

Comparison of large scale coating techniques for organic and hybrid films in polymer based solar cells

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ABSTRACT

Polymer based solar cells (PSC) can be manufactured in a continuous roll to roll process as a low cost regenerative energy source. Coating ink properties and film thicknesses of 30–200 nm are challenging with respect to the manufacturing process, which itself has an important impact on film properties and cell efficiencies. In this paper we compare the large area coating methods: knife coating, slot-die coating, and spray coating with laboratory spin coating. Properties of coating inks and a viscosity model for commercial PEDOT:PSS types are discussed. The significantly smaller viscosity to surface tension ratio, of typical coating inks for PSC compared to conventional coating inks, causes a different behavior during the coating process. Wet film thickness, homogeneity, and process stability and their dependence on process parameters are addressed for each coating method. Hole-conductive and photoactive layers, consisting of polymer-fullerene and polymer-nanoparticle blends, are then compared with respect to homogeneity, AFM topography and absorption spectra. First results indicate that the coating method itself has an impact on polymer-fullerene film morphology and opto-electric properties.

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1. Introduction

Rapidly increasing energy consumption as well as a decreasing fraction of energy generated from fossil fuels create a growing market for low cost photovoltaic technologies. The main advantage of polymer based solar cells (PSCs) is their potential to be produced by solvent based deposition in a continuous roll to roll process [1]. Hybrid cells may further increase the efficiency due to high absorbance, high conductivity, tunable size, and shape of semiconducting nanoparticles [2]. Power conversion efficiencies (PCE) are steadily increasing and have reached 9.2% and 3.6% on laboratory scale for polymer [3] and hybrid [4] cells, respectively. Though a number of groups have successfully proofed the principle of roll-to-roll manufacturing of PSC (e.g. [5–11]) with a record efficiency of 3.5% [5]; developing a stable process for large area coatings with high average efficiencies and reliability is still one of the major challenges for the technology.

The phase separation of n- and p-type semiconducting materials during processing from a homogeneous coating ink (Fig. 1) is controlled by material properties and processing conditions and determines the efficiency of the solar cell [12]. Device morphology is canonically optimized for lab scale spin coating which has little significance for a technical process, because it is limited to small areas and does not allow for an independent

variation of process parameters. The influence of drying kinetics has been investigated with a roll-to-roll compatible knife coating setup by Schmidt-Hansberg et al. for a poly(3-hexylthiophen-2,5-diyl):phenyl-C61-butyric-acid-methyl ester (P3HT:PCBM) blend [13]. As well the influence of annealing conditions has been documented by various groups (e.g. [14]). This sensibility of film properties for processing parameters suggests that coating parameters, or method, may also have an influence. Though polymer solar cells have been prepared by a variety of coating methods, a direct comparison and thus evaluation is difficult as different materials and processing conditions are used by different scientific groups [5,6,15–21].

In this paper, we aim to compare coating methods for the hole-conductive and active layer in polymer and hybrid solar cells, using an identical material batch and – as far as possible – identical processing parameters. A variety of coating methods are, in principle, suitable for preparation of one or more layers in solar cells, as discussed by Krebs [22]. Here we chose to focus on knife coating due to its simplicity and low holdup, slot-die coating due to its superior homogeneity and controllability, and spray coating as it allows coating of curved or rough substrates. Spin coated samples were prepared as a reference to laboratory experiments.

The experimental setup and procedure is described in Section 2. Typical values of fluid-dynamic properties for the applied coating inks are tabulated and a model for the viscosity of two common PEDOT:PSS types is proposed (Section 3). The properties of the applied materials, high requirements regarding film homogeneity and limited material availability differ from

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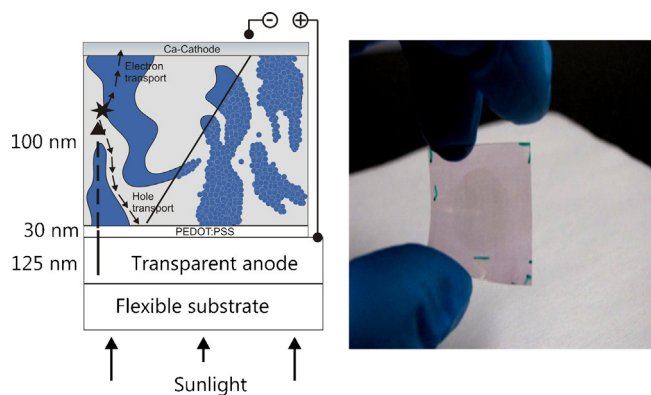


Fig. 1. Sketch of the working principle of polymer based solar cells (left). A flexible hybrid solar cell (prior to cathode deposition) produced by pilot scale slot-die coating.

processes with conventional coating and printing inks. This leads to different behavior regarding film thickness and stability of the coating process (Section 4). The semiconducting polymer and hybrid films were characterized with respect to homogeneity and surface roughness using a profiler and an atomic force microscope (AFM). As well, light absorption spectra are shown as a function of coating method, giving a first indication whether coating method has an influence on opto-electric film properties (Section 5).

2. Coating methods and procedures

The experimental setups for the investigated coating method are shown in Fig. 2. Knife coating was done with a commercial knife coater (ZAA2000, Zehntner) (Fig. 2a) typically at 70 μm gap and a coating speed of 5 mm/s. The miniature slot-die coater (Fig. 2b) used here is described elsewhere [23] and was applied in a speed range of 0.5–20 m/min and gap widths of 25 μm , 38.1 μm or 50 μm . A simple paintbrush (Vega2000, Thayer & Chandler, Fig. 2c) was used at 1 bar nitrogen differential pressure for spray coating and a spin coater (Fig. 2d) was employed at 500 1/s for PEDOT:PSS and 1000 1/s for active layers.

Either float glass or indium tin oxide (ITO) coated glass was employed as substrate, cleaned, treated with oxygen plasma and coated with PEDOT:PSS (Clevios PVP AI4083, HC-Starck). The PEDOT:PSS dispersion was diluted with water and methanol (MeOH) in a volume ratio of 1:1:0.25. The active layer consisted of the p-type polymer P3HT (6 mg/ml, Lisicon, Merck) and semiconducting nanoparticles (54 mg/ml, BayDots[®], Bayer Technology Services GmbH) or fullerene PC[60]BM (4.8 mg/ml, Solenne BV). The solids were dissolved in chlorobenzene (CB) and – for particle containing solutions – 5 vol.% pyridine (PYR) was added as stabilizing ligand. The thickness of the hole conducting layer (HCL) was set to \sim 30 nm and the hybrid (P3HT:QD) photoactive layer (AL) to 80 nm, for all coating methods except spray coating. Spray coated

films were thicker but due to their extreme roughness a precise determination of their film thickness was not possible. To allow for a direct comparison of organic and hybrid photoactive films, the P3HT content of the inks and the coating parameters were held constant, resulting in a dry film thickness of 40 nm for the organic (P3HT:PCBM) material system. Though the coatings processed in this study were large area (4–60 cm^2) the final fabricated solar cells had an active area of 0.24 cm^2 due to constraints of the experimental setup for cathode evaporation (100 nm Aluminum) and cell characterization under AM1.5 radiation (spectrally monitored ORIEL solar simulator). All layers were annealed for 1 h at 110 $^\circ\text{C}$ under nitrogen atmosphere.

3. Material properties

Table 1 gives an overview of the applied coating inks and their fluid dynamic properties at 20 $^\circ\text{C}$. As material prices are high (e.g. 653 $\text{€}/\text{g}$ regioregular P3HT [24]) and often not available in the required quantity for several test runs, model systems – consisting of polystyrene (PS) in xylene (XYL), and polyacrylamide (PAA) in water – were defined and used for fundamental coating experiments and calibration.

Where the organic active layer (AL)-inks and the model systems show pure Newtonian or only slightly shear thinning behavior (less than 66% decrease within the measured range), the PH500-PEDOT:PSS dispersion exhibits a shear thinning viscosity where the low shear rate is in the order of five times that of the low shear viscosity. All viscosities strongly depend on compositions and temperature. The composition of active layers is frequently changed and the values of viscosity and surface tension has to be determined for each coating ink. The inks for the hole-conductive layer are typically diluted from commercial PEDOT:PSS dispersions and can thus be described by a viscosity model.

The zero shear viscosity η_0 is determined by the type and concentration of the polymer in the dispersion and can be described at 20 $^\circ\text{C}$ by:

$$\eta_{0;20^\circ\text{C}}(x_{\text{Solid}}) = c_1 \cdot x_{\text{Solid}}^2 + c_2 \cdot x_{\text{Solid}} + \eta_\infty \quad (1)$$

where the solvent viscosity η_∞ can be measured or taken from the literature (e.g. [25]) if PEDOT:PSS is diluted with other solvents (e.g. methanol to modify wetting properties). x_{Solid} is the solid mass fraction in the PEDOT:PSS dispersion (e.g. 1.27 wt.% or 1.36 wt.% for undiluted PH500 and VPAI4083, respectively). The parameters c_1 and c_2 were determined for PH500 and VPAI4083 by a fit of experimental data shown in Fig. 3.

The shear rate dependent viscosity can be described by a Carreau-Yassuda model as a function of zero shear viscosity η_0 , solvent viscosity η_∞ , characteristic shear rate $1/\lambda$ and logarithmic slope ($n-1$):

$$\eta_{20^\circ\text{C}}(x_{\text{Solid}}, \dot{\gamma}) = \eta_{0;20^\circ\text{C}} + (\eta_\infty - \eta_{0;20^\circ\text{C}}) \cdot [1 + (\lambda \cdot \dot{\gamma})^2]^{(n-1)/2} \quad (2)$$

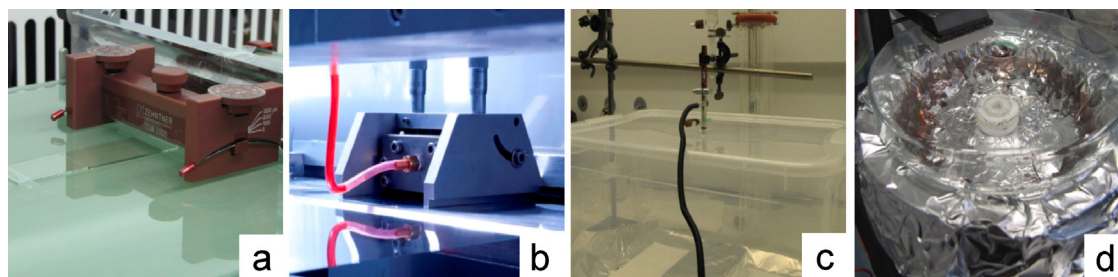


Fig. 2. Experimental setup for knife (a), slot-die (b), spray (c), and spin (d) coating of polymer based solar cells.

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