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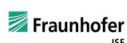




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1 Overview of the project

Important advantages of OPV against other PV technologies include low cost and easy manufacturing on different flexible substrates using roll-to-roll (R2R) printing and coating technologies. High volume production technologies enable cost-efficient industrial production compared to vacuum processes. Moreover, the OPV technology does not present raw material abundance or toxicity problems at the same level as other PV technologies do. In addition, printing brings the advantage of large area arbitrary size and shape processing which increases the freedom of product design and its integration into various types of applications.

OPV's main application areas today are in low power electronic devices but there are visions of OPV entering into Building Integrated PV (BIPV) area as a high cell efficiency is not essential where there is a large area available, and further, when technology develops to energy harvesting for standalone stations and grid energy harvesting. Europe has a leading role in the development of OPV technologies and ArtESun combines the multidisciplinary and complementary competences of top-level European research groups and industries to generate new innovations in the field of OPV and to stay in the front line of development. The partners combined represent the capability to make break-through advances in the development of high performance innovative materials for multi-junction OPVs suitable for cost-effective non-vacuum production of arbitrary size and shape modules with efficiency over 15% in relevant environment and lifetimes relevant to its expected future applications. Application potential has been demonstrated in the field of BIPV as ventilated facade elements, as an energy harvesting element for RFID, and as a photovoltaic antenna for wireless sensor network nodes.

The main objectives for the project are:

Materials developments to increase the efficiency of multi-junction architectures, up to 15% PCE in wavelengths up to 1000 nm. Novel material solutions for donor and acceptor materials will be developed as well as new solutions for interlayers and electrodes. For these novel compounds sustainable scale-up routes will be identified to clarify the potential for large-area processing of OPV modules with these materials on the longer term. Novel eco-friendly ink formulations for the photo-active layers will be targeted, avoiding the use of halogenated solvents.

Alternatives to the costly transparent IndiumTinOxide (ITO) electrode, especially processing technologies for ITO-free highly conductive transparent electrodes will be developed using two approaches, i) based on sputtered thin Ag layers (MO/Ag/MO), or ii) based on printing metal grids with metal nanoparticle inks and printing of Ag nanowires or conductive PEDOT materials on top of the grid structure.

Non-vacuum, low-cost multilayer printing, slot-die coating and self-patterning processes was investigated for the processing of the ITO-free transparent electrodes, photoactive layers and the ultra-thin interface and recombination layers, with a focus to produce OPV modules with arbitrary size and shape.

In this project focus was on developing stable materials and device structures, including also the barrier layer required for shielding the solar cell, as well as processing technologies for these materials.



Internal and external degradation mechanisms was analysed and characterized for example using special indoor and outdoor testing facilities. In the project ultra-thin flexible glass to guarantee ultra-high barrier properties against external degradation has been introduced.

2 Photo active materials development

One of the objectives was to develop high efficiency multi-junction OPV devices, exceeding 15% PCE, with novel donor and acceptor materials. In order to achieve these objectives, efforts under the active material development work package were focused on the development of both low and wide band gap donor polymers with complimentary absorption properties and developing novel small molecule acceptors as replacements for fullerene based acceptors. Our efforts to deliver photoactive materials capable of reaching 8% power conversion efficiency have been focused on three different classes of push-pull type medium-to-narrow band gap semiconducting polymers. Pleasingly, all three polymers have been optimised through synthetic modifications, polymer weight fractionations and OPV device optimisations to afford single-junction solar cells with efficiencies exceeding 8%.

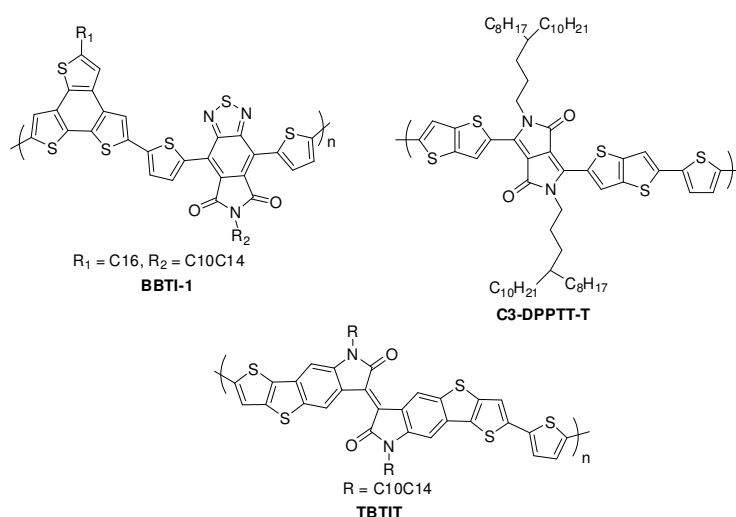


Figure 1 Structures of polymers synthesised.

A novel eight ring benzothieno[3,2-*b*]thiophene isoindigo acceptor unit was designed and a copolymer with thiophene was made to afford polymer **TBTIT-h** with a number average molecular weight of 152 kDa, an ionisation potential of 5.1 eV and an optical band gap of 1.6 eV (*Adv. Mater.* **2015**, **27**, 4702-4707). The single junction solar cell devices derived from **TBTIT-h**:PC₇₁BM blends (1:2 weight ratio) exhibit power conversion efficiencies up to 8.7 % with a high J_{sc} of 17.08 mAcm² and a high FF of 0.70 V for as-cast films without using any additives or post-solvent/ thermal annealing steps during the device fabrication.

Polymer **BBTI-1** was synthesised by coupling benzo[1,2-*b*:3,4-*b'*:5,6-*d'*]trithiophene (BTT) as the electron-rich unit and 2,1,3-benzothiadiazole-5,6-dicarboxylic imide (BTI) as the electron deficient unit (*Adv. Mater.* **2015**, **27**, 948-953). **BBTI-1** was obtained with a number average molecular weight (M_n) of 64 kDa and a polydispersity index (PDI) of 1.99. The photovoltaic properties of **BBTI-1** were examined in an inverted OPV device architecture with PC₇₁BM as the electron acceptor material. The photoactive layers comprised a 1:2 weight ratio of polymer to fullerene and were in the first instance



solution cast from neat *o*-dichlorobenzene (ODCB). Using these conditions, **BBTI-1** afforded a high power conversion efficiency (PCE) of 8.3% owing to a high short-circuit current (J_{sc}) of 16.45 mA/cm², a high V_{oc} of 0.80 V and a respectable fill factor (FF) of 0.63. Interestingly, when 1,8-diiodooctane (DIO) was employed as a solvent additive, the performance of **BBTI-1** significantly decreased (4.8% PCE) due to a loss in both current, voltage and fill factor.

In addition, we have also synthesised low band gap diketopyrrolopyrrole based polymer C3-DPPTT-T which affords a maximum power conversion efficiency of 8.8% resulting from a J_{sc} value of 23.5 mA/cm², a V_{oc} of 0.57 V and a fill factor of 0.66 (*J. Am. Chem. Soc.* **2015**, **137**, 1314–1321). Apart from medium and low band gap polymers we have also developed wide band gap polymer C2C6-NDT-BT with a band gap of 1.8 eV. A 1:2 (w/w) ratio blend of the polymer with PC₇₁BM in an inverted device architecture gave PCE of 7.5% (*Adv. Funct. Mater.* **2016**, **26**, 6961–6969).

The donor polymers developed under WP1 have been optimised to give efficiency close to 9% with PC₇₁BM, which have poor absorption in the visible region of the spectrum. Our efforts under WP1 to deliver photoactive materials capable of reaching 10% power conversion efficiency have been focused on developing non-fullerene based acceptors with better absorption as an alternative to fullerene based acceptors.

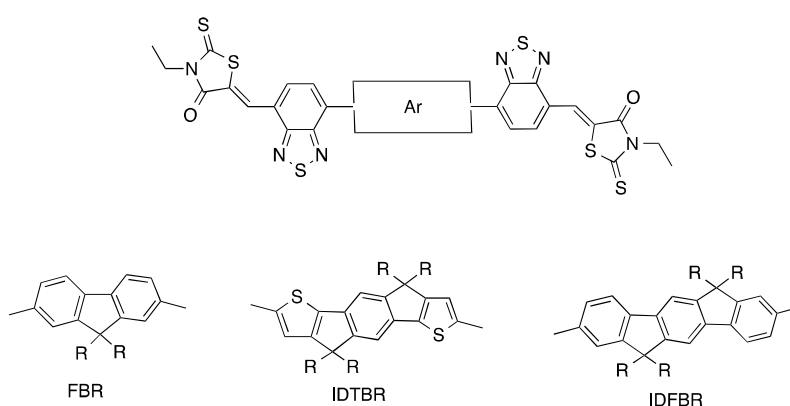


Figure 2 Chemical structure of non-fullerene acceptors.

Solution processable novel small molecule electron acceptors bearing 3-ethylrhodanine flanking groups were synthesized as an alternative for fullerene based acceptor. These molecules were designed to be synthetically simple and versatile, with wide scope to modify the structural, electronic, and morphological properties through chemical design. Incorporation of benzothiadiazole extends the conjugation, thus improving charge transport, as well as affording electron deficient character to the outside of the molecule. To cap the ends of the molecule, 3-ethylrhodanine was chosen as an electron withdrawing flanking group. Furthermore, these molecules are highly absorbing in the visible region and the absorption can be tuned by changing the central aromatic core, offering the potential for enhanced photocurrent generation in OPV devices.

When fluorene was used as the central unit, the molecule gave relatively higher PCE of 4.1% with P3HT as donor with high open circuit voltage (V_{oc} – 0.82 V) compared to PC₆₁BM as an acceptor (V_{oc} – 0.59V) (*J. Am. Chem. Soc.* **2015**, **137**, 898-904). The nonplanar molecular structure results in reduced tendency to crystallize, which helps to prevent large crystalline domains from forming in the bulk heterojunction blend composition over extended lifetimes. However, the inability to form pure domains results in highly intermixed donor and acceptor resulting in low short-circuit current (J_{sc}) in



these devices. In addition, the large extent of spectral overlap of FBR with P3HT and lack of long-wavelength absorption reduced the ability to harvest photons across the spectrum, further limiting the generated photocurrent.

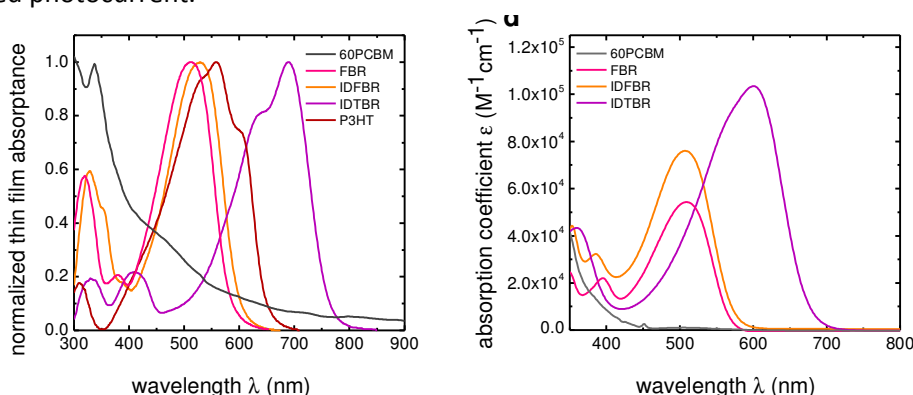


Figure 3 Optical properties of Non-fullerene acceptors in comparison with PC61BM.

In order to address both the spectral overlap and morphological issues associated with FBR the central fluorene unit was replaced with an indacenodithiophene unit. This has the effect of planarizing the molecular structure and thus significantly red-shifting the absorption as well as increasing the tendency to crystallize on length scales commensurate with charge separation and extraction. Power conversion efficiencies of up to 6.4% were achieved, which is, to the best of our knowledge, the highest reported for fullerene-free P3HT solar cells (*Nat. Commun.* **7:11585** doi: [10.1038/ncomms11585](https://doi.org/10.1038/ncomms11585) (2016)). The oxidative stability of these devices is also found to be superior to the benchmark P3HT:PC₆₀BM devices demonstrating this to be a robust and highly promising new materials combination for OPV.

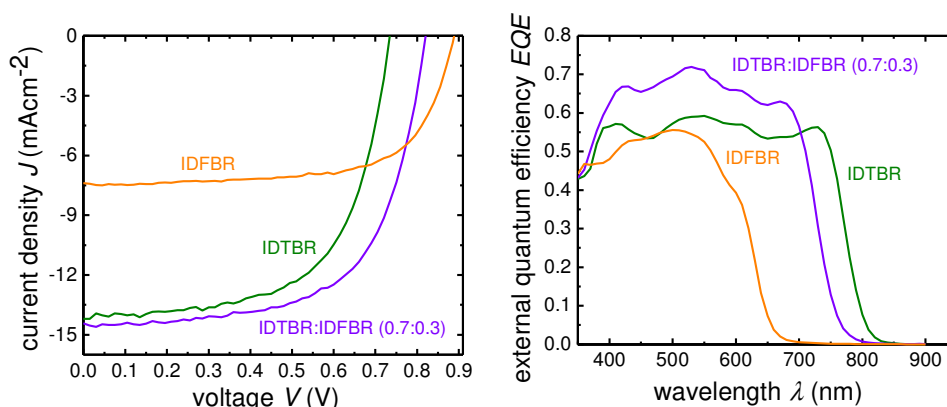


Figure 4 Photovoltaic performances and EQE profiles of binary and ternary devices.

In order to improve the efficiency of binary OPV devices a third component, which is either electron donor or acceptor material, is usually added. The third component is often chosen to have complementary absorption with respect to that of the two components. Similar approach was applied to improve the efficiency of P3HT:IDTBR devices using our novel small molecule acceptors. The addition of IDFBR into P3HT binary devices increases the device efficiencies up to 7.7% by reducing the recombination, improving photo-voltage, charge carrier mobility and lifetime in the ternary blend (*Nat. Mater.* doi:10.1038/nmat4797 (2016)). This is the highest efficiency value reported for P3HT based solar cells. Careful optimization of the third acceptor component in a P3HT ternary blend resulted in optimal phase morphology where the vitrification of the crystalline IDTBR phase, preservation of crystalline P3HT phase and a molecularly dispersed mixed phase creates an optimum



energetic landscape for charge separation. This optimal phase morphology yields extended lifetime and reduced bimolecular recombination resulting in simultaneous improvement in V_{oc} , J_{sc} and FF for P3HT:IDTBR:IDFBR devices.

3 Non-photo active materials and tandem architecture development

ArtESun also focus on the development of novel transport, barrier and recombination interlayers based on solution processing. In addition, also ITO-free alternative electrodes were studied; On one side transparent and semi-transparent structures comprising silver grids, silver nanowires and highly conducting PEDOT derivatives, and semi-metallic Cr/Al/Cr and AZO/Ag/AZO. In the other side, highly bottom reflective systems made of thicker Cr/Al/Cr layers were also investigated. In all cases, the main objective was to increase the PCE of resulting devices in comparison to traditional ITO based PEDOT-Ca structures for different active polymers. Finally, joining results for all material development, a task was dedicated to modelling single and junction tandem devices with the aim of achieving PCE over 15% under relevant illumination conditions.

In this way, the **most important results** from this WP are itemized below:

- ✓ Novel alternative transport and recombination interlayers for standard and inverted cell designs^{1,2}:
 - Standard architectures: Novel interlayers based on **C60-adduct** pushed **PCE** to **6.7%** in comparison to reference ITO/PEDOT-Ca devices (5.5%) made with ArtEsun reference polymer (ARP).
 - Inverted architectures: Novel **glycin** and **5-ethyl-thiophene-2-carboxylic acid** based interlayers equalized reference devices (**5.5%**).
- ✓ ITO-free alternative electrodes gave promising results:
 - Ag grids/PEDOT structures yielded 3.5% (Figure 5)
 - Ag NW/PEDOT showed PCE of 2.7%.
 - AZO/Ag/AZO resulted in 4.6% PCE devices.
- ✓ Highly bottom reflective systems also improved the efficiency of standard reference devices:
 - **Cr/Al/Cr** devices produced **5.9% PCE**.
- ✓ Combined optical and electrical modelling showed that ArtEsun **tandem devices** can yield **PCE of 18.5%** under indoor LED illumination conditions (Figure 6).

¹ U. Würfel et al. *How Molecules with Dipole Moments Enhance the Selectivity of Electrodes in Organic Solar Cells – A Combined Experimental and Theoretical Approach*. Adv. Energy Mater. 2016, 1600594, DOI: 10.1002/aenm.201600594

² A. Spies, U. Würfel et al. *On the Impact of Contact Selectivity and Charge Transport on the Open-Circuit Voltage of Organic Solar Cells*. Adv. Energy Mater. 2016, 1601750, DOI: 10.1002/aenm.201601750



- ✓ The best experimental **tandem** device showed a **PCE of 10.6%** under AM1.5G illumination conditions (Figure 7).

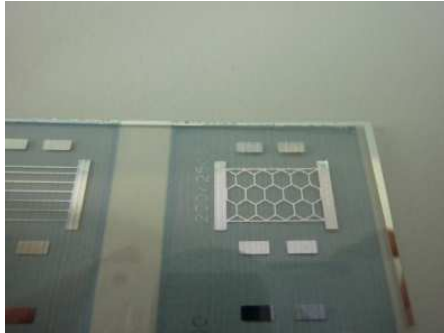


Figure 5 Novel ITO-free alternative electrodes based on silver grids and highly conducting PEDOT roll-to-roll printed onto PET flexible substrates.

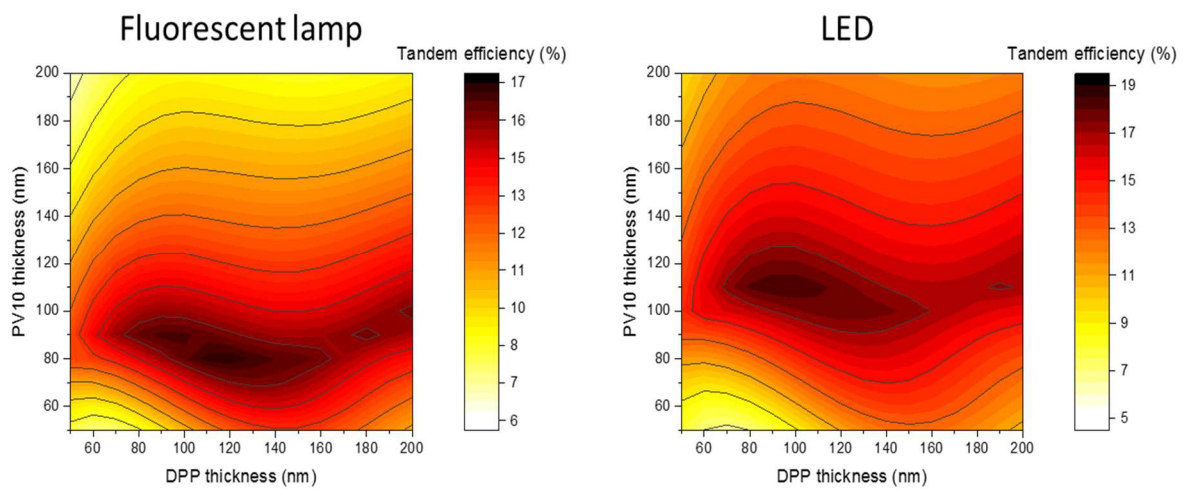


Figure 6 Main results from the combined optical and electrical modelling show that PCE can go beyond 18% under relevant illumination conditions for the experimentally developed tandem system.

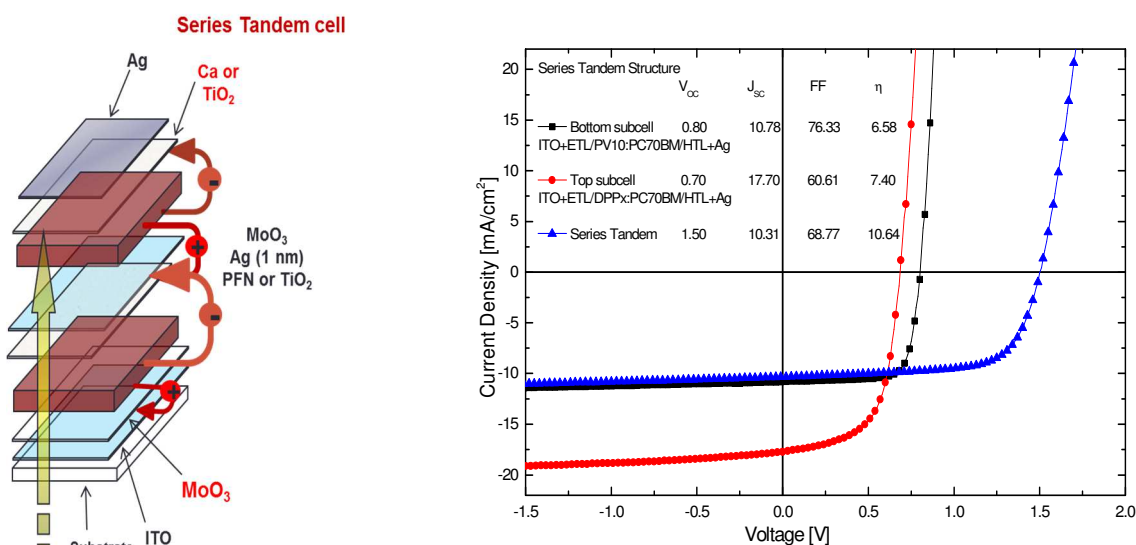


Figure 7 Design (left) and characterization (right) of the most efficient tandem cell (10.6%) fabricated within the project.

4 Printing and coating materials and processing development for R2R manufacturing of OPV modules

The main objectives related to device processing has been to develop and optimise low cost R2R printing (direct patterning) and coating processes for novel materials developed in the project, thus highlighting the environmental impact in terms of material use efficiency and safety and optimisation of energy consumption. The upscaling of process capacity to R2R production was accompanied by work on the lab scale for determination of process parameters. Experimental work was carried out with VTT, Fraunhofer, IMEC, Ikerlan and, closely connected to the demonstrators.

Specific goals can be summarized as:

- Formulation of environmental friendly printing and coating inks
- Electrical modelling of arbitrary shape OPV cells and modules
- Direct patterning (printing) deposition development for OPV layers and structures
- R2R slot die coating process development
- Self-assembled patterning of OPV modules
- Upscaling of R2R processing of OPV module production including cost calculations

Environmental friendly printing and coating ink formulations from photoactive materials were developed with the main focus on using non-halogenated environmental friendly solvents. The inks were designed according to the Hansen solubility parameters and boiling points considering the ink requirements for specific printing and coating techniques. Non-halogenated solvents are one step toward industrially viable printing of solution processed TFPV and solvent systems were based on thiophene, tetralin, 1,2,4-trimethylbenzene, anisole, and o-xylene. The general five-step evaluation was based on screening of solvents, examination of wetting and Hansen solubility parameters as well as preparation of lab-scale coating trials and device tests. Furthermore, the feasibility of the inks



containing novel photoactive materials and non-halogenated solvent was demonstrated as the upscaling of OPVs was realized in R2R process.

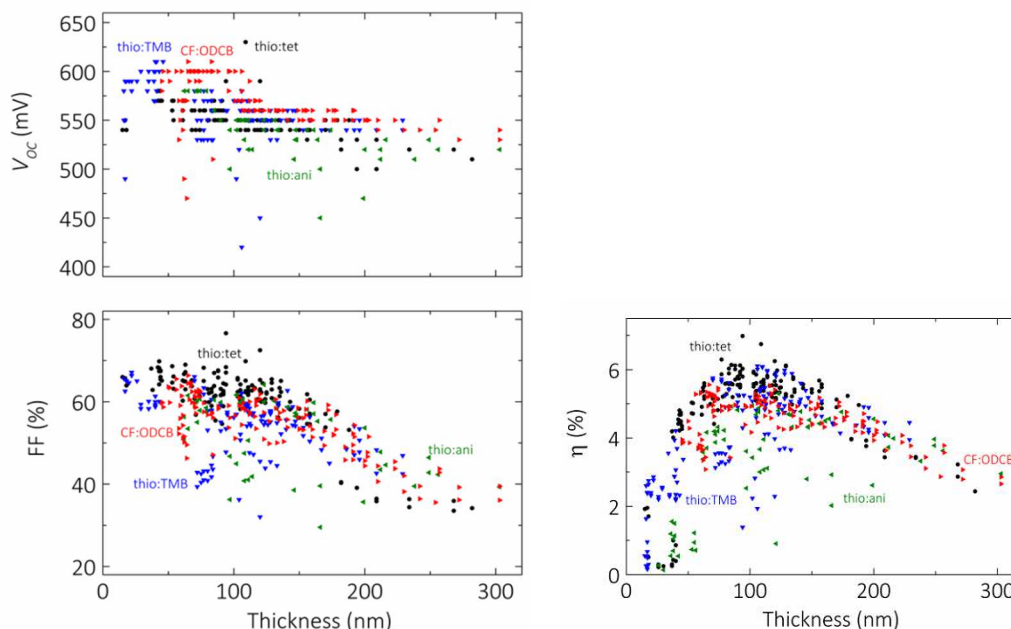


Figure 8 V_{oc} and FF vs. thickness of active layer, explaining the inability to utilize the interference maximum in current production at 230 nm of photoactive layer thickness.

The output power of a solar cell is depending on the incident light intensity and spectrum thus, it is important to know the expected lighting conditions and real JV characteristics under these conditions. For low light intensities a low parallel resistance can severely limit the performance, while its influence may be negligible under a full sun illumination. In addition, one important source of light is artificial lighting with about 500 lux light intensity that has been utilized as printed, arbitrary shaped modules for demonstrator case has been design using novel materials developed by Imperial College. Different direct and indirect methods printing methods were examined in order to pattern modules namely by using flexographic printing, screen printing, gravure printing, inkjet-printing or aerosol printing techniques. Based on these results the free form OPV modules were fabricated using gravure printing and screen printing techniques.

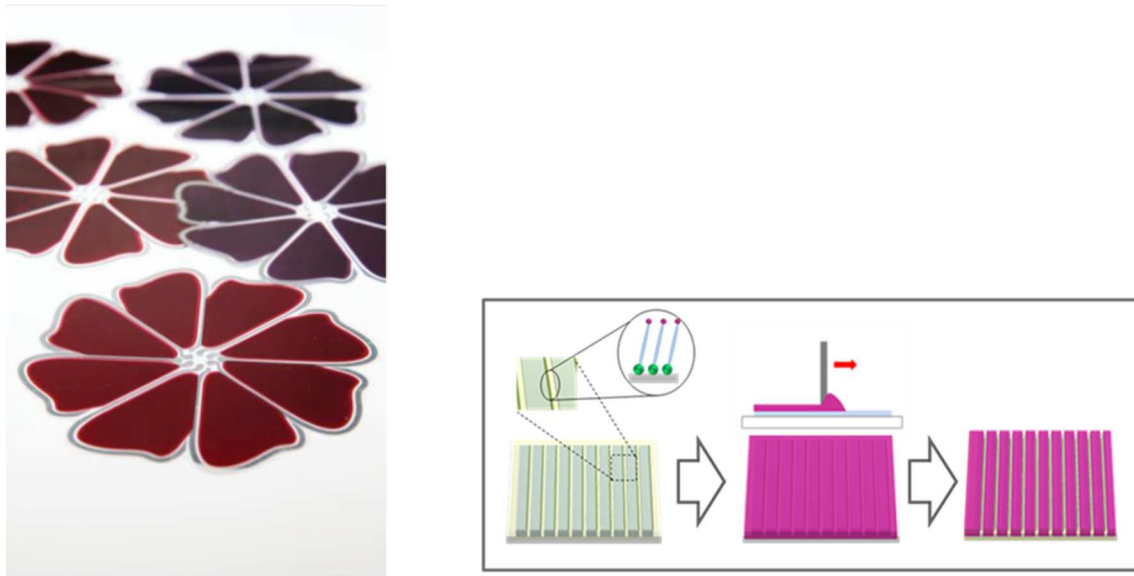


Figure 9 Patterning of OPVs using direct and indirect printing steps.

5 Evaluation of lifetime, encapsulation materials and processes

A separate work package was set up in order to demonstrate device processing using willow glass, optimize encapsulation strategies and identify the dominant degradation mechanisms of the OPV devices. The integration of flexible willow glass allows for a perfectly tight highly transparent front barrier preserving a certain degree of flexibility. A first important finding was uncovered by comparing degradation of thin films of Calcium and Benchmark OPV devices in different packages, like state of the art flexible barrier films and willow glass laminated with different glues as pressure sensitive adhesive tapes or filled epoxy glues. It could be clearly demonstrated that the main ingress of water and oxygen takes place through the adhesive or at the interface between adhesive and substrates (Figure 10). A clear advantage of a filled epoxy over PSA glues was observed especially at 85°C and 85% relative humidity, which is one of the IEC 61646 standard tests for thin film PV module qualification.



Figure 10 Result of the Ca test which clearly shows that main degradation channel is diffusion from the edges through the adhesive or at the adhesive/substrate interface.



The degradation of benchmark devices in the same packages was well correlated to the results of the Ca test. A large set of around ten photoactive materials and more than 15 different device stacks with and without ITO have been tested during the project, showing that each component influences the final device stability and that some materials are not compatible with certain device stacks, in which other function well and show best stability. The device optimization is therefore a highly complex task with a lot of interplay between materials and processing conditions. The goal was to identify a stable ITO-free device stack using a highly efficient organic photoactive layer which could be processed from non-chlorinated solvents in ambient atmosphere, to make the device compatible with low cost R2R production and suitable for real world applications. The best overall package was delivered by a novel non-fullerene acceptor ternary blend system which yields efficiencies above 5% even when processed in air from non-chlorinated solvents without using ITO and showed remarkable stability as shown in Figure 11. Other photo-active materials yielded higher efficiencies but degraded significantly faster or where not compatible with non-chlorinated solvents or air processing while others did not work properly in ITO-free devices. Only the newly developed material fulfilled all requirements at once [B. Zimmermann: *Efficient, large-area arbitrary shape solar energy*, European Photovoltaic Cluster General Assembly, 25th - 27th May 2016, Barcelona; B. Zimmermann: Highly Efficient and Stable ITO-free Organic Solar Cells - EU-Project ArtESun, Oral presentation at 2016 International Summit on OPV Stability, October 14, 2016].

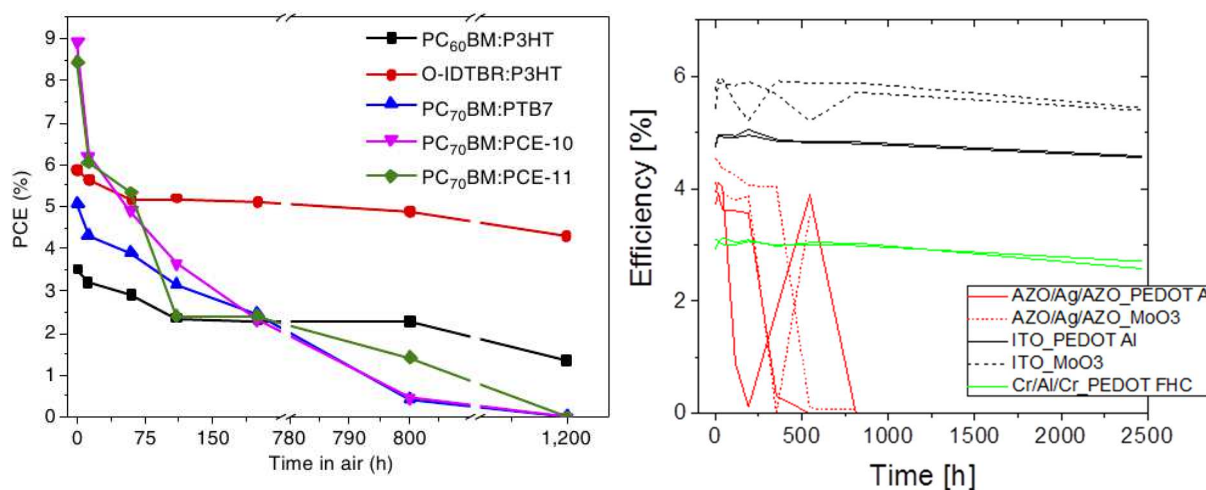


Figure 11 Stability of several highly efficient OPV-materials (left) and the novel ternary non-fullerene acceptor photoactive layer in different device stacks under continuous illumination (right). The material enables efficient and stable devices.

The second focus of the work package was to demonstrate processing of OPV devices directly onto Corning Willow glass. The handling of the thin glass needs a few precautions to avoid breakage, most importantly point impact must be avoided, but if this is carefully executed processing on the thin glass is reproducibly possible with the same quality as on thick glass. Figure 12 shows the JV curve and photos of the smartcard size OPV module for integration into the active RFID tag of Confidex, which works well at low light intensities [B. Zimmermann, (*Organic*) *Solar Cells for Indoor Applications*, invited presentation at MatHero Standardisation Workshop, 26.05.2016, Barcelona]. The rightmost picture shows the side view illustrating the very thin glass, the device shown is built on Willow glass and also encapsulated with Willow glass, at a total device thickness of 200 μm only. Roll-to-Roll handling of Willow glass was also investigated showing that minimum roller diameters should be 100 mm and that with a machine equipped like this, R2R processing of the Willow glass is indeed possible.

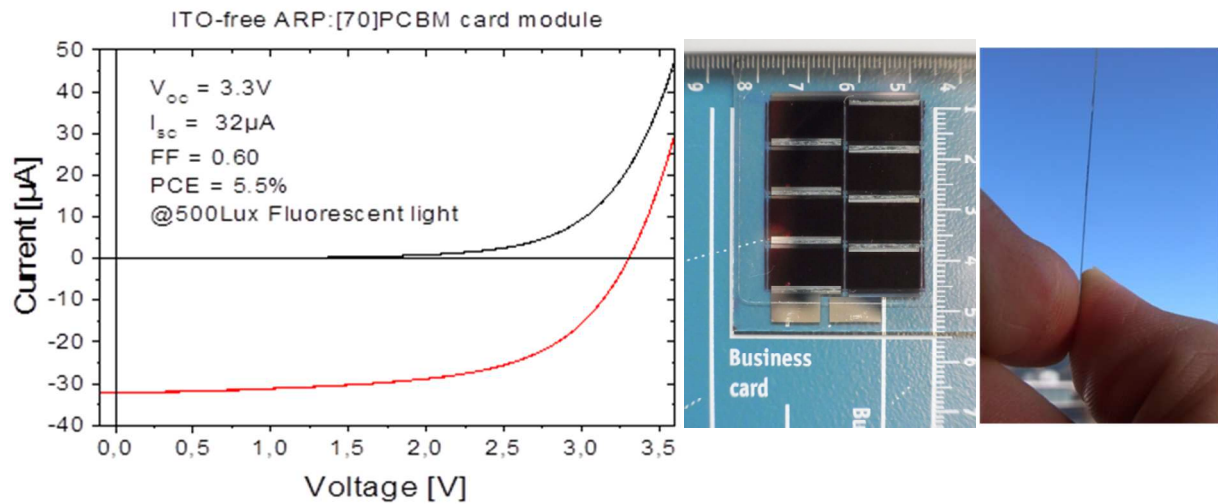


Figure 12 JV curve under 500 lux indoor illumination and photos of an OPV smartcard energy supply. The rightmost photo shows the of the device built on and encapsulated with 0.1mm thin Corning Willow glass seen from one edge.

The third main prototype related objective was to develop encapsulation strategies for the large scale BIPV Demonstrator. Two complementary approaches were developed. The first was to laminate a plastic film OPV module between two glasses to get a rigid and robust BIPV façade element as shown in Figure 13 left. Critical is the right choice of the encapsulant to ensure good adhesion. The second concept was to process the OPV directly onto a low cost flexible PV-Barrier Backsheet film and encapsulating with flexible Willow glass shown on the right hand side of Figure 13.



Figure 13 Photos of the large area Demonstrators, a plastic-film based module laminated into a BIPV façade element and a 30cm x 40cm large module processed onto a PV-barrier backsheet and encapsulated with 0.1mm thin willow glass.

6 Demonstrator prototypes

One of the key objectives were to produce three types of prototypes based on the ArtESun OPVs, where the applications is quite different, to demonstrate the flexibility and versatility of the technology.



6.1 RFID + OPV

Within the ArtESun project, it has been shown that the small rectangular OPVs of size 40 mm by 30 mm can successfully be integrated into the RFID tag together with auxiliary electronics including energy storage and overvoltage protection. This enabled the utilization of solar power as a power source for BAP operation and for powering sensor. Real measurements were conducted with the RFID-OPV prototype to evaluate the adaption to application requirements. It was measured that the RFID-OPV tag could achieve market acceptable performance level when it comes to vehicle identification outdoors and temperature sensor application indoors. The power utilized from the OPV could increase the vehicle read distance dramatically and the sensor information could be used for monitoring the cold chain for food and pharmaceuticals.

The RFID-OPV device shown below is essentially a flexible label, ready to be attached on both opaque and transparent platforms.



Figure 14 RFID-OPV device prototype with the label dimensions of 59 mm (W) x 90 mm (L) x 3 mm (T).

The temperature is read with a hand-held reader. This could represent the RFID-OPV operation in indoor sensor applications where the temperature is monitored. The outdoor vehicle identification is demonstrated by reading the label through a piece of car window glass with a fixed UHF RFID reader. The reading distance could be increased by a factor 10 when utilizing the solar power for BAP mode operation compared to passive mode operation.

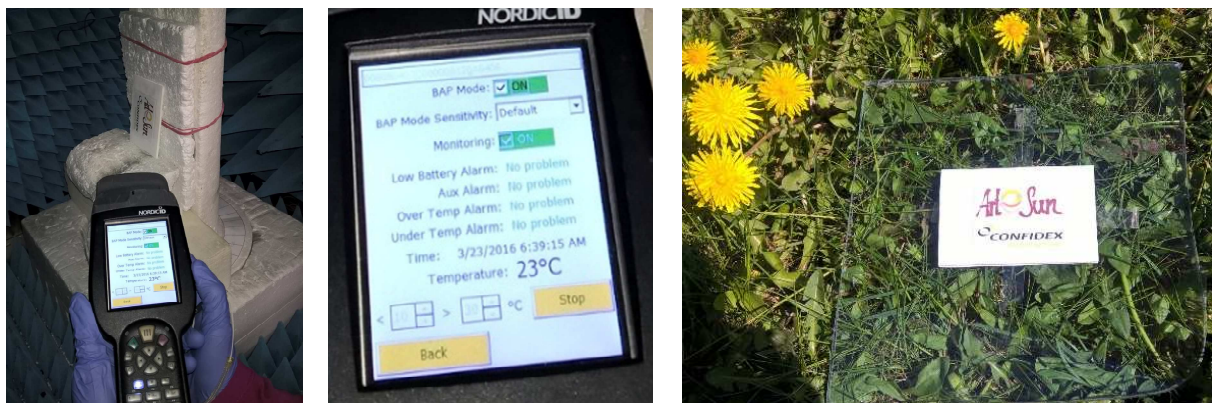


Figure 15 Indoor wirelessly temperature reading with use of a hand-held reader (left and middle) and outdoor long-range wireless identification of imaginary vehicle (right). Sensor communication and long-range reading are enabled with use of the solar power.

6.2 OPV + Antenna

It has been successfully demonstrated that an OPV having the shape of flower is directly integrated with a Bluetooth® low energy radio since the OPV is specifically designed to act as the antenna for the



wireless device. The OPV antenna has a diameter of 10 cm and provides enough electrical energy to continuously power the electronic circuitry indoors at low light intensity. The system comprises an environmental sensor measuring ambient temperature, humidity, and pressure, aiming at applications where precise weather monitoring is needed, such as in greenhouses. The Bluetooth® radio connects to a smart phone via a mobile app where sensor data can be read in real-time. The OPV wireless device has a range of 30 to 100 m. The OPV Smart Flower module is very thin and flexible, thus it can be stuck to an object, such as a bottle, or on a window. It radiates in all directions and can be easily deployed. Not only can the OPV Smart Flower be used for wireless sensing, but also as a broadcast beacon sending notifications or alerts, and for precise location tracking. With a Bluetooth® to WiFi or Ethernet hub, multiple devices connect to the internet and data is stored on a Cloud getting access to historical data and data analytics.



Figure 16 OPV antenna device prototype.

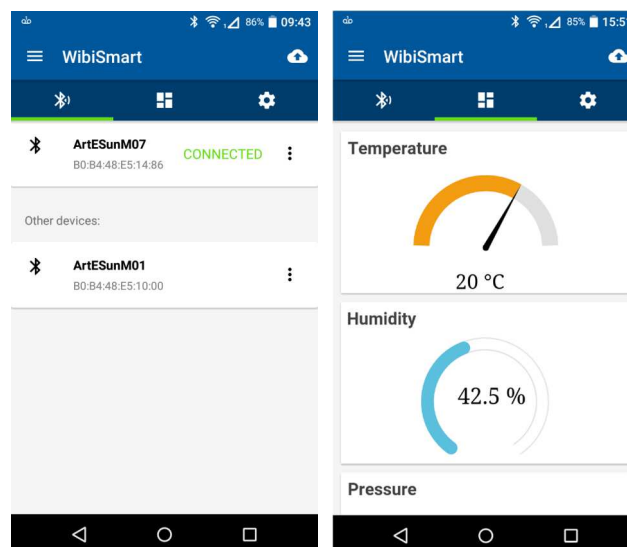


Figure 17 Using the WibiSmart™ mobile app from Wibicom, the OPV ArtESun device connects to a smart phone (left) and sensor data are visualized and updated in real-time (right).



6.3 Large Format BIOPV elements

ARTESUN project has allowed to prove that OPV fabricated by R2R can be laminated as a double glazing BIPV element within typical large scale manufacturability conditions.

In this case, a decrease in the OPV efficiency when transformed from cell to module was observed. This finding could be explained by means of the different measurement equipment (lab vs. industrial simulator), but most probably is linked to the limited contact surface between Ag paint/Al ribbons leading to an increase in Rs. Nevertheless, BIPV market acceptable efficiencies of 1,23% were obtained.

The BIPV element was developed as a vertical fin as large as 1610mmX380mm to be integrated as a ventilated façade within very well defined structural system. Potential market acceptability, in terms of overall subjective properties (robustness, colour, design, reflexion, etc.), was tested by means of a visual inspection experts' panel providing scores from 0 to 10. The result shows that overall a good acceptability of the product with ratings between 7-8 over 10.

Cost of ownership, actual and projected, for a BIOPV-based ventilated façade was compared to those found in first and second generation PV technologies. Overall it was found that BIOPV solutions can offer a suitable CoO that can build up a business case to the final client, being a promising technology on mid-term basis.

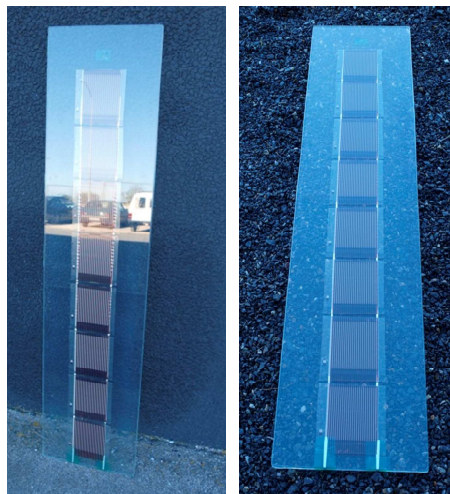


Figure 18 Outdoors view of a 1610mmX380mm BIOPV “fin”.



Figure 19 BIPV panels installed on the wall at Onyx in Ávila (Spain).

7 Summary

In conclusion, despite the very wide scope of the project all objectives were reached. As a final outcome three very different application prototypes were presented from the very small RFID to the large area BIPV application. Also a wide range of materials were synthesized and developed for different layers of the device.