving especially worthwhile candidates, most commonly making use of an electron donating conjugated polymer and an electron accepting fullerene unit to provide a flow of electrons to the electrodes. OPV BHJ devices are now reaching a power conversion efficiency (PCE) above certified 8% [2]; with even above 10% being reported [3].

One of the fundamental concerns for organic photovoltaics (OPVs) is compatibility with printing manufacturing. Only then can OPV commercialization be truly viable [4]. One such compatible technique is inkjet-printing. Its drop-on-demand (DoD)

technology allows the ink droplets to be deposited exactly where required on the substrate [5]. This technique demands specific processing parameters in order to achieve morphology control similar to OPV device fabricated with common lab-scale techniques, such as doctor blading or spin coating [6–9].

An inkjet printer can also be seamlessly integrated into a roll-to-roll (R2R) production process, which can produce a large throughput of cells or large area devices [10]. Inkjet printing has been used to manufacture all the main components of an OPV device—from the metal electrodes to transparent conductive materials and also the active layers. The processes used for each of these components differ greatly and competing techniques exist with regard to high throughput, degree of accuracy, material consumption and energy payback [11]. Inkjet printing mainly competes with screen printing or flexographic printing for R2R compatible deposition of the metal electrodes, such as a transparent grid structure to replace ITO or a bulk metal structure as a back electrode [12,13]. For high throughput active layer deposition slot-die coating may be viewed as preferable, however, inkjet printing is still advantageous with regard to low material consumption and flexibility because of its DoD properties.

Due to the fact that the intricate network of conjugated polymer donor and acceptor morphology within a BHJ blend determines the performance of an organic solar cell, it is important to preserve this intricate morphology when producing thin films from a liquid solution. It is therefore imperative to gain knowledge

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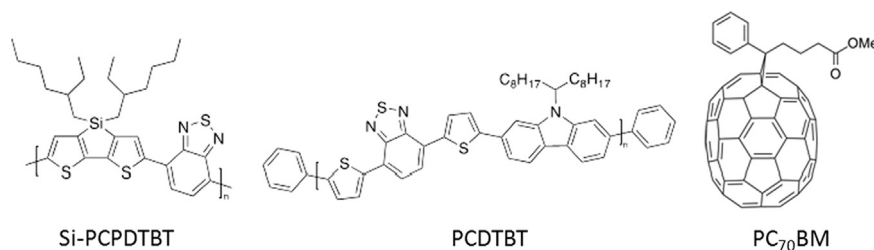


Fig. 1. Chemical structures of materials used in this study.

on desired inkjet-printing parameters to further improve obtained device performance [14]. The potential of combining inkjet-printed active layers with other inkjet-printed components of the OPV device, such as the bottom electrode currently mostly comprised of expensive indium tin oxide [15,16], can pave the way for highly efficient fully inkjet-printed devices.

To date, good inkjet-printed solar cells with a power conversion efficiency of approx. 3–4% have been achieved using active layer blends of the well-known poly(3-hexylthiophene) (P3HT) and phenyl-C71-butyric acid methyl ester (PC₇₀BM) [17–21]. However, high performing conjugated polymers have so far not been widely included in inkjet studies. One of these high performing conjugated polymers is poly[(4,4'-bis(2-ethylhexyl)dithieno[3,2-b:2',3'-d]silole)-2,6-diyl-*alt*-(4,7-bis(2-thienyl)-2,1,3-benzothiadiazole)-5,5'-diyl] (Si-PCPDTBT), yielding a certified efficiency of 5.24% with a doctor-bladed active layer [22], but a significantly lower efficiency of 0.64% when used in inkjet trials [6], primarily attributed to the low current density as well as fill factor achieved. This conjugated polymer is particularly interesting as it has a low bandgap of approx. 1.7 eV and therefore absorbs across a wide wavelength range around 700 nm [22]. This makes it a worthwhile candidate for several organic solar cell applications, including tandem cells [23]. The second conjugated polymer included in this study is poly[N-9'-hepta-decanyl-2,7-carbazole-*alt*-5,5'-(4,7'-di-2-thienyl-2',1',3'-benzothiadiazole)] (PCDTBT), which has been shown to achieve a power conversion efficiency of approx. 7% and high IQE values approaching 100% [24]. This conjugated polymer has so far not been utilised in studies involving inkjet printing.

In this paper we discuss the processing conditions to fabricate efficient BHJ organic solar cells based on inkjet-printed active layers using the materials shown in Fig. 1. By employing current/voltage, atomic force microscopy, photoluminescence quenching and external/internal quantum efficiency techniques we discuss the results in relation to doctor-bladed reference devices.

2. Experimental details

The organic solar cells were deposited on pre-patterned indium tin oxide (ITO) substrates (Psiotec, UK) using a normal device architecture. First, a ~50 nm hole transporting layer of Clevios PH poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) (1:3.2 by volume with isopropanol) was deposited using a doctor blade (Erichsen, Germany) and annealed for 20 min at 140 °C in air. On top of this the active layers consisting of, respectively, Si-PCPDTBT:PC₇₀BM (1:1.5 by weight) and PCDTBT:PC₇₀BM (1:2 by weight) were doctor-bladed (for the reference devices) or inkjet-printed using a commercially available printer fitted with a piezo-driven 16-nozzle printhead with integrated reservoir and a nominal drop volume of 10 pL (Fujifilm Dimatix, USA). Both inkjet-printed and doctor-bladed active layers were dried in air by remaining on the heated surface until visible solvent evaporation had occurred. In a final step, ~20 nm calcium and ~100 nm aluminium were evaporated under vacuum at a chamber base pressure of

10⁻⁶ mbar. The finished devices were encapsulated under a cover glass (Ossila, UK) using UV-activated adhesive (Dymax, USA). All components of the organic solar cells, except for the final metal evaporation step and encapsulation, were handled and deposited in air. Over 100 cells were produced for this study, each with an active device area of 9 mm², accurately defined by the shadow mask used for evaporating the top electrode.

Si-PCPDTBT was purchased from Konarka, Germany, PCDTBT from 1-material, Canada, PC₇₀BM from Solenne, The Netherlands, and Clevios PH PEDOT:PSS from Heraeus, Germany. Uniform layer thicknesses were measured using a Dektak 150 step profilometer (Veeco, USA). Atomic force microscopy to analyse layer surface roughness and morphology was carried out in tapping mode using an Easyscan 2 system (Nanosurf, Switzerland). Device parameters and current/voltage characteristics were obtained using a sourcemeter (Keithley, USA) and solar simulator calibrated to 100 mW/cm² light intensity (Newport, USA). External quantum efficiency (EQE) measurements were performed on a setup comprising a monochromated light source originating from a xenon lamp, a mechanical chopper, a preamplifier and a lock-in amplifier (Newport Oriel, USA).

Internal quantum efficiency (IQE) was calculated by dividing the external quantum efficiency of each cell under study by the absorption spectra of the corresponding active layer. The absorption spectra have been calculated using the transfer-matrix formalism [25] in a MATLAB script provided by Ref. [26] based on our OPV device stack of glass/ITO/PEDOT/active layer/Ca/Al. The refractive index n and extinction coefficient k in our polymer:fullerene systems was $n=2$ and $k=\lambda a/4\pi$ (with λ the wavelength and a the absorption coefficient in our active layer) as per Ref. [26]. Good agreement is seen between the calculated k values and those experimentally obtained using ellipsometry [27,28].

Steady-state photoluminescence (PL) was performed using a 0.75 m spectrometer equipped with a charge-coupled device (CCD) camera with spectral response up to 1100 nm. The PL was quasi-resonantly excited by a continuous wave (cw), power-stabilized He–Ne laser at 632 nm, operated at moderate excitation densities of ~15 mW/cm². All PL data were acquired with samples placed in vacuum conditions (ca. 10⁻⁵ mbar). The PL quenching measurements were performed on a side-by-side sample geometry to allow a direct intensity comparison. The emission of each film was normalized to its optical absorbance (optical density) to take into account film thickness and concentration variations. All PL spectra were corrected for the instrument response at the detection range.

3. Results and discussion

An inkjet printer, while providing several advantages such as drop-on-demand technology, therefore using minimal material quantities, requires careful adjusting of the processing conditions to allow fabrication of smooth films. Due to the inkjet printing process taking place line by line, it is important that the active

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