

Solution-Processed Organic Solar Cells

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Abstract

Organic solar cells, based on polymer/fullerene-blend films, are advancing rapidly toward commercial viability. In this article, we review recent progress on two issues critical for technological applications: device photovoltaic efficiencies and processing technologies for high-throughput production. In terms of device efficiencies, we consider advances in low-bandgap polymers, film morphology, and device structure aimed at increasing efficiencies beyond 5%. We then review recent progress in developing high-throughput, solution-printing-based processes for low-cost device fabrication.

Introduction

Today, after 50 years of challenges and excitement, the field of photovoltaics (PV) is becoming a significant success story, with a market value in 2007 of over \$10 billion and an annual growth rate of 30–40%. Although PV's contribution to the global electricity supply still remains less than 0.1%, the scale of the global solar resource (the solar irradiation incident upon Earth in one hour is equivalent to the world's annual energy demand) coupled with the rapid growth of the PV market suggests that PV could soon be playing a major role in the global energy supply.

The past 50 years have been dominated by silicon-based PV. Over the past decade, thin-film PV technologies, so called second-generation technologies such as a-Si (amorphous silicon), CIGS (copper indium gallium diselenide), and CdTe (cadmium telluride) have rapidly caught up in terms of performance and production technology and are now undergoing a rapid expansion in production. The next few years will determine whether this indicates the beginning of the end of the dominance of the PV market by crystalline Si.

The time frame for a PV technology to go from laboratory concept to market is approximately 20–30 years. With a-Si, CIGS, and CdTe now becoming established in the market, the question arises as to what photovoltaic technology platform(s) will follow. Research into such platforms is currently proceeding in two directions: ultrahigh efficiencies and ultralow production costs. For lowest cost PV production, solution-processable solar cell technologies based on both molecular and inorganic

light absorbers, including polymer-blend, small-molecule, dye-sensitized, and inorganic nanoparticulate devices, are all receiving extensive attention.

In this article, we focus on solution-processed organic solar cells (OSCs) based on polymer/fullerene-blend films (see Figure 1). Such devices have great potential for ultralow costs, not so much because of the costs of the material classes under consideration, but more because of the productivity of the printing and coating techniques compared to wafer-based production techniques. The only production technology that can achieve production rates of greater than 10,000 m²/h at production costs as low as even €(s)/m² is web coating or printing (including vacuum web coating). Web coating and printing are alternative names for roll-to-roll coating and printing. The potential low costs for a printed/coated PV technology are extremely encouraging and are motivating the academic and industrial research communities to develop this technology to market readiness within this decade.

The status of organic photovoltaics (OPV) was last reviewed in this journal in January 2005.^{1,2} In this article, we address some of the subsequent exciting new developments in this field, focusing on two key issues:

- selection of materials for the photoactive layer to achieve further advances in device performance and
- selection of a high-volume, high-speed, and high-yield production process.

This article is divided into two parts. The first part describes the state-of-the-art

performance for solution-processed PV, and the second part discusses production and application aspects in more detail.

Advances in Device Efficiency

The photoactive (light-absorbing) layer of solution-processed organic solar cells is typically based on a bicontinuous blend of two organic semiconductor materials (see Figure 1), most often a conjugated polymer blended with a soluble C₆₀ derivative. Offsets of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels of the two materials result in the formation of a bulk heterojunction with the polymer being the electron donor and C₆₀ the electron acceptor. Optical excitation of the blend results in photoinduced charge separation across the heterojunction, and subsequent charge transport through the two phases allows current output to an external circuit.

The current workhorse materials employed in OSCs are poly(3-hexylthiophene) (P3HT) and the C₆₀ derivative PCBM ([6,6]-phenyl-C61-butyric acid methyl ester), as shown in Figure 1. Several groups have now reported small-area device efficiencies in excess of 4% with P3HT/PCBM blends.^{3–5} This success can be attributed to several different properties of this materials combination. The P3HT optical bandgap (~1.9 eV) and strong absorption coefficient enable efficient light absorption up to 650 nm for film thicknesses of only ~200 nm. The LUMO level of PCBM is ~1 eV lower than that of P3HT, providing a sufficient driving force for interfacial charge separation. The free energy of the charge-separated state approximated by the difference in energy between the PCBM LUMO and P3HT HOMO levels, is sufficient to generate a reasonable output voltage of over 600 mV. The charge mobilities of the holes in P3HT and electrons in PCBM are on the order of 10⁻⁴ cm²/(V s) or higher, thus avoiding significant resistive losses during charge transport to the device electrodes.

Crucially, P3HT and PCBM are amenable to systematic optimization of blend morphology. They are reasonably miscible but show strong tendencies to crystallize into separate domains on the nanometer length scale^{3–5} and to form concentration gradients normal to the device electrodes. A range of different strategies have been employed to optimize film morphology and crystallinity, including control of polymer regioregularity, film deposition conditions, and postdeposition film processing.^{3–5} These strategies appear to have been successful in achieving film morphologies compatible with efficient charge separation and collection while

Figure 1. Structure of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM).

minimizing losses. The device is guided by these elements.

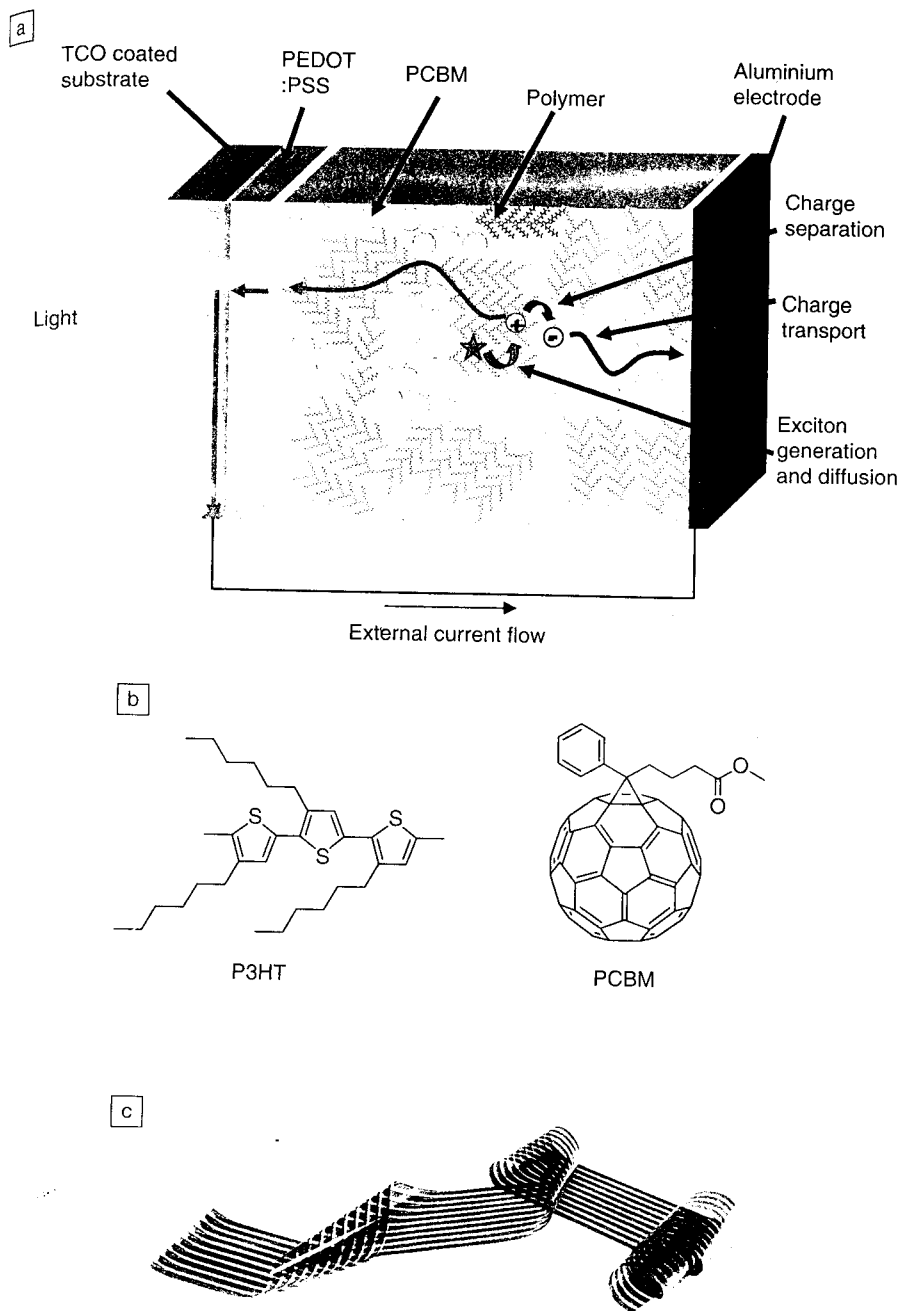


Figure 1. (a) Schematic layout of the function of a typical organic solar cell. (b) Chemical structures of typical donors and acceptors. (c) Photograph of reel-to-reel-fabricated organic solar cells. The active layer of the solar cells is a P3HT/PCBM blend. Note: P3HT is poly(3-hexylthiophene), PCBM is [6,6]-phenyl-C61-butyric acid methyl ester, PEDOT is poly(3,4-ethylenedioxythiophene), PSS is poly(4-styrenesulfonate), and TCO is transparent conductive oxide.

minimizing interfacial charge recombination losses, thereby achieving device efficiencies in excess of 4%.

Theoretical analyses of the optimum device performance achievable as a function of particular materials properties have guided strategies to make further improvements in device performance beyond

P3HT/PCBM.⁷ Such analyses have identified two key strategies to achieve further advances. One strategy aims at improving the voltage output of the solar cell. This voltage is limited by the free energy of the photoinduced charge-separated state and, therefore, by the energy difference between the polymer HOMO level and PCBM

LUMO level. Strategies to address this limitation have typically targeted the use of polymers with larger ionization potentials.

The alternative strategy is to improve the photocurrent by enhancing light absorption by the photoactive layer. This has typically been addressed by lowering the polymer's optical bandgap to enhance the absorption of longer wavelength light. The past few years have seen an intense program of polymer design, synthesis, and evaluation based on these two strategies (see, for example, Reference 8). In practice, it has proved to be remarkably challenging to improve device performance beyond that of P3HT/PCBM blends. We have, for example, recently suggested that achieving efficient dissociation of photogenerated charges might be a more significant challenge than had previously been appreciated, with dissociation typically requiring a relatively large free energy for charge separation to give initially generated polarons sufficient thermal energy to overcome their Coulombic attraction.⁹

Nevertheless, an exciting range of new materials have been evaluated for OSC applications, including not only homopolymers but copolymers and metallated conjugated polymers. The new materials and their evaluation lead not only to enhanced device performance but also to improved understanding of the underlying science relating materials properties to device performance. A particularly notable success has been the low-bandgap benzothiadiazole-fused thiophene copolymer PCPDTBT (poly[2,6-(4,4-bis-(2-ethylhexyl)-4H-cyclopenta[2,1-b,3,4-b']dithiophene)-alt-4,7-(2,1,3-benzothiadiazole)]).^{10,11} This class of materials led to a certified performance of over 5% as depicted in Figure 2. When blended with the C₇₀ analogue of PCBM and used with a chlorobenzene/octane dithiol cosolvent system for film deposition, this polymer yielded efficient photocurrent generation out to 850 nm and a white-light power conversion efficiency of up to 5.5%.¹¹

In parallel with these studies aimed at advancing device efficiencies by tuning polymer HOMO and LUMO levels, other groups have been trying a range of innovative approaches to achieve enhanced control over the morphology of the photoactive layer, including the use of block-copolymer and dendritic structures.¹² Although such approaches have, to date, led to only modest device efficiencies, the more systematic control of blend morphology that can be achieved with such structures is very promising.

Aside from improvements in the photoactive layer, several studies have targeted advances in device efficiency through overall device design. One

approach is to use light management approaches to enhance light absorption. The top contact of OPV devices is typically a reflective, metallic electrode, opening the potential for multiple light pathways through the active layer, as well as interference effects. Examples of light management strategies already showing promise include the use of TiO_x optical spacer layers to optimize the spatial distribution of light within the device¹³ and the use of corrugated device structures to ensure multiple light paths through the active layer.¹⁴ An alternative approach is to employ tandem device structures. For example, a 6% device efficiency has recently been reported for an all-solution-processed tandem polymer cell employing a TiO_x interface layer between the two cells.¹⁵

Production Technologies

Highest productivity at lowest process costs is gained from roll-to-roll processing. Roll-to-roll processing can be divided into solid-state (transfer), gas-phase (vacuum), and liquid-state deposition techniques. Production of photovoltaic modules requires at least the deposition of stripes of active layers using a pattern on to a web. The method electrically separates the active layers into individual cells, which later are interconnected in series (Figure 3). All three classes of deposition processes can produce either patterned or unpatterned films. Techniques for patterning of thin films can be divided into subtractive or additive methods. In the first case, an unpatterned film becomes patterned in a separate process step, by removing film from undesired areas (e.g., by lithography, laser patterning, embossing, or imprinting, to name a few of the more popular technologies for postpatterning). In the case of additive methods, film deposition and patterning are done in the same process step.

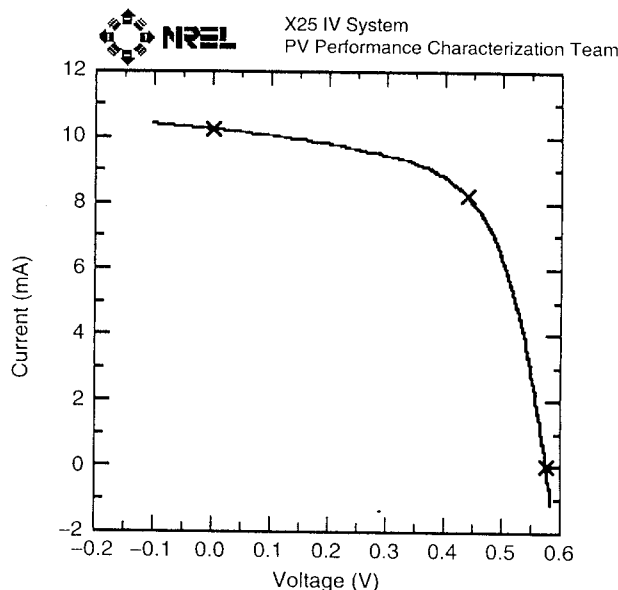
Solid-state printing typically starts from already prepatterned films. Alternatively, the pattern can be generated during the transfer process by a technique such as a laser transfer method, in which a laser beam ablates material from the web.

Patterned vacuum deposition is realized by shadow masking or by lift-off processes. Both processes have limitations in terms of resolution and registration. Shadow-mask processing requires specific precautions to prevent particles and to control the distance between the web and the mask. More sophisticated lift-off processes are used for the manufacturing of patterned Al films. Here, a fast-evaporating ink (e.g., an oil) is printed right before the film is run through the evaporation unit. During evaporation, the

Konarka Technologies Organic Cell

Device ID: LS1
Jul 12, 2007 13:58
Spectrum: AM1.5-G (IEC 60904)

Device Temperature: $25.0 \pm 1.0^\circ\text{C}$
Device Area: 0.685 cm^2
Irradiance: 1000.0 W/m^2



$V_{oc} = 0.5756 \text{ V}$
 $I_{sc} = 10.218 \text{ mA}$
 $J_{sc} = 14.917 \text{ mA/cm}^2$
Fill Factor = 61.05%

$I_{max} = 8.2047 \text{ mA}$
 $V_{max} = 0.4376 \text{ V}$
 $P_{max} = 3.5908 \text{ mW}$
Efficiency = 5.24%

Figure 2. National Renewable Energy Laboratory (NREL) certificate for a solar cell based on an electron-donating polymer from the class of bridged bithiophene copolymers.²⁹ This type of polymer already exhibits short-circuit current densities of up to 15 mA/cm^2 at external quantum efficiency (EQE) values of 60% at 800 nm. A fully optimized polymer with an EQE as high as 80% has the potential to deliver short-circuit currents of up to $18\text{--}20 \text{ mA/cm}^2$ and efficiencies of around 7%.

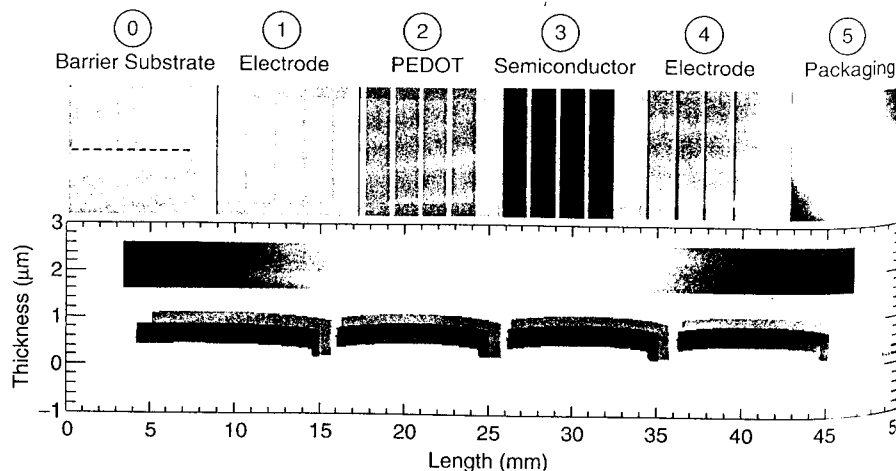


Figure 3. Cross section through a four-stripe organic photovoltaic module. Note that the x and y axes have different scales. Production requires five major steps: (1) deposition of the electrode, (2) deposition of the electron blocking layer [i.e., poly(3,4-ethylenedioxythiophene) (PEDOT)], (3) deposition of the semiconductor, (4) deposition of the top electrode, and (5) packaging.

ink released from the substrate, and adhesion of the metal is prevented. The best resolution and registration are gained with classical lift-off or photolithography methods, both of which require printing of a suitable photoresist.

Liquid-state printing results directly in patterned films and is typically used where high resolution and registration are required. In contrast, liquid deposition methods that result in unpatterned films are commonly called "coating" methods. Coated, unpatterned films can be converted into patterned films by subsequent combination with (subtractive) patterning steps (e.g., mechanical scribing or laser patterning).

Production of Solar Cells by Coating Techniques

Over the years, the processing of bulk heterojunction solar cells has become increasingly sophisticated. The first reports on solution-processed devices utilized spin coating as the manufacturing process.^{16,17} Spin coating is a well-understood process for the deposition of thin film coatings with high precision. The method requires little formulation work and is quite forgiving over a considerable range of viscosities. Wetting can be guaranteed by surface pretreatment of the substrate. The film thickness is controlled by a combination of spinning speed, acceleration, solution viscosity, and temperature: typical spin-coated films are between 50 and 5,000 nm. Typical spin-coating machines handle substrates up to 6–8 in. in diameter. The disadvantages of spin coating are the large material losses (>90%) and the incompatibility with roll-to-roll processing. Whereas the material losses can be minimized by collecting and purifying the residuals from the spin flow, the incompatibility with roll-to-roll processing remains.

Shortly after the first presentation of spin-coated organic solar cells, doctor blading was demonstrated to overcome many of the disadvantages of spin coating.^{18,19} Doctor blading is a coating technology that is comparable to knife coating, whereby a knife homogeneously spreads ink as the blade is drawn over a substrate. Doctor blading is fully compatible with both batch and roll-to-roll processing. Ink formulation for doctor blading is quite comparable to that for spin coating. Four parameters determine the film formation: the concentration (responsible for the viscosity of the ink), the temperature of the solution, the speed of the blade, and the distance between the blade and the substrate (i.e., the volume of ink being deposited). The temperature profile and

the drying kinetics of the wet film during blading are quite different from those encountered during spin coating and can lead to different solid-state morphologies.²⁰ However, detailed investigations have shown that blading can result in fully optimized solar cells with performances identical to those of cells prepared by spin coating.¹⁹

Another coating technology, spray coating, which is even easier to perform than doctor blading, has been suggested as an elegant alternative.²¹ Spray coating transfers small droplets (from microns to tens of microns) of ink onto a substrate, where they dry upon impact. Ink formulation is not an issue, as the method works well for highly diluted solutions. Spray coating typically results in rough thin-film surfaces. Because of the transfer of rather large droplets that individually dry on the substrate without first forming a closed film, the surface roughness is typically in the tens of nanometers (compared to a few nanometers for bladed or spin-coated films). Nevertheless, coherent films and fully functional solar cells and photodetectors have been obtained by spray coating the organic semiconductor layer to a dry film thickness of 3–4 times the surface roughness.²²

All of these pioneering trials in coating organic semiconductor layers and converting them into solar cells have indicated that reel-to-reel coating of organic solar cells is possible. Indeed, the successful manufacturing of organic solar cells using reel-to-reel slot-die coating has been reported.²³ Both of the active layers—the electron-blocking layer [PEDOT:PSS, where PEDOT is poly(3,4-ethylenedioxythiophene)] and PSS is poly(4-styrenesulfonate)] and the semiconductor layer (P3HT:PCBM)—were coated at a speed of meter(s) per minute on a 50-mm-wide indium tin oxide (ITO)-coated polyester film. Small areas of the web-coated film were converted into functional solar cells by vacuum deposition of Al as the top electrode (cathode), resulting in device efficiencies of 1%.

The fabrication of modules requires some patterning of the coated layers. At least the interconnect region, where adjacent cells are electrically connected to each other in series, needs to be kept clean from coated material. Most of the coating techniques do have a limited potential to deposit patterned layers. The patterning of long stripes in the down web direction as specified for photovoltaic modules is fortunately compatible with the specifications of various coating methods (e.g., by using more complex coating heads with multiple die channels). Nevertheless, the resolution and registration of such a process are somewhere in the submillimeter range, which is nearly one order of magnitude less than desired. An elegant way to overcome this limitation is to post-pattern coated films. Postpatterning of flood-coated films can be done by either mechanical or optical methods. Laser patterning of thin films is well-established in the inorganic thin-film photovoltaic community and was probed for the production of organic solar cells in combination with large-area coating. Initial trials indicated the high potential of this method with respect to resolution, speed, and costs.²⁴

Production of Solar Cells by Printing Techniques

Printing is, of course, the most straightforward technology for creating patterned films. However, most printing methods require relatively high-viscosity inks. Coating is more compatible with low-viscosity inks, as it does not generate a pattern that needs to stay on the substrate while drying. Each of the printing methods operates best for specific viscosities, and fully optimized inks can produce the resolutions and productivities listed in Table 1. There are three printing methods that work well with low-viscosity inks: inkjet, flexo, and gravure printing. These methods would be natural choices for the production of organic solar cells.

The first report on printed organic solar cells used a screen printing process to

Table 1: Characteristic Properties of Printing Methods.

Method	Resolution (μm)	Layer Thickness (μm)	Ink Viscosity (Pa s)	Maximum Throughput (m^2/h)
Inkjet	>50	0.1–20	0.001–0.04	>50
Screen	>100	1–15	0.5–50	>100
Flexo	>50	0.5–2.5	0.05–0.5	>50,000
Gravure	>30	0.5–8	0.05–0.2	>100,000

Note: The table summarizes typical performance values for the printing methods. However, deviations can be found in the case of specialized inks or processes.

deposit the active layer. In screen printing, a stencil is patterned by closing selected areas of a screen. Upon imprint, ink is forced through open meshes by means of a blade. Screen printing typically requires 10–100 times higher viscosity inks than those used for spin coating or doctor blading. Nevertheless, even the very first report on screen-printed solar cells described excellent performance. Cells in which the active layer was screen printed were directly comparable to the spin-coated reference.²⁵ Screen printing of OPV has been reported a few more times,²⁶ but the requirement for high viscosities limits the available materials. Only conjugated polymers with high molecular weights and high solubilities are suitable for high-quality screen printing. Today's commercial screen printing inks are quite complex fluid systems, in which various binders, thickeners, and thixotropy and viscosity modifiers are mixed with the pigment. Thixotropy modifiers decrease the viscosity of the ink upon applied stress. These additives certainly can convert conjugated-polymer-based fluids into well-defined screen printing inks, but so far, no studies have been conducted to determine whether these additives have an impact on the photovoltaic performance.

Inkjet printing has evolved as the most popular printing method for the production of multicolor polymer light-emitting diode (PLED) displays and, by now, is probably the most widely used printing method in organic electronics. The two basic methods for droplet formation

and control are continuous and drop-on-demand (DoD) systems. In the former case, a continuous liquid jet through a nozzle is split into drops, for example, by means of an acoustic wave generated by a piezoelectric device. Ejected droplets are then electrically charged and deflected according to the desired pattern. In DoD systems, droplets are discontinuously created by acoustic (mostly for non-water-based inks) or heat (mostly for water-based inks) pulses when needed. Inkjet systems can reproducibly handle droplets of a few picoliters up to nanoliters, corresponding to pixel sizes of at least a few microns.

Inkjet printing of solar cells has different requirements than inkjet printing of PLEDs. Photovoltaic applications require large and homogenous coated areas. DoD inkjet printers generate single pixels with diameters according to the droplet size and to the number of droplets fired, making the printers unsuitable for PV applications. Successful inkjet printing of photovoltaic cells requires the development of inks that dry sufficiently slowly, in combination with a printing scheme that fires droplets sufficiently narrowly spaced to make single pixels coalesce over a reasonably large area before drying. Following these design rules, high-performance P3HT/PCBM solar cells were inkjet printed from a solvent mix of a high-boiling and a low-boiling solvent (Figure 4).²⁷ It was further shown that such solvent combinations can significantly influence the solid-state morphol-

ogy formation of the bulk heterojunction composite during the drying phase, especially with respect to vertical phase separation of the polymer to the fullerene.

Only a few research groups worldwide work on the application of mass printing for the fabrication of large-area, flexible electronics. This is probably due to the extensive know-how and comparatively high effort required for adapting new ink systems for mass printing.

Two widely discussed mass printing methods for printed electronics are gravure and flexography printing. In flexography printing, soft and flexible printing plates are used in combination with low-viscosity inks and low transfer pressures. The flexibility of the printing plates contributes to reducing the resolution and layer quality. No other printing method, however, can deposit low-viscosity inks with comparatively high and variable thicknesses up to several micrometers on both flexible and rigid thin films.

In gravure printing, the printing pattern is engraved into the printing plate (or film/cylinder). During inking, the engraved cells of the plate are filled, and during imprinting, the substrate moves and takes up the ink from the cells. Gravure printing can give outstandingly high resolution and registration for relatively low-viscosity inks. These properties in combination with a high-throughput production capacity, make gravure printing a promising method for the mass production of OPV. The first gravure printing trials of P3HT/PCBM blends reported excellent film quality.²⁸ The homogeneity and the surface roughness of gravure-printed P3HT/PCBM films were comparable to that of spin-coated layers. The gravure-printed solar cells had good diode properties and sufficient shunts. However, the short-circuit current of the gravure-printed solar cells was lower than average for P3HT/PCBM cells and might result from the formation of a less favorable morphology during the drying process. Nevertheless, these preliminary studies clearly demonstrated that organic solar cells can be mass-produced by printing methods.

Conclusions

Developments in organic photovoltaics are moving rapidly. The best laboratory efficiencies are currently in the 5–10% range and the community has developed a clear understanding and strategy of how to further evolve performance. No fundamental obstacles have been identified to achieving efficiencies of 10–15%, and the general expectation is that the further progress will come from both novel optimized materials and more efficient device geometries.

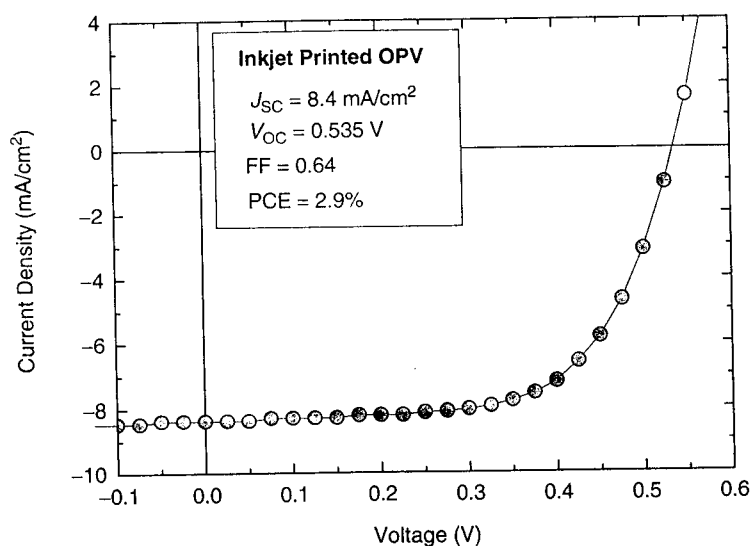


Figure 4. Current-voltage (J - V) curves for an inkjet-printed organic solar cell under illumination. The semiconductor solution, a poly(3-hexylthiophene)/[6,6]-phenyl-C61-butyric acid methyl ester (P3HT/PCBM) composite, was printed from a mesitylene/*ortho*-dichlorobenzene mixture. Note: J_{sc} is short-circuit current, V_{oc} is open-circuit voltage, FF is fill factor, and PCE is power conversion efficiency.

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breakthrough has been achieved in the development of production methods for organic solar cells during the past few years. Multiple printing and coating methods have been demonstrated to yield the required film specifications for device operation. Moreover, one company recently succeeded in the roll-to-roll production of photovoltaic modules, where all layers were deposited by printing and coating methods. Roll-to-roll manufacturing of functional modules was the last missing piece of technology between development and commercialization, and the first organic solar cell products are expected to enter the market within the short term.

Acknowledgments

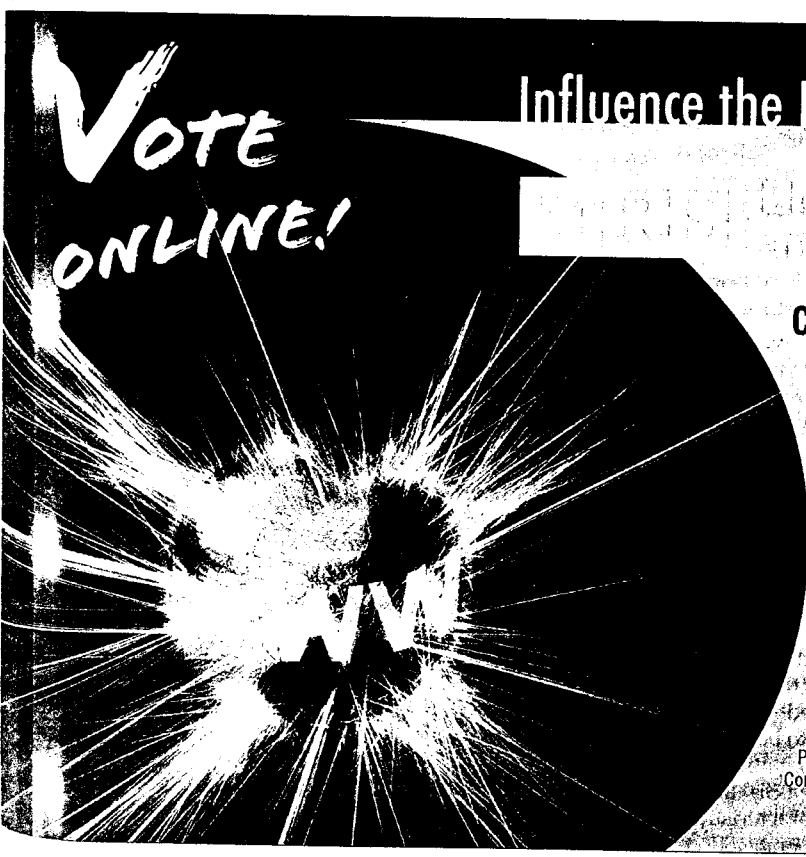
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Visit www.mrs.org/2008_election
for details on the candidates

Paper ballots available upon request
Contact Kathy D'Biagio, dbiagio@mrs.org, 724.779.3004 x102