RESEARCH ARTICLE

Fully printed organic solar modules with bottom and top silver nanowire electrodes

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Abstract
One of the advantages of organic photovoltaics (OPV) over other contemporary technologies is its relative ease of processing. There are, however, very few works that have realized fully printed devices, including the bottom electrode, let alone with a scalable process in a reasonable device size (>1 cm²). In this work, design steps and optimization processes towards fully printed OPV modules with scalable processes are demonstrated for the first time. An overview on issues related to upscaling with printed electrodes is first provided. The various issues are then addressed by a rational design process supported by measurements and calculations. Finally, a set of fully printed OPV modules are fabricated using these optimized parameters that have over 3.5-cm² active area with 5% efficiency. For the first time, this work has also demonstrated the process compatibility of fully printed device structures with non-fullerene acceptor systems, which enables more design opportunities for the current generation of high-performance OPV materials.

KEYWORDS
modularization, organic photovoltaics, printed electronics, silver nanowire, upscaling

1 INTRODUCTION

Solution processed thin film opto-electronic devices are particularly attractive for photovoltaic applications due to the high throughput and potential cost savings associated with roll-to-roll manufacturing. In the case of thin film solar cells, tremendous effort has been put into demonstrating that high power conversion efficiencies can be achieved for small lab scale devices (active area <~1 cm²).1,2 However, the vast majority of reports considers only partially solution processable architectures, that is, one or several of the interlayers and electrodes are most often processed by physical vapour deposition, for example, thermal evaporation, e-beam evaporation, ALD and sputtering, among others.3-5 These architectures utilize substrates that are precoated and prepatterned with indium-tin-oxide (ITO) or ITO/Ag/ITO (IMI) sandwich structures, a process that relies on energy intensive sputtering process and requires indium, which is listed as a rare element.6 Thus, it becomes necessary to either rely on external providers or to set up both, solution process and vacuum deposition, in a single production line, which unavoidably raises the cost of production. In contrast, printing bottom electrodes allows for all layers to be deposited by solution processable roll-to-roll printing method, which will greatly simplify the production process.

In the case of organic photovoltaics (OPV) and thin film printed photovoltaic technologies in general, the production costs are a key criterion that determines market competitiveness. Given that transparent conductors usually constitute a large part of the bill of materials (BOM), replacing materials processed via costly physical vapour techniques with potentially inexpensive solution processed
alternatives will constitute an important cost advantage.\textsuperscript{7,8} Besides the inherent economic benefit of solution processing, the possibility of depositing all layers of a device from the liquid phase, including both electrodes, will allow devices to be printed also on discretionary, untreated surfaces in an in-line production process, precluding the need for transferring the devices between different deposition and/or patterning equipment.

Fully printed OPV cells have already been demonstrated by various works in literature.\textsuperscript{9–14} The printed electrodes, at least one of which must be semitransparent in order to allow light to pass through, are realized by various materials, methods and technologies. A contemporary solution to this challenge is to print a combination of opaque silver grid and transparent poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) film.\textsuperscript{15,16} This method however requires two processing steps and can cause short circuits.\textsuperscript{17} Furthermore, the optical disturbance that is introduced by the grids also affects the appearance of the devices. While there are already many alternatives proposed to address these shortcomings,\textsuperscript{18,19} one of the most promising technology is based on mesh network electrodes formed by highly conductive nanowire dispersion, such as silver (AgNW).\textsuperscript{9,12,14,20,21} There are already numerous works published by us or other groups on the optimization of this type of electrode for organic solar cell application but most are still limited to smaller device areas.\textsuperscript{3,9,12–14,22}

Besides the well-reported surface roughness issue associated with AgNW electrodes,\textsuperscript{11,21,23,24} one apparent problem as device area increases is the increase in resistive losses associated with the electrodes. This necessitates the patterning of a single large cell into a number of smaller but interconnected cells, that is, a solar module.\textsuperscript{25,26} An organic solar module is fabricated by patterning a large solar cell into smaller stripes and then serially connect them monolithically. An illustration of the interconnection scheme is shown in Figure 1. Three distinct patterning steps are introduced during the fabrication process, namely, the P1, P2 and P3 steps. The first step, P1, separates the bottom electrode into discontinuous conductive strips. The P2 step is performed after PEDOT:PSS layer deposition. This creates a gap in all the interlayers to allow the top electrode to connect to the bottom electrode during the deposition process. Finally, the P3 step separates the top electrode into discontinuous conductive strips so that individual cells are finally connected in series.

Despite all the progress in small-scale fully printed solar cells, fully printed organic solar modules with AgNW electrodes have yet to be demonstrated. In comparison, vacuum-deposited ITO or ITO/Metal/ITO bottom electrode and printed AgNW top electrode combinations have successfully been utilized to fabricate solar modules with high power conversion efficiencies (PCEs) and high geometric fill factor (GFF).\textsuperscript{27–30} Few reports on fully printed organic solar modules utilize exclusively silver grid/PEDOT:PSS composite electrodes, which have various aforementioned issues.\textsuperscript{34–36} In this paper, we will first identify the challenges to realize fully printed organic solar modules, which will be addressed through meticulous optimization of the AgNW/AgNW interconnection and cell structure through a combination of roll-to-roll (R2R) compatible solution processing and R2R compatible high-resolution, nanosecond pulsed LASER patterning. This work will then, for active layer systems frequently used in large scale production, demonstrate fully solution processed semitransparent modules with power conversion efficiencies (PCE) of up to 5% in an ambient environment, with limited upscaling losses compared with more conventional electrodes.
FULLY PRINTED OPV CELLS

In order to realize fully printed OPV modules, the cell architecture has to be optimized first. The first issue with AgNW as bottom electrode lies in its relatively rough surface morphology that causes devices to shunt, as reported by other works. Similar to Morgenstern et al., the AgNW electrode is planarized by the zinc oxide nanoparticles (ZnO), which form the electron extraction layer. However, this layer thickness must be minimized to reduce the electrical resistance and optical absorption, and hence the negative impact on fill factor (FF) and short circuit current density ($j_{sc}$), respectively. In order to optimize the ZnO layer thickness with respect to these two requirements, ZnO layers with different thicknesses were coated on top of AgNW films of sheet resistances of roughly 12 $\Omega/\square$. Figure 2A–D shows the respective SEM images for AgNW electrodes coated with ZnO layers of different thicknesses. It becomes obvious from these images that ZnO layers of at least around 100 nm in thickness are needed for complete coverage of the AgNW layer.

Standard inverted structure cells are subsequently fabricated on top of the planarized AgNW films. P3HT:ol-DTBR was chosen as the active layer for this investigation with film thicknesses referenced from previous works. The top AgNW electrode has a sheet resistance of roughly 6 $\Omega/\square$. The transmittance data of all these electrodes are shown in Figure S2. ITO based devices are also included for comparison. Figure 3 shows the current–voltage characteristics for cells with different ZnO layer thicknesses, whereas Figure 4 lists their respective performance parameters. All semitransparent devices have lower $j_{sc}$ and FF due to the low reflectance and conductivity of AgNW compared with evaporated silver (EvapAg). The result confirms what has already been suggested by the SEM images shown in Figure 2, that is, that AgNW bottom electrodes without ZnO layers of sufficient thickness are in direct contact with the active layer. This results in shorts between both electrodes, which lowers the open circuit voltage ($V_{oc}$), as also illustrated by other works. AgNW electrodes with ZnO layer thickness above 100 nm perform similarly to those with ITO bottom electrodes (Figure 4). However, such a thick planarization layer has also slightly reduced the short circuit current density ($j_{sc}$). This is due to the absorption by the thick ZnO layer, which reduces the transmission of light into the photoactive layer. Overall, the transition from ITO to AgNW as bottom electrode with ZnO planarization has incurred on average about 10% loss in power conversion efficiencies (PCE), resulting in fully solution processed semitransparent devices with efficiencies of 4.6%. This is an acceptable tradeoff considering the substantial changes to the device structure needed.

FROM CELLS TO MODULES—ISSUES WITH AgNW/AgNW INTERCONNECTION

Despite the successful demonstration of OPV cells, the path towards modularization, however, is far from straightforward when compared with some of our previous works. Unlike other film electrodes

FIGURE 2  SEM images of AgNW electrodes without filler before and after coating ZnO layers of different thicknesses on top. (A) Bare AgNW film; (B) 40-nm ZnO; (C) 80-nm ZnO; (D) 120-nm ZnO
such as ITO or PEDOT:PSS, AgNW electrodes have only limited surface area coverage. Thus, it is geometrically more challenging to make highly conductive interconnections between two AgNW networks when the contact area (the P2 opening gap in Figure 1) is very limited. Initial attempts to fabricate fully printed organic solar modules using LASER patterning parameters and interconnection dimensions (total fluency 0.923 J/cm² at 70 μm width) from previous works have resulted in non-connected modules or modules with very low fill factors. When comparing the slope and the shape of the JV curve between the modules in Figure 5A to their cell counterparts in Figure 3, it is clear that individual cells are connected via highly resistive interconnections. Thus, one of the challenges on fabricating fully printed organic solar modules lies on the critical pulsed LASER ablation process to establish such an efficient interconnection. However, the dark field microscopy image after the P2 LASER patterning step in Figure 5B indicates that the bottom AgNW electrode is severely damaged, which has disconnected individual cells in the module. This also reveals that the LASER ablation threshold of AgNW bottom electrodes is significantly lower than that of vacuum-deposited materials, such as ITO. Further attempts with lower total fluency at 0.626 J/cm² and much wider interconnection still result in non-connected modules. The coloured AgNWs in Figure 5C indicate the interconnections are still covered by the stacked layers that are printed on top of the AgNW electrode (see Figure 1). This has drastically increased the interconnection resistance, which severely limits the FF and even Voc of the module as shown in Figure 5A for both cases. The series resistance of the interconnection network, which separately limits the FF and Voc, is very high.

In the previous section, thicker ZnO layers have been deposited.

### 3.1 LASER power optimization

A test is first performed to determine the upper limit of the total patterning LASER energy per unit area (total fluency) before the electrode is damaged. Using a nanosecond pulse LASER, patterning lines are drawn perpendicularly across layered stacks as depicted in Figure 6 and the change in electrical resistance across the bottom AgNWs of the electrodes is measured. Two different layer stacks are tested in Figure 6, and the change in electrical resistance across the bottom AgNWs of the electrodes is measured. Upon completion of these LASER patterning steps, all the AgNWs are cut into discrete stripes at the bottom electrode, similar to the top electrode before the LASER patterning step in the dark field microscopy image after the P2 LASER patterning step in the module. The top electrode with its conductive individual cells in the module is also patterned with this disconnection individual cells in the module. This also reveals that the LASER ablation threshold of AgNW bottom electrodes is significantly lower than that of vacuum-deposited materials, such as ITO.

In the previous section, thicker ZnO layers have been deposited. However, the impact of ZnO thickness variation on the LASER energy intensity has not been investigated. The P1 step (Figure 6A) should therefore be repeated. The current vs. voltage (JV) curves of organic solar cells (bottom electrode/ZnO/P3HT:oIDTBR/PEDOT:PSS/AgNW) with AgNW (solid line) and ITO (dotted line) bottom electrodes covered by ZnO layers of different thicknesses, measured under 1 sun, illuminated through the bottom electrode, and in dark (bottom curve). Evaporated silver as top electrode is also included for reference (thin grey line). The detailed layout of the cells structure is provided in Figure S3.
However, the total fluencies chosen should be the minimum applicable energy to minimize ablation damage to the substrate. It is clear from the data that the total fluencies required to cut open the AgNW electrode are significantly higher for electrodes with thicker ZnO layers, which could be an issue for less resilient substrates, such as PET. The ZnO blended AgNW electrode, however, provides a critical processing advantage over bare AgNW electrode. Most printed electrodes such as AgNW cannot be washed or ultrasonicated after P1 patterning step to remove conductive debris, unlike ITO electrodes. One of the ways to reduce these conductive debris from being generated is by intermixing the AgNW electrode with another non-conductive layer, which is the ZnO layer in this case.

The second LASER patterning step, P2 (Figure 6B) is responsible for the removal of multiple stacked layers that are above the AgNW electrode without damaging it. The interconnection between cells is then established by the coincident AgNWs between the two electrodes after the deposition of the top electrode. Thus, this patterning step should ensure the electrical resistance of the interconnection be low enough so that it does not hamper the module performance. This means the total fluencies applied should be as high as possible.
without significantly increasing the bottom AgNW electrode resistance. Figure 7B shows the change in the resistance of the electrode with different total fluencies applied during this step. Similar to the P1 step, ZnO layers of different thicknesses are provided for comparison. While the shaded area highlights all total fluencies in which the electrical resistance remains unchanged after patterning, the LASER parameter chosen should be very close to the damage threshold in order to achieve lowest possible interconnection resistance \( (R_c) \). This is proven in the previous section when low total fluency does not manage to remove all the interlayers or high total fluency causes too much damage to the AgNW electrode (Figure 5). Their respective values are also indicated in Figure 7B.

Beside process optimization, a few findings can also be extracted from the data. First, the total fluencies required to cut open the AgNW network are significantly higher for the P1 step than for the P2 step by more than two orders of magnitude (>100 J/cm² for P1 vs. <1 J/cm² for P2). This is due to the higher photoabsorption coefficient of the active layer compared with ZnO and AgNW at the LASER wavelength. For P1, most of the energy is transmitted through the ZnO/AgNW film. For P2, however, most of the energy is absorbed by the active layer. Since the heat diffusion length of the heat zone created by a nanosecond LASER in the organic material is in the range of tens of nanometers, some of the thermal energy will unavoidably transfer to the ZnO and thereafter the AgNW layer. This causes an induced ablation of the ZnO:AgNW mixture underneath, which is similar to observations from other works in other photovoltaic systems. This indicates the bottom AgNW electrode is more readily damaged during the P2 step when compared with the P1 step. This issue is somewhat alleviated by the thick ZnO layer that fully covers the AgNW surface (0.5 J/cm² for 40-nm ZnO vs. 0.8 J/cm² for 120-nm ZnO), which behaves similarly in the P1 step. In fact, the total fluencies required to cut open the AgNW network are always higher for thicker ZnO layers, regardless of the patterning step (Figure 7A,B). Given the stark contrast in ZnO coverage between the two AgNW

**FIGURE 5** Characteristics of fully printed OPV modules with excessive or insufficient LASER ablation; (A) current voltage characteristics of fully printed OPV modules with highly resistive interconnections of 70 μm and 170 μm in width. The modules consist of 4 cells with total area of 3.512 cm². The detailed layout of the module is given in Figure S5; (B,C) top down dark field images of the bottom AgNW electrode after P2 patterning; (B) 0.923 J/cm². This image shows that most, if not all AgNWs in the patterning area are melted into particles; (C) 0.626 J/cm². This image shows that most, if not all AgNW still exist. However, the brownish colour of the AgNWs indicates the stacked layer above is not completely removed

**FIGURE 6** Measurement device structure for determining the total laser fluency limit: (A) for patterning the AgNW bottom electrode, that is, the P1 step; (B) for creating an opening for the connection between both electrodes, that is, the P2 step
electrodes shown in Figure 2, it is reasonable to conclude the ablation threshold of the AgNW electrode is indeed enhanced by such coverage.

While the experiments in Figure 6B have indicated the LASER intensity threshold for the P2 patterning step, the critical relation between the interconnection resistance and the total fluency is still missing. In our previous work, we have devised a measurement structure, the cross Kelvin bridge resistor (CKBR), to measure such interconnection resistances of a thin film solar module.31 In this work, we have fabricated an array of CKBRs to measure the effect of total fluencies on the interconnection resistance ($R_c$). The layout of the measurement structure, together with the calculation of $R_c$ and of the specific interconnection resistances $p_c$ are provided in Figure S4. The interconnection resistances resulting from P2 ablation using different laser fluencies are tabulated in Table 1 with their respective openings before top electrode deposition shown in Figure 8. Within all conditions tested, the wires are still clearly visible after LASER patterning. Samples that have sustained higher intensity irradiation appear to have more distorted and fused wires when compared with samples irradiated with pulses of lower energy. This indicates that all LASER intensities tested are indeed high enough to remove the active layer and the ETL but still low enough not to remove the AgNWs at the bottom. However, even when all total fluencies tested are close to but within the threshold limit discussed earlier, $p_c$ and $R_c$ still show a significant variation across the range tested. This indicates that there is only a narrow process window for P2 ablation, which is limited by insufficient removal of active layer and ETL on one hand (Figure 8A) and serious damage to the AgNW bottom electrode on the other hand (Figure 8C). The residues at insufficient LASER power consist mainly of ZnO. Nonetheless, an optimal total fluency can still be found at 0.8 J/cm² with roughly $R_c = 2 \Omega$. Thus, total fluencies of 3000 and 0.8 J/cm² are selected for the P1 and P2 steps, respectively, which translates to the experimental parameters given in the experimental section.

### 3.2 Interconnection structure optimization

While the $p_c$ obtained is well within the acceptable range for OPV module fabrication, $R_c$ is still relatively large for the module geometry similar to the ones used in this work. As a general rule of thumb, in order to ensure minimal resistive losses at the interconnection, the $R_c$

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**FIGURE 7** Plots of resistance change of the AgNW bottom electrode after LASER patterning for the measurement structure in (A) Figure 6A, which corresponds to the P1 patterning step and in (B) Figure 6A, which corresponds to the P2 patterning step. Two different ZnO layer thicknesses were tested, which are represented by two different colours. The shaded areas represent the total fluencies which can be utilized for the particular patterning step. The horizontal lines at 200 and 50 Ω represent the thresholds in which the AgNW networks are considered to be cut open in steps P1 and P2, respectively

| Total Fluency [J/cm²] | Average $R_c$ (Ω) | $p_c$ (Ω * cm²) | Average $R_c$ (Ω) | $p_c$ (Ω * cm²) |
|-----------------------|--------------------|-----------------|--------------------|-----------------|
| 0.74                  | 12.60              | $23 \times 10^{-03}$ | 17.8               | $3.26 \times 10^{-03}$ |
| 0.80                  | 35.75              | $69 \times 10^{-03}$ |

**TABLE 1** $R_c$ and $p_c$ of CBKRs with different LASER power

Note: Each resistor has an interconnection length of 2.65 mm and a width of roughly 70 μm (taken from Figure 7). Bottom electrode sheet resistance: 12 Ω/□; top electrode sheet resistance: 6 Ω/□. The test current per unit length is 38 mA/cm.
of an OPV module should be smaller than 0.7 \( \Omega \) for the CKBR geometry in test, as calculated in our previous work.\(^3\) The AgNW electrode is a conductive wire mesh network with only limited surface area coverage. This limits the effective interconnection area between the bottom and top electrode to the number of coincident wires within the area. Thus, the interconnection area must be enlarged accordingly to ensure that an acceptable number of wire connections is formed, thereby reducing the resistive loss of the interconnection. An additional advantage with increased contact area lies on the reduced current density at the interconnection. This minimizes the chance of damaging the interconnection when a limited number of coincident nanowires are conveying electrical current.

In this work, a number of CKBR arrays are fabricated to investigate and optimize the interconnection width. Similar to Figure 8, the silver nanowires are still clearly visible after LASER patterning with slight distortion. The SEM image further confirms the existence and continuity of the AgNW network across the opening produced by laser ablation. For determining the interconnection resistances, two different current densities are applied to the resistors for 32 s and the potential difference is measured every 50 ms to also analyse the stability of the interconnection. The widths of the openings after the P2 steps are first measured in Figure 9. Table 2 then shows the relation of \( R_c \) and \( p_c \) with different applied current and different interconnection widths in Figure 9.

All images in Figure 9 have indicated the presence of debris along the edges of the interconnection line. This is due to the Gaussian energy distribution in a LASER spot.\(^{27,28}\) The higher intensity at the centre causes the wires to distort slightly while the edges are still partially covered. Undoubtedly, this has reduced the effective interconnection area and thus increased \( p_c \). The partially covered areas are roughly the same in size for all samples. Thus, \( p_c \) would be reduced by roughly 10% for the wider interconnections but by more than 30% for the 70 \( \mu \)m wide interconnection if they were not considered.

The result also reveals \( p_c \) decreases as the interconnection width increases. This can be attributed to the increasing number of coincident AgNWs as the interconnection width increases. The reduction in \( p_c \), however, is not proportional to the interconnection area. It experiences the greatest reduction from 70 to 100 \( \mu \)m and starts to level off at 140 \( \mu \)m. The current is not uniformly distributed across the interconnection width but it rather depends on the localized \( p_c \) as well as the distance from the edge of P1, that is, the current is crowded at the edge of the interconnection.\(^4\) This puts an upper limit on the utilization of the interconnection width. This effect is best visualized in Figure 10. The distance over which most of the current transfer occurs can be considered as the characteristic transfer length \( L_T \) of the contact and the ratio between interconnection resistance and sheet resistance of the electrode determines this distance. Unlike evaporated metallic electrodes in which the contact properties were widely studied, AgNW electrodes have much larger sheet resistance, which significantly reduces the current crowding and increases the transfer length to tens of micrometre already. Apparently though, with such a wide interconnection width (>100 \( \mu \)m) the area closest from the P3 line is still not being utilized for current transfer.

As evident from Table 1, \( p_c \) also reduces as the current per unit width of the interconnection increases. Joule heating of the AgNW network increases the resistance of the wires, but this effect is overcompensated by the welding of the nanowire junctions.\(^{43-46}\) However, as indicated in other works, the sustained application of high current through the interconnection will result in damage to AgNW junctions and the eventual destruction of the network.

In order to quantify the effects described above, the changes in potential difference in seconds across the AgNW/AgNW interconnection under a constant applied current were monitored in Figure 11. Two levels of current are tested in here. At 38 mA/cm\(^2\) (Figure 11A), it corresponds to the current applied to the interconnection when a solar module is at its short circuit condition, while at 377 mA/cm\(^2\)
Thus, the interconnection must remain stable permanently under the loading condition postulated in Figure 11A while it must endure, at least for a brief period of time, the condition postulated in Figure 11B. The results are qualitatively similar to what has been presented in other works.46 A brief summary of their findings are provided here. When high current density is applied, the thermal stress will weld individual wires together due to the higher junction resistance than individual wires. This reduces the overall resistance of the network. However, when such stress is too high, nanowires will be fused into silver nanoparticles, destroying the junction and wires altogether.46–48 This explains the hikes in resistance. These two effects counteract each other continuously and result in the jumps in Figure 11B.

At 377 mA/cm, the 70-μm wide interconnection is destroyed in less than 2 s due to thermal stress, whereas the wider interconnections show stepwise reduction of potential difference, indicating a reduction of $R_c$ and $p_c$. At 38 mA/cm, the 70-μm wide interconnection indicates a slight decrease in $R_c$ and $p_c$ after about 30 s, while the two wider interconnections do not show any changes. These results indicate that interconnection widths of 100 and 140 μm are feasible candidates for AgNW/AgNW interconnections. As the enhancement at 140 μm width when compared with 100 μm width is marginal and both are within the $R_c$ of 0.7 Ω prescribed, the interconnection width of 100 μm is thus selected for its higher geometrical fill factor (GFF; module layout is given in Figure S5). GFF is the ratio between the active area producing photocurrent and the total area in a solar module (see Figure 1). While a high GFF module designs minimize the loss due to inactive area, it also amplifies the current density at the interconnection region by a factor of active area over the contact area. The average current density $J_i$ at interconnections of different widths is calculated as

\[
J_i = \frac{I}{A_{active}}
\]
Thus, $J_I$ of the module layout in Figure S5 that has an active cell width of 5 mm with the maximum current density in Figure 4 is 0.54 and 0.77 A/cm² for interconnection widths of 100 and 70 μm, respectively. Thus, the power density of the interconnection region is calculated as 82 mW/cm² and 1.05 W/cm² using the $R_c$ values from Table 2 (at 38 mA/cm). The stark difference in power density is contributed by both higher $R_c$ and narrower interconnections, and accounts for the huge difference on their overall electrical stability.
3.3 Impact of AgNW formulation on interconnection resistance

There are numerous AgNW formulations from the literature and in the commercial market. The scope of the discussion thus far has been limited to AgNW formulations without additives. However, there are also AgNW inks that contain fillers to enhance their adhesion to substrate surfaces, as revealed by our previous works.9,12,27 While these formulations have successfully been used to fabricate top transparent electrodes for organic solar cells and modules, there are currently no studies on their suitability as bottom electrodes of OPV modules.9,27 One potential issue with the additional filler lies on the formation of an insulating layer on top of the nanowire network after drying, which prevents direct contact with individual wires after P2 patterning, especially when the interconnection width is very limited. However, there are currently no investigations on the impact of these filler materials on the $R_c$ and $p_c$ of the interconnection. This work will provide an insight into such issues to determine if AgNWs films with fillers can be used as bottom electrodes for fully printed organic solar modules.

The presence of such fillers on the AgNW network is first confirmed by the SEM images in Figure S1a-c, which also compares the surface morphology before and after coating ZnO layers at different thicknesses. The filler within the AgNW film is clearly visible when Figure S1a is compared with Figure S1c. The reduced contrast between wires as well as the appearance of bubbles Figure S1a are good indications that the AgNWs are at least partially submerged in the filler matrix. This is best illustrated in Figure S1b and Figure 2B. Notice that the AgNWs are well intermixed and in more direct contact with the ZnO nanoparticles for AgNW films without fillers compared with those with fillers.

The AgNW electrodes with fillers are then used to fabricate the same CBKR structures that were presented before. The results presented in Table 3 indicate that both $R_c$ and $p_c$ are orders of magnitude higher than the results for the AgNW films without fillers presented in Table 2. The insulating fillers, as can be observed by the blueish colour

| Interconnection width | 70 μm | 140 μm | 200 μm |
|-----------------------|-------|--------|--------|
| Current per unit length (mA/cm) | Average $R_c$ (Ω) | $p_c$ (Ω * cm$^2$) | Average $R_c$ (Ω) | $p_c$ (Ω * cm$^2$) | Average $R_c$ (Ω) | $p_c$ (Ω * cm$^2$) |
| 38 | 261.25 | 1.37 | 232.50 | 0.92 | 202.50 | 0.39 |
| 377 | 97.50 | 0.51 | 88.50 | 0.35 | 68.33 | 0.13 |

Note: Each resistor has an interconnection length of 2.65 mm. Bottom AgNW electrodes are prepared from inks with filler polymer. Bottom electrode sheet resistance: 12 Ω/□; top electrode sheet resistance: 6 Ω/□.

The performance parameters for the fully printed OPV modules are listed in Table 4. The results indicate that the use of AgNW films with fillers results in lower efficiency compared to those without fillers.

### Table 4 Performance parameters for the fully printed OPV modules

| Material          | $j_{sc}$ per cell (mA/cm$^2$) | $V_{oc}$ (V) | $FF$ | Injection @ 4.8 V (mA/cm$^2$) | PCE (%) | Transmission at 550 nm (%) |
|-------------------|-------------------------------|--------------|------|-------------------------------|---------|---------------------------|
| P3HT:OIDTBR       | 9.27                          | 2.79         | 0.70 | 0.58                          | 57.12   | 1.75                      |
| PV2000:PCBM       | 13.27                         | 2.93         | 0.73 | 0.50                          | 43.43   | 6.25                      |

Note: Active module area is 3.512 cm$^2$. Note: PCE and $j_{sc}$ are calculated from active device area. Overall transmittance is measured at 550 nm for an encapsulated fully printed organic solar module.
around the AgNWs in Figure 12 as compared with Figure 9, are indeed hampering the contact between both electrodes. Given the high current density at the interconnection, such a high $R_c$ will cause too much potential drop and render the OPV module unusable as described in Section 3.2.

### 4 | FULLY PRINTED OPV MODULE

To demonstrate the processability of the fully printed semitransparent solar modules with AgNW/AgNW interconnections, two types of OPV modules were fabricated by a combination of the process parameters developed in the last two sections. In order to demonstrate the industrial relevance of our approach, two active layer systems were chosen which are used in manufacturing large area OPV modules. Besides P3HT:oIDTBR, PV2000:PCBM that has demonstrated high performance in some of our previous works, was also utilized as photoactive layer for module fabrication.\(^{22,49,50}\) The characteristics of PV2000:PCBM based devices of 0.1 cm\(^2\) cell area are provided in Figure S6 and Table S1. The same cell structure and the same LASER patterning parameters as for P3HT:oIDTBR were employed for the PV2000:PCBM system. This proves the general applicability of the cell architecture and the processing method to both fullerene and nonfullerene-based systems.

The performance data are tabulated in Table 4 with their respective current voltage characteristics shown in Figure 13. The modularization of both material systems has incurred roughly 0.7% efficiency loss (absolute) with respect to the test cells, which is on par with other conventional electrode combinations we have demonstrated in the past.\(^{26,29}\) Despite the additional optical losses due to the thicker ZnO layer, both modules have demonstrated reasonable transparency compared with the previous ITO/IMI based OPV modules.

**CONCLUSION**

This work has provided a guideline on the way to design and optimize fully printed semitransparent OPV modules with scalable process technologies. We have demonstrated, from cell to modules, the route we have taken to achieve a fully printed OPV module with respectable performance with fully printed ITO-free electrodes. AgNW have proven to be an excellent electrode material for small scale lab cells.\(^{9–14}\) However, in terms of fabrication of large area modules, AgNW electrodes pose a few challenges. First, their wire mesh surface is rough and provides only limited contact surfaces in the critical interconnection region between cells. The former results in the formation of shunts while the latter results in high contact resistances and limited current carrying capacities. Another challenge with AgNW electrodes lies on the narrow processing window for the creation of conductive interconnects between cells, which is due to the relatively low ablation threshold of AgNWs. In this work, we have demonstrated how these issues can be resolved by rational module layout design and proper process optimization.

First, the process window of laser ablation is enlarged by coating a 120-nm thick ZnO ETL on top of the AgNW bottom electrode, which provides protection against laser ablation and additionally reduces shunting by planarizing AgNW layer. By careful and systematic optimization of the laser power, we have managed to establish reproducibly a low resistive interconnection between cells by selective removal of active layer and ETL without significant damage to the fragile AgNW bottom electrode.

Second, the critical interconnection is further optimized to achieve less than 0.7 $\Omega$ in resistance by analysing its change with respect to the width of interconnection. We have also experimentally proven interconnection width of at least 100 $\mu$m is needed to sustain the current density of continuous operation in OPV modules. Finally, the importance of appropriate AgNW formulation is demonstrated. AgNW bottom electrodes with fillers have significantly higher interconnection resistance and are unsuitable for bottom electrodes for OPV modules.

Based on these findings, fully printed OPV modules with two different active materials are fabricated with less than 0.7% loss in PCE compared with test cells. Overall, fully printed OPV modules in this work has managed to reach 5% in PCE. Further performance enhancement can be achieved by the utilization of the state-of-the-art photoactive material with little changes to the module structure and process parameters as the current design already has sufficient design margin to handle the higher current of these systems.\(^2\) Better optical alignment for all patterning steps will improve the GFF beyond the \(~90\) of the fully printed OPV modules in this work.

This work has proven that printed electrodes, in particular AgNW, can indeed be used as both bottom and top electrodes for OPV module production with no drawbacks in device performance. This opens the way to fully roll-to-roll manufacturing of optically attractive and flexible semitransparent photovoltaic modules, which are ideally suited for building and textile integration. Moreover, the ability to fully print OPV modules at room temperature will widen applications to any discretionary surfaces, even on 3D objects.
6 | EXPERIMENTAL SECTION

6.1 | Preparation

Float glass sheets from Weidner Glas were used as substrates for cells (25-mm square), CBKR and modules (50-mm square). ITO substrates were also purchased from the same company for reference device fabrication. Before printing, the substrates were rinsed with DI water and sonicated in acetone (technical grade >99%, Chemsolute) as well as isopropanol (technical grade >99%, Chemsolute) for 5 min.

AgNW solution were purchased from Cambrios Advanced Materials Inc. AgNW ink without fillers, branded as Ink-Y were diluted to 80% vol. concentration with isopropanol (ACS Grade >99% Sigma Aldrich). AgNW ink with fillers, branded as ink-N NKA710 was diluted in 1:1 ratio by DI water. All solutions are then filtered by a 30 μm polycarbonate mesh syringe filter before use.

Donor polymer (PV2000, Raynergy Tek), acceptor6 phenyl C61-butryc acid methyl ester (PC60BM 99%, Solenne BV) in 2:3 ratio, which were dissolved in a 2:3 (vol.) mixture of xylene (>98%, Merck Chemicals) at 80°C under constant stirring for 24 h.

Donor polymer (poly-3hexylthiophene [P3HT], Merck), acceptor rhodanine-benzothiadiazole-coupled indacenodithiophene (oIDTBR, Flexink Ltd.) which were dissolved in a 95:5 mixture of xylene (>98%, Merck Chemicals) and p-bromoanisole (>99%, Sigma-Aldrich) at 60°C under constant stirring for 2 h.

PEDOT:PSS, that is, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate solution was prepared by dilution of HTL Solar from Heraeus with DI water at 1:1 ratio; 0.1 vol.% of Fluorosurfactant Capstone FS-31 and 0.05 vol.% of Silane Silquest A-187 were subsequently added and mixed.

ZnO nanoparticle dispersion, N-10 were purchased from Avantama AG. The solution was filtered by 0.22-μm PVDF syringe filter before use.

6.2 | Equipment

The doctor blade (Model ZAA2300) and applicator (Model ZUA 2000) are from Zehntner GmbH. Laser patterning is done by LS-7xxP from LS Laser Systems GmbH equipped with a Q-Switched LASER from InnoLas Photonics GmbH (Nano air 532-10-V, 532-nm 20-ns pulse). Thermal evaporation is done in Oerlikon Leybold UNIVEX 250 system.

Characterization of the solar cells and modules were done with LS0916 AAA solar simulator from LOT Quantum Design. The power output was calibrated prior to the measurement using a reference single-crystal silicon solar cell from the same company. Diode current voltage characteristics were obtained from B2901A sourcemeter from Keysight Technologies Inc. The current from device was measured when the voltage range swept from –1 to 1.5 V using 20-mV steps for cell devices while the voltage range swept from –2 to 6 V using the same voltage steps for module devices. All device area was confirmed by confocal microscope μsurf custom from NanoFocus AG. SEM images and dark field microscope images were obtained from JSM-7610F from JEOL Ltd and MX51 from Olympus Corporation respectively. 34461A digital multimeter from Keysight was used to measure the voltage across CBKR while B2901A sourcemeter was used to provide constant current during measurement. Sheet resistance of films were measured by a noncontact sheet resistance meters from Delcom Instruments Inc. Film thicknesses were measured by Alpha-Step D-100 Stylus Profiler by KLA-Tencor Inc. Individual film thicknesses of all cells and modules are checked to ensure they have roughly the same film thicknesses during fabrication. Transmittance data were acquired with a Lambda 850 UV/VIS Spectrometer from PerkinElmer Inc. Transmission spectra were recorded between 300 and 800 nm with a step size of 1 nm using an integrating sphere.

6.3 | Cell/module fabrication

ZnO, photoactive layer and PEDOT:PSS layers were coated from left to right direction in Figure 55 (perpendicular to the direction of current flow), whereas AgNW were coated from top to bottom direction (in the direction of current flow). Films coated by doctor blade coating are usually thicker at the beginning than at the end. A gradient of film thickness in the direction of current flow would lower \( j_{sc} \) of the module. On the other hand, AgNW films usually have a directional orientation along the blading direction due to the shear force applied. Thus, blade coating along the current flow direction of the organic solar module can reduce the overall series resistance of the device.

Bottom AgNW electrode was fabricated by doctor blade blading AgNW solution at 12 mm/s 30°C at 400-μm gap for AgNW ink without filler and 10 mm/s 65°C at 400-μm gap for AgNW ink with filler. The film was then annealed at 120°C for 5 min.

ZnO was coated at 5 mm/s 30°C at 400-μm gap to achieve 40-nm film thickness. The process was repeated three times in opposite directions to achieve 120-μm film thickness. The film was then annealed again at 120°C for 5 min. The bottom electrodes were then patterned by LASER at 4 W 60 kHz at 500 mm/s. The same procedure is also applied as the P1 step in module fabrication.

The photoactive layer was printed from one of the two formulations described above. P3HT:oIDTBR solution was coated at 33 mm/s 65°C at 400-μm gap. PV2000:PC60BM solution was coated at 30 mm/s 65°C at 400-μm gap. PEDOT:PSS layer was then coated at 5 mm/s 65°C at 400-μm gap. All cells were annealed at 140°C for 5 min under nitrogen atmosphere. For organic solar modules, the samples were then transferred back to ambient for P2 LASER structuring at 800 mW 60 kHz at 500 mm/s. One LASER structuring lines is roughly 70 μm. Wider interconnection is achieved by overlapping each patterning line at 5-μm distance.

Top AgNW layer was coated at 20 mm/s 30°C at 400-μm gap with formulation that has no fillers. The top electrodes were patterned by LASER at 800 mW 60 kHz at 500 mm/s. The same procedure is also applied as the P3 step in module fabrication. The film was then annealed again at 120°C for 5 min under nitrogen atmosphere.
EvapAg layer was coated in evaporator under pressure $<6 \times 10^{-6}$ bar for 200 nm at a rate $<0.05$ nm/s.

To make efficient contacts with the bottom electrode after fabrication, Q-tips that are soaked in toluene are used to gently wipe the active layer off. Silver paste is then painted at the contact positions of the exposed bottom electrode and at the contact positions of the top electrodes.

After, fabrication cells were encapsulated by adding a barrier glass on top of the device with the UV curable epoxy Delo-Katiobond® LP655 for 2 min.

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CONFLICT OF INTEREST
There are no conflicts to declare.

DATA AVAILABILITY STATEMENT
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information may be found in the online version of the article at the publisher’s website.

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