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Optimized Process of Fully Spray-Coated Organic Solar Cells on Woven Polyester Cotton Fabrics

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Abstract

This paper presents the novel use of spray coating to fabricate organic solar cells on fabrics for wearable energy harvesting applications and optimises photovoltaic efficiency. A fully spray coated photovoltaic (PV) device fabricated on fabric has been successfully demonstrated with comparable power conversion efficiency to glass based counterparts. All the PV devices are characterised under AM 1.5 (100mW/cm2) irradiation using an ABET solar simulator. Device morphologies are examined by scanning electron microscopy (SEM). The aim of this study is to develop and optimise a method to obtain reproducible photovoltaic textiles using a fully spray coating processing at low temperature (<150 oC) on a standard 65/35 polyester cotton fabric. The main challenge when spray coating solar cells of less than a few micron thickness is the surface roughness of the polyester cotton fabric which is of the order of 150 µm. We report a maximum optimised efficiency of 2.7% achieved on a glass substrate and 0.02% on woven fabrics, respectively. This approach is suitable for the low cost integration of PV devices into clothing and other decorative textiles.

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*Keywords:* Organic solar cells; textile solar cells; wearable and flexible solar cells

1. Introduction

This paper concerns the development of organic solar cells on flexible fabric substrates. The fabric substrate places many constraints on the fabrication of the devices so existing processes and technologies cannot be applied to textiles. First generation solar cells based on silicon are the market leader in the PV industry.[1] However, first generation cells are rigid, costly and consume high levels of energy in production and are not compatible with textiles. The second generation of thin film based copper indium gallium selenite (CIGS), cadmium tellurium (CdTe) solar cells are attracting more attention due to reduced materials usage, low cost preparation techniques and broad solar coverage, compared to first generation devices. These solar cells have already reached 20% efficiency on rigid substrates.[1, 2] However, the fabrication of second generation solar cells still involves high temperature treatments and vacuum processes which are incompatible with textile substrates. In addition, there is a growing concern about toxicity and after life disposal which is a barrier to commercialization. Third generation solar cells are based on solution processed organic materials which are used to fabricate dye sensitized solar cells (DSSCs), perovskite solar cells and polymer based organic solar cells (OSCs).[3-5] The low cost preparation techniques are making third generation solar cells more attractive in flexible solar cell applications offering an excellent potential for large area power generation. In particular, there is considerable ongoing research in OSCs towards improving device efficiency and fabrication processes.[6-8] These processes and materials do have the potential for the realization of solar cells on fabric substrates.

In recent years, wearable technologies derived from e-textiles have been developed for various applications, for example, medical, sports and military clothing.[9-12] The topic of energy harvesting is concerned with the conversion of ambient energy (e.g. kinetic, thermal or light) into electrical energy for use in powering autonomous systems. There is naturally considerable interest in using energy harvesting in wearable applications, which can extend the life, or potentially replace, standard battery based power supplies. Fabric solar cells are one form of energy harvesting that has great potential for powering wearable devices. However, incorporating solar cells on fabric substrates is not straightforward. Fabrics are highly flexible substrates with different mechanical structures depending upon, for example, the weave and yarn parameters. The surface of a fabric is rough compared to a plastic substrate such as polyimide film (Kapton, trade name of Dupont) and the use of fabric limits the maximum temperature that can be used in device processing. Existing examples of solar cells on fabrics use conventional rigid silicon or plastic solar cells, as standalone PV devices, which are attached (stitched or glued) onto the fabric as a functional patch.[13] This approach makes the fabric relatively inflexible and alters the feel of the textile dramatically and the fabric itself has no added functionality. However, a new generation of flexible OSCs offer the potential for integrating the energy harvesting capability into the fabric itself providing a lightweight solution that maintains the feel of the fabric. Integrating OSCs on fabric substrates has many challenges such as, achieving suitable device flexibility and durability, acceptable conversion efficiency and fabrication using processes compatible with the textile industry.

Research in the fabrication of flexible OSCs integrated into fabrics has explored several approaches. Fabric OSCs were fabricated using a combination of evaporation and spin-coating by   
Bedeloglu *et al.*[14-17] This work actually used a non-woven polypropylene textile tape as the substrate which is not representative of typical wearable woven fabrics. These devices did work and achieved   
0.2 % efficiency which is the highest value reported to date from a coated organic PV textile. Krebs *et al.*[18] used a standard woven textile and smoothed the surface by laminating a polyethylene film prior to OSCs fabrication. This film has a low surface energy and requires a plasma treatment to enable subsequent films to be deposited. These films were deposited by a combination of screen printing and evaporation and did not function due to short circuiting. Another approach by Lee *et al.*[19] fabricated OSCs on a flexible PET/ITO substrate which was then attached to a conductive fabric which acted as the bottom electrode. This approach does not add functionality to the textile itself and uses evaporation processes for some of the films. Other research has explored fabricating a functional organic PV fibre which can then be woven into a textile.[20-23] This approach demonstrated a maximum efficiency of 0.5%, but the method fundamentally limits the output of the solar cell because, once woven into a textile, the PV layer is inevitably partially shaded.

Whilst the organic functional layers in OSCs are deposited using solution-based processes such as spin‐coating, spray-coating, precision-die coating, inkjet printing and dip‐coating, the cathode and anode metal layers have typically been deposited using vacuum based thermal evaporation.[24-28] This was due to the absence of a suitable solution based process for electrodes that give a low work function. Recently, however, several research groups have evaluated silver nanowire (AgNW) solutions for use as flexible electrodes to fabricate OSCs. These have demonstrated a comparable power conversion efficiency to those using indium tin oxide (ITO) and other metal evaporated electrodes.[29-32] Most recently, Guo *et al.* reported solar cells on glass substrates fabricated entirely by solution based processing with AgNW as top and bottom electrodes.[33, 34] The principle of spray-coating is to atomise the dispersion or solution, therefore enabling thin films to be deposited which is essential to achieve functional OSCs. In this work, we have produced fabric based solar cells using a fully spray-coated method to obtain functional photovoltaic textiles that are processed in low temperature conditions (<150 oC) on a standard 65/35 polyester cotton fabric.

1. Materials and Experimental methods
   1. Materials

The approach described in detail below involves using a screen printable polyurethane based interface paste (Fabink-UV-IF1, supplied by Smart Fabric Inks Ltd) to smooth the fabric surface. A metallic AgNW suspension in isopropyl alcohol (IPA), supplied by Nanopyxis, was used to fabricate the electron and hole collecting electrodes. The thin electron transport layer was fabricated with a ZnO-NP dispersion (40 wt% in ethanol) with an average particle size <35nm, supplied by Nanograde. The hole transport layer was fabricated with a PEDOT:PSS dispersion in water HTL/Solar-2), supplied by Heraeus. A blend of poly (3-hexylthiophene) (P3HT): indene-C60 bisadduct (ICBA), dissolved in 1, 2 dichlorobenzene were supplied by Plextronics was used to fabricate the photoactive layer. Patterned ITO glass substrates were supplied by Solaronix. The standard 65/35 polyester cotton fabric was supplied by Klopman International. These materials were used as supplied with no further modifications being required in order to use them in the spray coating process.

* 1. Fabrication of OSCs

The construction of the fabric solar cell begins by screen printing an interface layer onto the fabric substrate. The purpose of the interface layer is to reduce the surface roughness of the fabric and present a smooth layer to support the subsequent spray-coated films. The screen design ensures that the interface layer is only printed where required thereby maintaining the fabric’s flexibility and maximizing breathability when compared to commercial pre-coated fabrics. The printer squeegee pressure setting was set to 6 kg and the printing gap was 0.8 to 1 mm. The film is cured with a UV dose of 1500 mJ/cm2 thereby avoiding a thermal curing process that would release potentially harmful volatile organic compounds. The printed interface layer has a surface free energy of ~35 mN/m which was measured using a Kruss DSA30B tensiometer. This value confirms that the surface promotes the wettability of the majority of solvent based functional electronic inks. Figure 1(a) shows a cross-section of the fabrication process, comprising two deposition stages for the interface and one for each functional layer. Figure 1 (i)-(iv) shows the screen printing of the interface layer on the fabric substrate. As there are typically five functional layers in the solar cell structure, stage (v) to (viii) were repeated 4 more times after the first functional layer deposition to obtain the multilayer spray-coated fabric solar cells shown in figure 1 (b). Figure 1 (c) shows the plan view of the spray coated solar cells on the fabric with 8 pixels being fabricated in one device. Figure 1(d) shows the plan rear view of the fabric solar cell which demonstrates that the addition of the interface and the spray coated solar cells does not change the feel and appearance of the underside of the fabric. All the spray coating steps were performed in a nitrogen atmosphere glovebox with a differential pressure inlet/outlet of 0.3 bar and dried on a hotplate inside the glovebox. The first functional layer of the fabric solar cell is the spray-coated bottom AgNW electrode which is more flexible than evaporated thin metal layers in which micro cracks can occur as a result of bending.[35] The spray-coated AgNW layer was dried at 130 °C for 5 minutes on a hotplate to obtain an AgNW film with a thickness of ~100 nm. The ZnO-NP dispersion was successively spray-coated on top of the AgNW bottom electrode and dried at 60°C for 10 minutes on a hotplate to obtain a solidified layer. Next, the PV layer of P3HT: ICBA was spray-coated onto the top of the ZnO-NP layer. The deposited layers were subsequently dried on a hotplate for 10 minutes. The spray distance is 20 cm for the AgNW, P3HT:ICBA and PEDOT:PSS layers and 30 cm for the ZnO-NP layer.

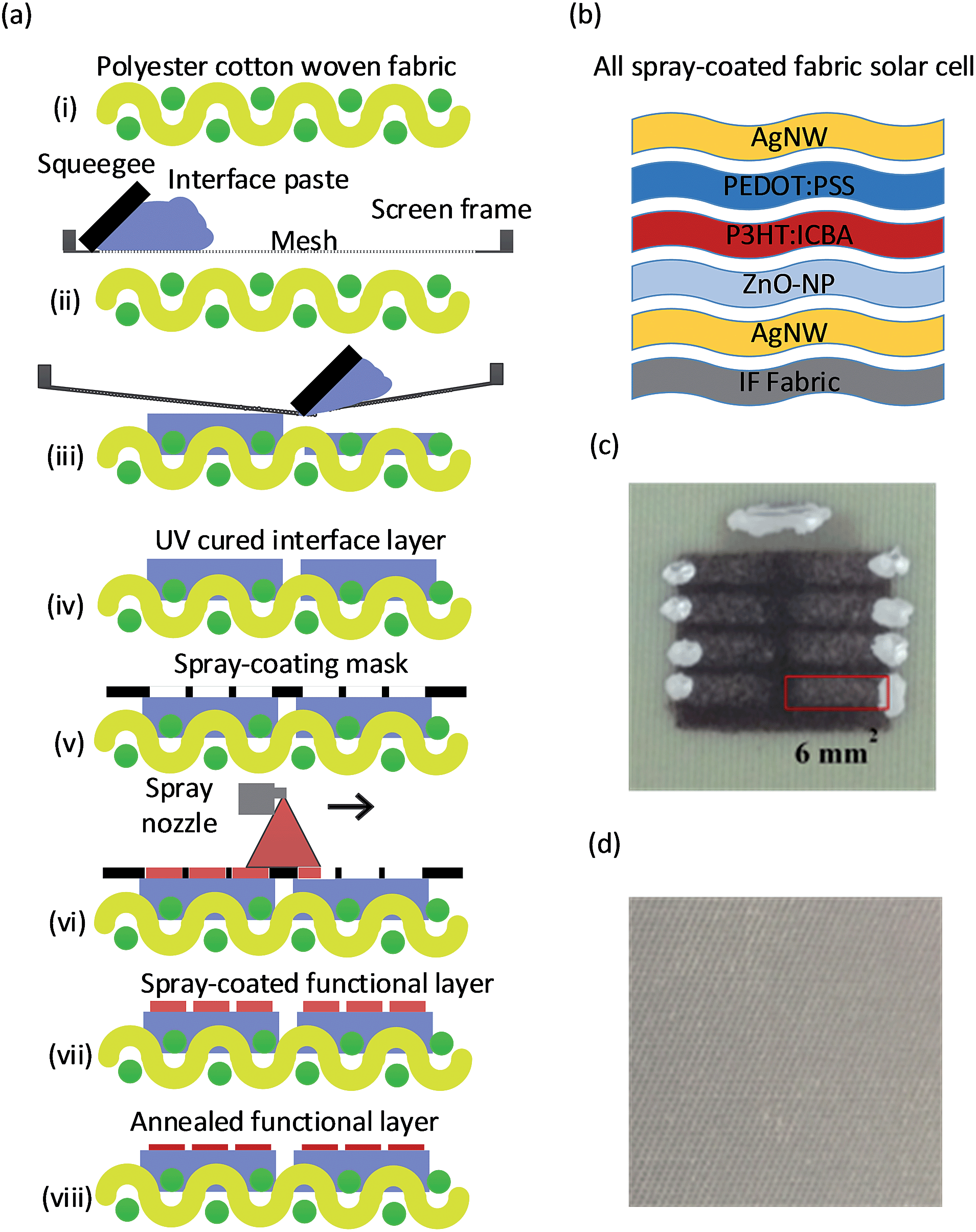


Figure 1: (a) Cross-sectional view of the fabrication process of spray-coated fabric solar cells; (b) Device structure of a fully solution-processed spray-coated fabric substrate; (c) The plan view of an optimised fabric solar cell; (d) The plan rear view of the fabric solar cell.

Then, the PEDOT:PSS hole transport layer was spray-coated and baked at 100°C for 5 minutes. To complete the device fabrication, a final semi-transparent AgNW electrode was spray-coated on top of the PEDOT:PSS layer. During processing, the fabric substrates were glued to an alumina tile that supported the fabric to maintain flatness for each subsequent functional layer deposition. The performance of the fabric solar cells was tested after peeling it off the alumina tile. The peel off angle to the alumina tile is about 60 degrees. In addition, pre-heating the alumina tile to ~50 oC on a hotplate facilitated the peel-off process, avoiding potential damage that might be caused by strain. The standalone fabric solar cells showed good flexibility after peeling off. For the purposes of comparison, we also fabricated OSCs by spray-coating onto glass substrates using the same parameters. There were 48 devices made for each device type and each device was measured in an ambient atmosphere immediately after fabrication. However, we only report the best performing cell in terms of power conversion efficiency (PCE) for each device type. The measurement results of the other devices show relatively low conversion efficiencies of up to 1 to 2 orders of magnitude lower, compared to the best performing device. The differences are due to inconsistent processing and uneven film coverage.

* 1. Characterization and measurement

The current density versus voltage (J/V) curves of photovoltaic devices were obtained using a Keithley 2400 source meter unit. The photocurrent was measured under AM 1.5 (100mW/cm2) irradiation using an ABET solar simulator, calibrated with a standard Si solar cell. The effective area of each cell is 3 mm2 and was defined by the shadow mask. The efficiency of standard solar cells was calculated by the equation:

where VOC is the open-circuit voltage, ISC is the short circuit current, FF is the fill factor, Pin is the power of the incident light and ηmax is the PCE. The surface morphology of the AgNW and fabric solar cells was examined by field emission scanning electron microscopy (FESEM) analysis using a JEOL JSM 7500F instrument.

1. Results and discussion



Figure 2. (a) Cross-sectional view of woven 65/35 polyester cotton fabric substrate; (b) Interface coated fabric substrate; (c) FE-SEM image of spray-coated bottom AgNW electrode on fabric; (d) cross-sectional SEM image showing the spray-coated layer sequence on the fabric substrate.

Figure 2 (a) shows a cross-sectional view of the woven 65/35 polyester cotton fabric structure with interlacing warp and weft yarns, illustrating the rough surface profile of the material. Figure 2 (b) shows the fabric after printing three layers of the interface with two depositions in each. This is required to obtain a sufficiently smooth interface surface with an average thickness of 150 µm. Figure 2 (c) presents an SEM image of the spray-coated AgNW electrode viewed from above on the fabric. The nature of the randomly dispersed AgNW forms overlapping wires, each a few tens of microns in length and a few tens of nanometres in diameter. As shown in figure 2 (d), subsequent deposition of the ZnO-NP layer reduces the surface roughness of the AgNW due to the ZnO-NP filling up the scaffold structure of the AgNW. Figure 2 (d) clearly shows that the flattened AgNW is covered by the ZnO-NP and successfully coated by the spray-deposition of a P3HT:ICBA active layer. The P3HT:ICBA blend used in this study has achieved the optimised thickness of around 200 nm.

The J/V measurements of the OSCs fabricated in this work are shown in figure 3 and the results are summarised in table 1. It was initially found, however, that the thin ZnO-NP and P3HT:ICBA films fill the scaffold structure of the bottom electrode AgNW but fail to sufficiently separate the top and bottom electrodes which leads to a short circuit. In order to avoid a short circuit, the bottom AgNW layer was first flattened by compressing the nanowires while annealing the fabric devices at 150oC for 15 minutes in an oven. The additional layers were then spray-coated subsequently to complete the device fabrication. This approach successfully prevented the short circuit. Device type 1 was fabricated on the fabric and gave a maximum PCE of 0.02% with a FF of 0.25, VOC of 0.41 V and JSC of 0.26 mA/cm2, as shown in table 1. For comparison, device type 2 was spray-coated with the same functional layers on a glass substrate, which gave a maximum PCE of 2.7% with a FF of 0.23, VOC of 0.40 V and JSC of 28.3 mA/cm2. As displayed in figure 3, the J/V curve of device type 2 indicates a higher rectification, which suggests better diode behaviour due to the smoother surface of the active layer and the uniform coverage of the PEDOT:PSS layer. VOC and FF values of the spray-coated OSCs on both fabric and glass substrates are nearly identical, but the JSC is lower for the fabric OSCs. This may be attributed to the bending of the fabric that happens during peeling-off from the alumina tiles after fabrication. Bending caused by peeling may generate micro-sized cracks on the functional layers which will increase the resistance across the junction to reduce the JSC current. Thus the fabric device (type 1) leads to a lower PCE compared to the glass counterpart device (type 2).

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| --- | --- | --- | --- | --- | --- | --- |
| **Devices** | **Device configuration** | **VOC (V)** | **FF** | **JSC (mA/cm2)** | **PCE (%)** | **Area** |
| Type 1 | Fabric/ Interface /Pressed AgNW/ZnO-NP/P3HT: ICBA/PEDOT:PSS/AgNW | 0.41 | 0.25 | 0.26 | 0.02 | 6mm2 |
| Type 2 | Glass/pressed AgNW/ZnO-NP/P3HT: ICBA/PEDOT:PSS/AgNW | 0.40 | 0.23 | 28.3 | 2.7 | 3mm2 |

Table 1. Summary of the spray-coated solar cell characteristics on both fabric and glass substrates.

Figure 3: J/V characteristics of OSCs fabricated on (a) fabric and (b) glass substrate using the spray-coating method, which represent device type 1 and type 2, respectively.

**Conclusions**

In summary, fully spray-coated fabric solar cells on standard polyester cotton fabrics have been demonstrated. The standard polyester cotton fabric was pre-treated with a screen printed interface layer to significantly reduce surface roughness and obtain a compatible wettability for the subsequent deposition of functional inks. The results gave a PCE of 0.02% for all the solution-processed spray-coated fabric solar cells and 2.7% for the solar cells on the glass substrate. Compressing the bottom AgNW layer during the annealing stage prevents short circuits, whilst reducing the thickness of the ZnO-NP layer in the optimised device also improved device performance by reducing the resistance. An optimised solution may be used to manufacture energy harvesting textiles to integrate into, and supply, the power source for wearable electronics systems.

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