



Photonic sintering of inkjet printed current collecting grids for organic solar cell applications

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ABSTRACT

ITO-free organic solar cells with inkjet printed current collecting grids are demonstrated. For sintering those grids, thermal treatment and its faster alternative, photonic flash sintering, are applied and the characteristics of the resulting metal structures are compared with each other. The electrical potentials and resulting currents in the devices with different sintering conditions are calculated. The flash sintered current collecting grids exhibit clear advantages over thermally sintered grids in terms of geometry and conductivity. Similar conductivities are obtained after 5 s of flash sintering and 6 h of thermal sintering. This finding demonstrates the great potential of flash sintering for the roll-to-roll manufacturing of printed organic solar cells on flexible substrates.

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1. Introduction

The ITO-free approach for manufacturing organic solar cells has attracted a lot of interest in the last few years [1–3]. Vacuum deposited ITO with a relatively limited conductivity can be substituted by high conductive poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS) combined with current collecting metal grids [1,4–6]. Printed current collecting grids [1,7] are an interesting alternative for lithographic and evaporated grids. Printing constitutes a fast and cost-efficient method for the deposition of such metal structures [8,9]. Currently, the most commonly used conductive inks for those purposes are based on silver nanoparticle dispersions [8,10,11]. The disadvantage of printed metal structures is the necessity for post-deposition sintering to achieve sufficient conductivities.

The most common approach to achieve high conductivities in printed silver nanoparticle structures is thermal treatment, e.g. in a hot air oven [10,12,13]. During this process, the polymer shells that prevent the silver nanoparticles from agglomeration and thus stabilize the ink, are partially removed. Subsequently, the nanoparticles fuse and form a continuous network of conductive pathways. Generally, however, this process is rather slow, especially when inexpensive heat sensitive substrates are used, which limit the maximum process temperature. Typical examples are polyethylene terephthalate (PET) and polyethylene naphthalate (PEN), which are commonly used as a substrate for flexible OPV devices and can withstand heating up to only 110–130 °C without too much deformation. At these temperatures, however, process times to achieve conductivities of ten or more per cent of the value for bulk silver are generally in the order of several minutes or even longer. This is a severe restriction for the fast and cost-efficient high volume production of printed organic solar cells on foil, e.g. by roll-to-roll processes [14,15].

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Alternative sintering methods that are able to accelerate the entire process without damaging the plastic substrates are therefore in high demand. One possibility is the selective heating of the ink, in contrast to the indiscriminate heating of both foil and ink by thermal treatment. Laser sintering [16–18] offers a method to produce conductive tracks with limited heating of the substrate, by following the printed structures and sintering them selectively. However, this method is technically very complex and inefficient in terms of time and cost. Electrical sintering [19] is a method where the metal layer is sintered by applying a direct-current voltage over the printed structures. This causes current flow through the structure, resulting in local heating by resistive energy dissipation. Metallic structures with structural complexities higher than simple lines, however, will suffer from inhomogeneous sintering due to random local variations in the initial conductivities. Additionally, the direct-current method requires mechanical contact between the voltage source and the structure to be sintered, which makes it very difficult to apply in a roll-to-roll process. Microwave sintering [20–23], by contrast, is a non-contact method since it is based on the interaction with alternating electric fields. It is fast and can result in very high conductivities. However, it functions efficiently only in combination with a pre-sintering step (e.g. thermal treatment) that provides the metallic structure already with some initial conductivity. Furthermore, microwave sintering of complex metallic structures is very sensitive to field inhomogeneities that will almost inevitably lead to local overheating and damage.

Another novel sintering method compatible with low-cost plastic foils is the exposure of the printed patterns to a low pressure Ar plasma, which degrades the polymer shells around the nanoparticles [24], but its speed is at current status comparable with thermal sintering. Instead of by plasma exposure, the shells can also be removed by the addition of certain chemicals [8], but this process is expected to result in a bad long-term stability of the ink.

In photonic flash sintering [25], the distinctly different absorption properties of the conductive ink and the plastic substrate in the visible region are exploited. Whereas common plastic foils are typically transparent, silver nanoparticle inks are deeply coloured and strongly absorb visible light. Consequently, by choosing a lamp with an appropriate emission spectrum, energy can be coupled selectively into the printed ink structures without directly affecting the substrate. Obviously, heat conduction from the hot ink to the cool foil will tend to increase the temperature of the plastic as well, but excessive heating and thermal damage can be prevented by applying the energy in short pulses. Using this technique, temperatures can be achieved in the ink lines that are much higher than those reached by thermal treatment, resulting in a strong acceleration of the sintering process with only very limited deformation of the substrate. This method of sintering significantly reduces the processing time, compared to conventional thermal sintering.

In this paper, we present an approach to apply photonic sintering in the processing of printed current collecting grids in an organic solar cell. This method is not only much

faster than the traditional thermal treatment, but the resulting metal structures even give rise to enhanced performance characteristics of the OPV devices, such as short circuit current and fill factor. This method of sintering is applicable for both devices produced on glass and flexible barrier coated PEN substrates.

2. Experimental section

The current collecting grids were inkjet printed on both 0.7 mm thick glass substrates and thin-film barrier coated [26] PEN foil with a thickness of 200 μm using a piezoelectric drop-on-demand system (Fujifilm Dimatix DMP-2800). The printer can handle 16 nozzles and drop volume of 1 or 10 pL. Current collecting grids were printed using 10 pL cartridges (DMC-11610) filled with Suntronic U5603 silver nanoparticle ink. The Ag nanoparticles (average diameter 30–50 nm) are stabilized with PVP (poly(vinylpyrrolidone)) in a mixture of ethylene glycol, ethanol and glycerol. The droplets ejected from the nozzles have a diameter of about 20 μm . Uniform droplet formation along the 16 nozzles was performed by applying a jetting frequency of 10 kHz and a nozzle voltage of 24 V. Immediately after printing, the current collecting grids were sintered in an oven or flash sintered. Thermal sintering was carried out in a Memmert hot air oven, set to a temperature of 130 °C. Photonic flash sintering was carried out in a home-made apparatus, the details of which are disclosed in [27]. The inner surface of the elliptic reflector was covered by a highly reflective foil (reflectivity > 98%). The Xenon flash lamps (XOP-50, Philips, The Netherlands) have a maximum power of 1000 W, an emission spectrum ranging from 350 to 900 nm and a flashing frequency of up to 17 Hz. The resulting current collecting grid lines were visually inspected with a microscope, followed by measuring a cross-section area (Veeco Dektak Profilometer) of the lines on 5 different places of the grid.

High conductivity PEDOT:PSS (Orgacon™ from Agfa-Gevaert, Belgium) was inkjet printed on top of the current collecting grids with a Spectra Galaxy 256 print-head. The printed layers were baked at 130 °C for 10 min. The resulting PEDOT:PSS layer, with a thickness of 100 nm and a conductivity of 200 S/cm provided a sheet resistance of 500 Ω/sq .

The PEDOT:PSS layer was over-coated by a P3HT/PCBM blend. Poly(3-hexylthiophene) (P3HT) (Plexcore OS 2100, purchased from Plextronics, US) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) (99%, purchased from Solenne BV, The Netherlands) were dissolved in 1,2-dichlorobenzene with a mixing ratio of 1:1 by weight. The solution was stirred for 3 h at 90 °C. The photoactive layer was obtained by spin coating a 4 wt.% blend at 1000 rpm for 30 s, resulting in a film thickness of 220 nm. The thicknesses of the films were measured using a Dektak profilometer. The metal cathode (1 nm LiF, 100 nm Al) was thermally evaporated in a vacuum chamber through a shadow mask. The finished OPV devices, prepared on glass substrate, were encapsulated with stainless steel lids using Huntsman Araldite® 2014-1 sealer. Flexible devices were encapsulated with thin film barrier, com-

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