Laminated fabric as top electrode for organic photovoltaics

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A simple lamination technique for conductive and semitransparent fabrics on top of organic photovoltaic cells is presented. Conductive fabrics consisted of metal wires woven in a fabric with polymeric fibers. The lamination of this conductive fabric with help of a high conductive poly(3,4-ethylenedioxythiophene) polystyrene sulfonate formulation results in well aligned low resistive metal wires as top electrode. Semitransparent flexible organic photovoltaic cells were processed with laminated fabrics as top electrode and sputtered layers of aluminum doped zinc oxide and Ag as bottom electrode. The organic photovoltaic cells showed similar performance when illuminated through the bottom or top electrode. Optical simulations were performed to investigate light scattering effects of the fabric. Results are very promising for photovoltaic and lightning devices as well as for all kinds of devices where semitransparent, highly conductive, and non-vacuum processed electrode materials are needed. © 2015 AIP Publishing LLC.

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Organic photovoltaic (OPV) technology is attractive to solve future energy supply scenarios. OPV combines the attractiveness of harvesting energy from the sun with solar panels that can act as design element when integrated into buildings¹ or used for mobile applications. The certified power conversion efficiency (PCE) increased to over 11% for single devices² and semitransparent³ and lightweight cells have been demonstrated. Beside the further optimization of the active OPV cell materials, the improvement of the electrode and barrier materials is mandatory for more efficient, stable, and economic devices. Different concepts for electrode materials have been demonstrated. Included are electrode materials based on indium tin oxide (ITO), thin metal layers, carbon nanotubes (CNTs),^{4–6} Ag nanowires (AgNW),^{7,8} graphene,^{9,10} and conductive fabrics.^{11–14} In addition, the combination of different technologies has been reported, e.g., high conductive poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (Pedot:PSS) in combination with metal grid electrodes to assist the conductivity of the semitransparent layer.^{15–18} The figure of merit includes both, high transparency and a high conductivity. A high conductivity allows for larger area cells without dramatic losses in performance and therefore higher geometrical fill factors (FFs), while high transparency minimizes losses due to absorption. In addition, for solution processed organic electronic devices, the use of non-vacuum processes for deposition of the top electrode is important. The combination of organic solar cells with textiles is an interesting approach giving more functionality to apparels or using fabrics as flexible substrate.¹⁴ The well-known weaving process from the textile

industry can be adapted and the fabrics provide flexibility and mechanical stability. Two general approaches have been reported. Fibers or tapes are coated with the electrode and active materials and woven thereafter to fabrics.^{14,19,20} The other approach is coating^{11,12,21} or stitching^{22,23} the OPV cell on the fabric. In this publication, we concentrate on OPV devices with fabric electrodes, i.e., conductive metal wires and polymer fibers in a fabric (provided by Sefar AG, Thal, Switzerland).²⁴ The usage of a fabric as bottom electrode for OPV was shown in the previous publications, where the open spaces in the mesh were filled with a transparent and inert polymer, resulting in a very smooth surface with only the top of the metal fibers exposed to air.^{11,12} The bottom electrode consisted of the embedded fabric coated with a highly conductive Pedot:PSS for further planarization and acting as lateral transport layer for charge carriers. In this publication, a semitransparent non-vacuum processed top electrode for OPV is presented, the integration of the fabric without filler. The advantages over conventional electrode materials as top electrode are discussed.

Flexible OPV cells were solution processed on substrates made of sputtered layers of aluminum doped zinc oxide (AZO) and Ag (AZO/Ag/AZO) on a polyethylene terephthalate (PET) foil (provided by ROWO Coating Gesellschaft für Beschichtung GmbH, Herbolzheim, Germany and Fraunhofer ISE, Freiburg, Germany). The substrate has a conductivity of 10 Ω/□. PEDOT:PSS Clevios HTL Solar (Pedot:PSS HTL (1 S/cm)) and Pedot:PSS Clevios F ET (Pedot:PSS F ET (200 S/cm)) (Heraeus, Leverkusen, Germany), poly(3-hexylthiophene) (P3HT, Lisicon SP001,

Merck Chemicals, Darmstadt, Germany), and 1-(3-methoxy carbonyl)-propyl-1-phenyl-(6,6) C_{61} (PCBM, Solenne BV, Groningen, The Netherlands) were used as received. The substrate was cleaned by isopropanol and a sticky role. The solution processed layers were deposited by doctor blading (Zehntner ZAA 2300, Sissach, Switzerland) and the sequence of the layers is illustrated in Figure 1.

First, a layer of P3HT:PCBM (20 mg:16 mg dissolved in 1 ml *o*-chlorobenzene (anhydrous 99.8%, Sigma-Aldrich), 90 nm) was coated on the substrate. A layer (160 nm) of Pedot:PSS HTL was blade coated on top and the half-finished cell was annealed at 140 °C inside a nitrogen filled glovebox. Pedot:PSS F ET was blade coated on top of the Pedot:PSS HTL layer and the fabric was laminated onto the wet Pedot:PSS F ET film using finger pressure. Alternatively, the fabric was put on the dried Pedot:PSS HTL film directly and Pedot:PSS F ET was blade coated over the fabric with excessive material being removed by a cleanroom towel. By heating the device stack to 60 °C, the Pedot:PSS F ET strongly adhered to the fabric and formed contact between the metal wires and the OPV cell. The full OPV cell was then encapsulated between glasses with an epoxy based liquid adhesive. White light J-V characteristics were measured using 100 mW cm^{-2} simulated AM1.5G solar irradiation on a calibrated solar simulator from Spectra-Nova. The external quantum efficiency (EQE) was measured using a monochromator and the light from a 300 W Xe lamp together with an AM1.5G filter set which was built-up in the lab. The monochromatic light intensity was determined using a calibrated Si-diode. Typical active cell areas were in the range of 1 to 2 cm^2 .

In this work, a fabric made out of polymeric fibers (PEN = poly (ethylene 2,6 naphthalate) with Ag coated metallic wires woven in, provided by the company Sefar, is used. This fabric acts as top electrode, i.e., the metallic wires transport the current with little ohmic losses. Figure 2 shows a scanning electron microscope (SEM) image of the fabric laminated on top of an OPV cell. The conductive metal wires (white) are woven in one direction with a metal/polymer ratio of 1:3. The protruding metal touches and makes electrical contact with the high conductive Pedot:PSS F ET film. Since the wetting of the Pedot F ET is not well on the hydrophobic P3HT:PCBM surface (contact angle of 71°), we worked with a bilayer of 2 different formulations of

FIG. 1. Schematic on the device structure. OPV cells were processed on the substrate PET/AZO/Ag/AZO in the inverted device structure. On the substrate, the layers P3HT:PCBM, Pedot:PSS HTL and F ET were processed from solution and a fabric was applied by lamination.

FIG. 2. (a) SEM image of a laminated fabric electrode on top of an OPV cell. The 2 white threads represent 2 metal fibers, while the other threads are polymeric fibers. The imprint of the metal fiber into the Pedot:PSS layer is shown from the top (b).

Pedot:PSS. Pedot:PSS HTL wets very well on the P3HT:PCBM surface (contact angle of 32°) and therefore acts as interfacial layer between P3HT:PCBM and the Pedot:PSS F ET. In addition, this layer protects the underlying P3HT:PCBM layer from mechanical damage when aligning the fabric during the lamination process. This mechanical protection was also reported for screen printing Ag electrodes on OPV cells.²⁵ The highly conductive Pedot:PSS F ET contains an organic binder which gives strong adhesion between the fabric to the underlying Pedot:PSS HTL layer, which is still true when storing the cells for 15 weeks under ambient conditions. To demonstrate the wetting between Pedot:PSS and the metal wire, the finished solar cell was cut and the metal wire was partly detached. The SEM image is shown in Figure 2(b). The subjacent Pedot:PSS layer clearly shows the imprint of the metal wire. Capillary forces acting during lamination and drying become evident via the protruding polymer skin at the metal/Pedot:PSS interface. This effect is beneficial since the contact area between Pedot:PSS and the metal fiber increases and hereby reducing the contact resistance.

Finished OPV cells were glass-encapsulated using an epoxy based liquid adhesive. Figure 3 shows the current-voltage (JV) characteristics of a typical semitransparent OPV cell once illuminated through the fabric top electrode and once

FIG. 3. JV characteristics of a glass-encapsulated OPV cell. The same cell was illuminated ones through the AZO/Ag/AZO substrate (black rectangles) and ones illuminated through the fabric electrode (red circles).

illuminated through the AZO/Ag/AZO bottom electrode. EQE results are shown in the inset of Figure 3. The calculated short-circuit current (J_{sc}) was 5 mA/cm^2 for illumination through the fabric top electrode and 5.6 mA/cm^2 when illuminated through the AZO/Ag/AZO bottom electrode. Illumination through the AZO/Ag/AZO electrode resulted in an open circuit voltage (V_{oc}) = 0.57 V , $J_{sc} = 5.6 \text{ mA/cm}^2$, FF = 50%, and a PCE = 1.6%. Illumination through the fabric top electrode resulted in $V_{oc} = 0.57 \text{ V}$, $J_{sc} = 5.0 \text{ mA/cm}^2$, FF = 53%, and PCE = 1.5%. The performance is almost similar with a slightly reduced J_{sc} when illuminated through the fabric top electrode. This is due to the higher absorption of the adhesive/fabric/Pedot:PSS layer compared to the adhesive/PET/AZO/Ag/AZO layer.

Figure 4 presents the JV characteristics of the same cell when contacting the fabric at different distances from the active area of the cell.

The JV characteristics do not depend on the distance since the highly conductive metal fibers transport the current. The conductivity of the metal fibers is $0.0171 \Omega \text{ mm}^2/\text{m}$ with a diameter of each metal fiber of $45 \mu\text{m}$. This means that the resistivity of the wire increases by $27 \text{ m}\Omega$ per cm of wire. Taking into account that several wires are in parallel with a distance of 1.08 mm , the resistivity will decrease by the factor of the number of wires, e.g., $2.5 \text{ m}\Omega$ per cm of wire for a cell with the width of 1.08 cm . JV curves in Figure 4 were measured using a cell with a width of 1.1 cm . Therefore, the serial resistivity increases by $7.5 \text{ m}\Omega$ when measuring at a distance of 4 cm instead of 1 cm . For the JV characteristics, this effect can be neglected. A fabric with 1 cm in width and 1 m in length comprising 11 wires would add a resistivity of 0.25Ω only. Therefore, for moderate lengths, the resistivity of the wire is insignificant. A critical point for the overall resistivity, however, is the lateral surface resistivity between wires that is determined by the resistivity of the Pedot:PSS and the contact resistivity between Pedot:PSS and the metal fibers. The high conductive Pedot:PSS F ET formulation helped to minimize ohmic losses and adhered strongly to the metal wire.

These results suggest that conductive fabrics can be well used as laminated top electrodes for OPV cells and also acting

FIG. 4. JV characteristics of the same OPV cell when collecting the electron current at different distances from the light spot (1 cm , black rectangles; 2.5 cm , red circles; and 4 cm , green triangles).

as semitransparent bus bars transporting the current. This semitransparency helps to integrate the fabrics into, e.g., windows transporting the current through parts of this window. These high conductive fabrics open possibilities in module design and also the potential to optimize the PCE related to the used area (geometrical fill factor). Typically, the width of solar cells in modules is determined both by the conductivity of the electrode materials and the loss of active area due to interconnecting several cells to modules. Since the fabric metal wires have a very high conductivity, larger cells are possible and therefore the loss of inactive area can be reduced. Additionally, the shading effect due to the metal wires is small compared to other concepts for printed top electrodes. Typically, grid electrodes are used, i.e., printed metal grids on top of a current spreading layer such as Pedot:PSS. The printed lines typically have a larger line width and are less conductive¹⁶ than the metal wires of the fabric. Therefore, the lamination of the fabric has beneficial aspects compared to conventional solution processed top electrodes.

The fabric is scattering the impinging light.¹¹ We determined the angle dependence of the light scattering for free standing fabric meshes. Therefore, samples were illuminated and the intensity of the transmitted light was measured at variable angles using a silicon photodiode. We found that approximately 12% of light was scattered for angles $>2^\circ$, 7% for angles $>10^\circ$, 4% for angles $>20^\circ$, and 2% for angles $>30^\circ$. We created an optical model to identify whether the increased path length of the scattered light increases the absorption substantially and therefore is beneficial for the PCE. The commercial software setfos (Fluxim, Inc.) was used for the optical modeling. The model takes into account the layer sequence in Figure 1, spectroscopic ellipsometry data of all materials, and the degree of light scattering (Haze) at the fabric. Details on the optical model are given in the literature.²⁶ The EQE of the OPV cell was simulated with light incident through the AZO/Ag/AZO bottom electrode and with light incident through the fabric top electrode. Assuming 85% extraction efficiency, we obtained a good agreement between simulated and measured EQE for both illumination directions. For illumination through the fabric electrode, we obtained $J_{sc} = 5.6 \text{ mA/cm}^2$ for an exaggerated haze of 50%, $J_{sc} = 5.4 \text{ mA/cm}^2$ with a haze of 12% (as measured), and $J_{sc} = 5.4 \text{ mA/cm}^2$ without light scattering. We thus conclude that the effect of light scattering in this cell configuration is very small and that the observed difference in spectral shape can be attributed to the shading of the Pedot:PSS layer for top illumination and interference effects. For future optimization of the fabrics, we suggest to reduce the stitch density to increase the direct transmission rather than increasing the light scattering.

In conclusion, a simple lamination technique for fabrics on top of OPV cells resulting in a highly conductive transparent electrode material is presented. The conductivity originates from metal fibers which are woven in a fabric with polymeric fibers. With the help of solution processed layers of Pedot:PSS, the fabrics are glued on top of OPV cells resulting in an excellent electrical contact of the metal fibers to the Pedot:PSS and therefore to the OPV cell. For comparison, results are discussed on semitransparent cells with AZO/Ag/AZO as bottom electrode and conductive fabrics as

top electrode. The performance is almost independent on illumination through the bottom or top electrode, respectively. The resistivity added by the length of the metal fibers when contacting the fabric at different distances from the cell is very small. A conductive wire of a length of 1 m would add a resistivity of 2.7 Ω . The fabrics show light scattering of approximately 12%. Optical simulations show negligible influence of light scattering. For future developments, we therefore suggest to reduce the stitch density for increasing the direct transmission rather than increasing the light scattering. Summarizing, laminated conductive fabric electrodes are promising non-vacuum processed high conductive electrode materials for OPV but also for other PV and lighting technologies.

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