



## PRODI

### WP1 Organic solar cell (OSC) manufacturing requirements

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Summary		
<p>This report describes the results of a literature and workshops carried out to generate and collect requirements on roll-to-roll (R2R) manufacturing equipment for organic solar cells (OSC). Based on the identified requirements, our task within the WP1 was to assess the applicability of different printing type manufacturing methods for producing organic solar cells. In doing so we first gathered information on the state of the art technical requirements for the fabrication of OSCs. This being done, we moved on to generating and defining future-oriented target requirements from same parameters and properties. Consequently, the present report provides state of the art and target requirements for the following OSC fabrication steps: substrate pre-treatments and converting, front side barrier deposition and curing, anode patterning and curing, hole injection layer patterning and curing, photoactive layer patterning and curing, cathode patterning and curing, contact pads and bus bars deposition and curing, and finally, adhesive printing and lamination. In case of each fabrication step, analytic discussion is provided to pinpoint the most fundamental problems, associated cause-and-effect relationships and potential solutions to overcome these identified technological challenges. As a conclusion, also the most potential production methods, including their advantages and disadvantages, per fabrication step are highlighted.</p> <p>As the key conclusion of the work done we find that the most important and challenging layers that require significant development work include barrier and electrode layers. The barrier layers protect the OSC layers from oxygen and moisture extremely well, thus maximizing and maintaining the cell lifetime and efficiency. However, the barrier properties (WVTR, OTR) are still rather poor as a result of which further material, deposition method, and layer structure developments are required. In addition, the transparency requirement of the front side barrier layer limits the material choice and impairs the barrier properties. The barrier layer should be homogeneous since any cracks or pinholes create a path to moisture and oxygen into the OSC structure. An additional challenge arises from the fact that the substrate onto which the barrier layer is deposited should be clean and smooth so that defect-free and homogeneous barrier layer is obtained. The non-vacuum R2R deposition makes also the achievement of the barrier properties challenging. The electrodes should be highly conductive to obtain high lifetime and efficiency of the OSC. Therefore, the electrode layers should be smooth, homogeneous, and stable. Electrodes might also require inert atmosphere processing, since they oxidise easily, thus diminishing conductivity. This places great demands for the R2R processing. The anode should also be transparent and have excellent resolution as well as sharp edges. The transparency and conductivity requirements require still material and process developments since highly conductive but non-transparent metals cannot be used. The cathode layer should be rather thick in order to protect the active layer from oxidation. Cathode inks are still under development since the lower work function of aluminium would be a more preferable choice than silver and commercial aluminium inks are not available. The final morphology of the cathode is determined in the sintering stage, thus making the sintering stage highly important. A thin electron transport layer might also be required under the cathode.</p>		
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## Contents

1	Organic solar cells technology overview .....	4
1.1	State of the art .....	4
1.2	Applications .....	4
1.3	Component structure.....	5
1.4	Manufacturing process description .....	6
2	Organic solar cell manufacturing requirements .....	8
2.1	Technological requirements and challenges .....	8
2.2	Production process requirements and challenges.....	9
2.2.1	Substrate pre-treatments and converting .....	9
2.2.2	Front side barrier deposition and curing.....	10
2.2.3	Anode patterning and curing.....	13
2.2.4	Hole injection layer patterning and curing .....	17
2.2.5	Photoactive layer patterning and curing .....	19
2.2.6	Cathode patterning and curing.....	21
2.2.7	Contact pads and bus bars (metallization) deposition and curing .....	24
2.2.8	Adhesive printing and lamination .....	26
3	Conclusions .....	29
	References .....	31

# 1 Organic solar cells technology overview

## 1.1 State of the art

Low cost organic photovoltaic (OPV) have been an object of intensive research during the last 10 years. Especially, due to its promise of low-cost manufacturing by high volume production methods such as printing and coating on flexible substrates, OPV has a powerful potential to become a novel optoelectronic device using as renewable energy source. Solution-processed solar cells are typically based on a conjugated polymer as a donor material blended with a soluble fullerene derivative as an acceptor material.

By spin-coating technique, the power conversion efficiencies of polymer-based bulk heterojunction (BHJ) solar cell, poly-3-hexylthiophene (P3HT) as a donor and phenyl-c61-butyric acid methyl ester (PCBM) as an acceptor, have reached up to 4-5 % under 1.5 AM (100 mW/cm<sup>2</sup>) illumination [10,11]. The non-tandem based organic photovoltaic device with active layer consisting of an alternating co-polymer--poly[N-9"-hepta-decanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole) (PCDTBT) in bulk heterojunction composites with the fullerene derivative [6,6]-phenyl C70-butyric acid methyl ester (PC70BM) has lead to an improved power conversion efficiency 5.5 % [12]. In order to improve the efficiency further, stacking of low and high band gap materials into a tandem solar cell has been reported. Currently, the highest reported power conversion efficiency is 6.5 % [13]. 233 cm<sup>2</sup> module processed by spin coating having 1,1 % total area efficiency have been developed by Plextronics [14]. The disadvantage of spin-coating technology is the high material loss and incompatibility with high volume roll-to-roll processing.

It has been reported that several printing and coating methods are suitable for processing organic solar cells, including gravure printing, inkjet printing, screen printing, pad printing, doctor blading, slot die coating, spray coating etc. Ding et al. have obtained a power conversion efficiency of 1.68 % with organic solar cells containing rotogravure printed hole injection layer and active layer with an active area of 0.03 cm<sup>2</sup> [15]. There has been also development on inkjet side and increased efficiencies up to 3 % have been reported [16-18]. Krebs et al. have introduced pad printing as a technique for preparing the active layer [19]. Screen printing, knife-over-edge coating and slot-die coating have been combined in order to prepare an organic solar cell module with an active area of 75 cm<sup>2</sup>. [20] Giroto et al [21] and Hoth [22] et al have demonstrated the feasibility of spray coating to process organic solar cells. Power conversion efficiency ~3 % has been reached with spray coated active layer with an active area of 20 mm<sup>2</sup>. [21,22]. In addition, doctor blading [23] and screen printing [24] have been successfully used for preparing organic solar cells. Recently also an extensive review of OPV processing technologies have been published [5].

## 1.2 Applications

Organic solar cells are produced by utilising roll-to-roll manufacturing methods capable to mass production that is attractive solution from cost standpoint. Although the efficiency, device stability and lifetime do not reach the values obtained with inorganic based solar cells, there is market for organic solar cells. The main benefit of organic solar cells produced with roll-to-roll manufacturing methods is that it requires less capital investment in comparison to Si-based devices. [25]

In EC funded EURO-PSB project, flexible, a self-rechargeable polymer solar battery was developed. In addition, potential market applications have been evaluated which are summarised in Table 3. As it can be pointed out, the main parameters affecting the requirements for the solar cell module to be used in certain application are operational voltage and current, typical working duration per day/week, need for energy storage, conditions for light exposure including indoor/outdoor applications and requirements for the lifetime. [26]

Table 1. Potential market applications for organic solar cells. [26]

[1]	[2]	[3]	[4]	[5]	[6]	[7]	[8]
Power range	Product description	Voltage	Running current	Typical use/day	Energy consumption/day	Storage needed	Conditions of light exposure
$\mu\text{W h/day}$	Basic calculator	1.4 V	10 $\mu\text{A}$	0.5 h	7 $\mu\text{W h}$	(NO)	Indoor/drawer
	Kitchen weighting scale	3 V	20 $\mu\text{A}$	10 min	10 $\mu\text{W h}$	NO	Indoor/cupboard
	Simple wrist-watch	1.5 V	0.4 $\mu\text{A}$	24 h	15 $\mu\text{W h}$	YES	Indoor/outdoor
	TV remote control	3 V	10 mA	1 min	500 $\mu\text{W h}$	YES	Indoor/drawer
	Alarm clock	1.5 V	Clock + ringing	24 h	800 $\mu\text{W h}$	YES	Indoor
$\text{mW h/day}$	PC keyboard (low power)	3 V	4 mA	1 h	12 mW h	YES	Indoor
	PC mouse (low power)	3 V	8 mA	2 h	48 mW h	YES	Indoor/under the hand!
	PDA	3 V	15–75 mA	20 min	15–75 mW h	YES	Indoor/pocket
	Small radio	1.2 V	15–150 mA	1 h	20–180 mW h	YES	Indoor/outdoor
	LEDs torch	3.6 V	20–100 mA	1 h	72–360 mW h	YES	Any
	Mobile phone	3.6 V	200 mA	0.5 h	360 mW h	YES	Indoor /pocket
$\text{W h/day}$	Mobile phone	3.6 V	200 mA	3 h	2160 mW h	YES	Indoor/pocket
	Laptop (personnal computer)	12 V	3 A	1 h	36 W h	YES	Indoor
	Energy pack	3/6/ 12 V	Personnal ultralight charger			YES	Outdoor

Possible applications for organic solar cells can be divided into four categories which have different requirements for the performance and lifetime:

- Category I: Low efficiency solar cells, short lifetime (1.5-3 years), mainly for indoor usage (consumer electronics)
- Category II: Low efficiency solar cells, longer lifetime than category I (3-5 years) outdoor usage
- Category III: Higher efficiency than with categories I and II, higher requirements for lifetime (5-10 years)
- Category IV: High efficiency, long lifetime (> 10 years), outdoor usage, organic solar cells for energy production in buildings (installed in the roofs of the buildings)

### 1.3 Component structure

Polymer solar cells compose typically of multilayer structure as shown in Figure 4. To simplify, an organic solar cell composes of following layers: substrate, anode, hole injection layer, active layer, cathode. Because the active materials are sensitive to moisture and oxygen, barrier materials should be deposited on both sides, front and back side, of the structure. Hole injection layer and active layer are sandwiched between two electrodes, anode and cathode. Transparent conductive oxides (TCO) such as indium tin oxide (ITO) are typically used as anode material. Due to the limited conductivity of TCOs and the high price of the material, alternative layer materials have been investigated. One of the approaches is to use high conductivity PEDOT and metal grid layer to replace the TCO. The hole injection layer (HIL) smoothens the anode layer and enhance the OSC performance by creating a proper interfacial contact between the anode and photoactive layers, i.e., facilitating the charge transport. The most commonly used structure is bulk heterojunction device architecture in which the active layer consisted of blending two organic semiconductors having donor and acceptor properties in solution. In this deliverable, it is assumed that the cathode is prepared with roll-to-roll printing or coating methods. In order to improve the charge transport to the top electrode, possible interlayer will be prepared between the active layer and cathode layer. In addition, contact pads/lines will be prepared in order to get a contact to electrodes and connect OPV modules/component together. There are also more advanced structures; one of those is a tandem cell structure in which both high and low bandgap materials are stacked.

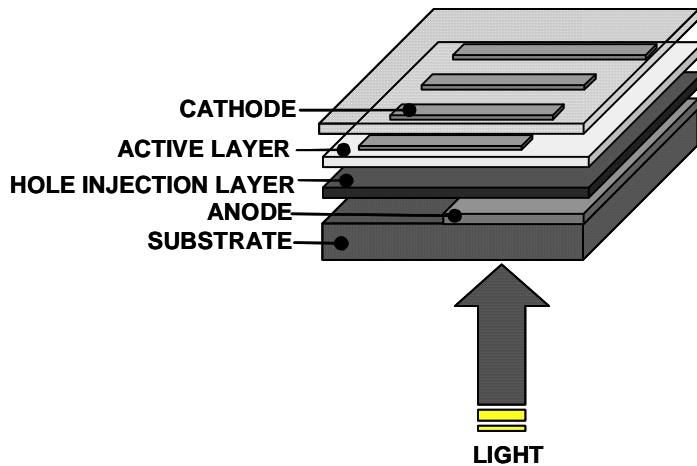


Figure 1. Structure of single OPV component.

Single organic solar cells produce output voltage at maximum  $\sim 0.5$  V. However, in most of the applications, higher voltages is needed, so single organic solar cells need to be connected in series in order to produce higher output voltages. In series connection the anode of the first cell is connected to the cathode of the next cell as shown in Figure 2. The output current is directly proportional to the active area so in order to increase the produced electrical power the active area needs to be increased. Parallel connection can be used in order to improve the reliability of the solar cell module. If one single solar cell fails, the module still works if parallel connection is used in the design of solar cell module.

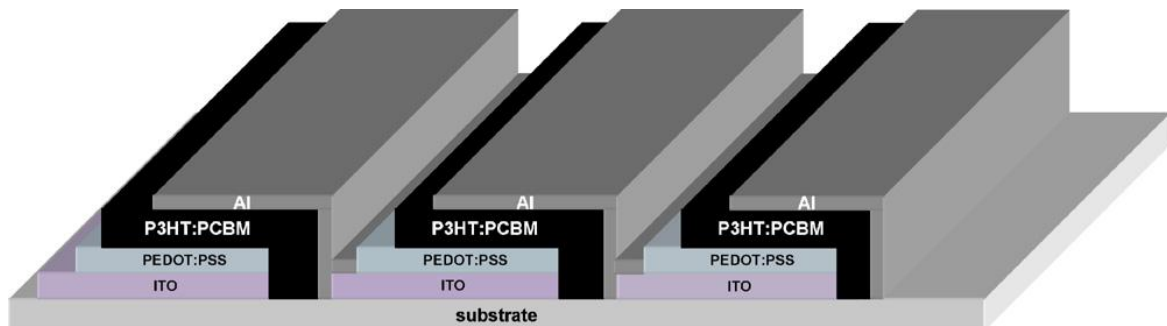


Figure 2. Series connected organic solar cells prepared on one substrate. [27]

## 1.4 Manufacturing process description

As already described in the previous chapter, the organic solar cell is a multilayer structure and thus different roll-to-roll printing or coating methods can be used for separate layers. In this chapter, a summary of the OSC manufacturing process will be described. More detailed analysis will be presented in Chapter 3. In roll-to-roll OSC manufacturing process, flexible substrate will be used as a basis. Cleaning, deionization and corona/plasma treatment will be done before the barrier layer deposition. The objective of cleaning and deionization is to remove particles and inhibit new particles to stick on the surface of the substrate. Corona/plasma treatment increases the surface energy of the substrate. As a first layer, a barrier is deposited on top of flexible substrate by coating methods since no patterning is required. Since the active materials and cathode materials are sensitive to water and oxygen, barrier inhibits those reagents to penetrate to the active regions. The barrier is dried either with hot air or UV. Registration marks will be prepared for all the layers prepared after the barrier layers since the active areas need to be aligned to each other in order to process patterned structures. Especially due to the high transparency of anode and hole injection layer, the recognition of registration marks sets challenges to the measurement equipment. On top of barrier, anode will be prepared with roll-to-roll printing methods since the anode needs to be patterned. The printed anode layer will be dried with blowing hot air or by UV. Next, hole injection layer (normally PEDOT:PSS is used) is processed by printing methods such as gravure, flexography or ink jet printing and dried with

hot air. The photoactive layer is prepared with gravure, flexography or ink jet printing method due to the low solid content of the photoactive ink. Due to the sensitivity of active materials to UV, hot air will be the choice for the drying method. The top electrode i.e. cathode will be most likely to be prepared with screen printing due to the high solid content and viscosity of the cathode ink and dried with hot air. As a next step conductivity lines will be prepared in order to have contacts to anode and cathode and connect organic solar cell modules to each other. The OPV components/modules/systems will be encapsulated by using a back side barrier layer which is laminated to the plastic foil composing of other processed layers. Finally, the OPV modules/components/systems will be cut into pieces.

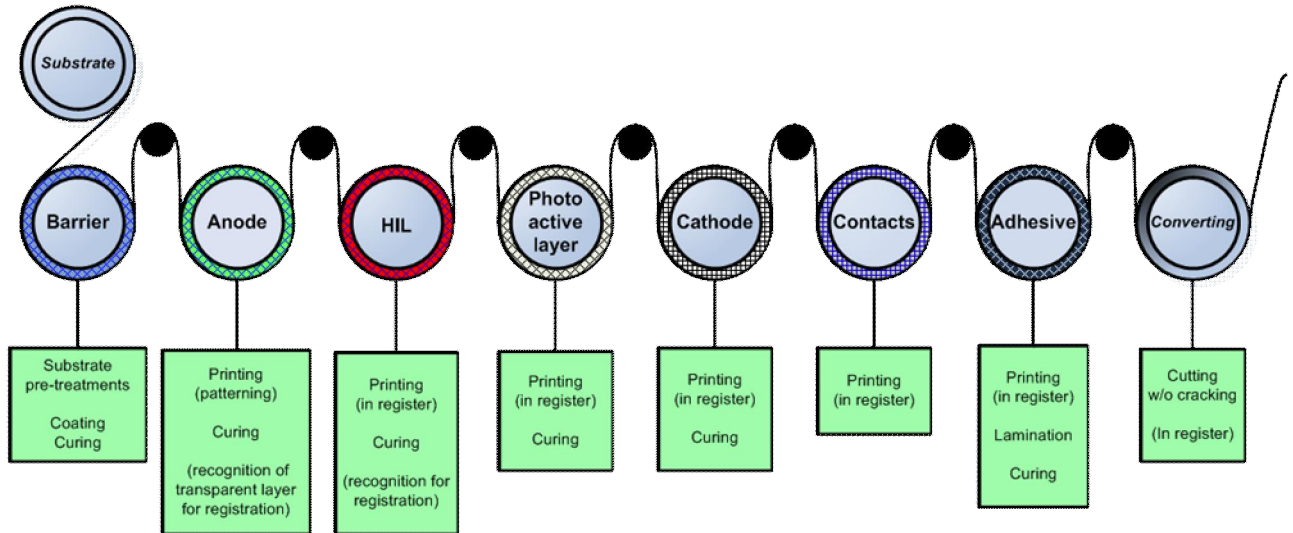


Figure 3. Process steps of OPV component manufacturing.

## 2 Organic solar cell manufacturing requirements

### 2.1 Technological requirements and challenges

Low cost organic photovoltaic (OPV) have been an object of intensive research during the last 10 years. Low cost can be obtained by producing organic solar cells by roll-to-roll production methods leading to mass production of organic solar cells. However, low performance and poor stability are issues that need to be solved before organic solar cells can hit into energy markets in large scale. In addition, the material costs need to be reduced and development of materials is required in order to diminish the environmental impact of toxic materials and solvents and fulfil the environmental compliance. In addition, since one of the targets is to utilise organic solar cells in packaging industry and other consumer products, the products are required to be consumer safe. Cell efficiency has a moderated impact and monolithic cell integration slight impact on low cost solar cells.

The total lifetime of the organic solar cells is highly dependent on the shelf lifetime and operational lifetime of OPV component. In addition, the ink and material stability, especially in the case of the cathode and photoactive materials, is one of the most impacting factors which determines the total lifetime of OPV component. Furthermore, the mechanical strength has moderate effect on OPV lifetime. However, the requirements for the mechanical strength are dependent on the chosen application. Additionally, the durability of the monolithic integration affects the total lifetime.

Cell encapsulation is one of the main parameters that affect significantly on total efficiency of produced system. The development of barrier materials is needed in order to obtain a lifetime at least 1.5 years for the consumer electronic applications. Cell efficiency of separate cell either with low or high illumination affect the total efficiency obtained with whole system. The active layer of OPV device absorbs photons in certain wavelength region, so the efficiency is highly dependent on the active layer thickness. In addition, ink and material stability is of high importance when considering total efficiency. The objective is to use non-toxic materials and solvents which have moderate impact on total efficiency. The monolithic cell integration has moderate impact on total efficiency. In addition, OPV mechanical strength has slight effect.

Since the application is not defined more precisely in this report, the organic solar cell component/modules of large and small areas can be handled. Roll-to-roll processability, cell encapsulation and ink and material stability have a dominant impact on mass production of large area and small area organic solar cells. Furthermore, the total OPV thickness affect moderately on mass producible organic solar cells. With small area devices, OPV mechanical strength has a moderate effect on mass production. However, in larger areas the impact of mechanical strength is dominant. With small area devices, the components need to be monolithically integrated to other components.

The organic solar cells need to be easily integrated in other different devices in order to utilise the electrical power produced from the organic solar cells. Ease of system integration is strongly related to OPV mechanical strength and moderately related to monolithic cell integration, cell encapsulation and total OPV thickness.

## 2.2 Production process requirements and challenges

### 2.2.1 Substrate pre-treatments and converting

The plastic substrate should be as clean as possible when entering the OSC manufacturing process. Therefore, several pre-treatment steps are made to fulfil the strict cleanliness requirements of the substrate. The substrate is typically first washed with water and/or solvents, such as acetone and alcohols, in an ultrasonic bath to detach and rinse away contaminants (e.g. dust, grease, debris) from the surface. The washing liquid is then evaporated. After the cleaning, static and other charges that attract contaminants and decrease runnability are removed in the de-ionization stage. These charges tend to re-develop over time. In the final pre-treatment stage, the surface energy of the substrate is increased using corona or plasma treatment to allow proper wetting, spreading, and adhesion of the material deposited in the following stages of the manufacturing. At the same time, dust is removed from the surface.

After the R2R manufacturing process of the OSC modules, converting is also necessary. Solar cells or modules are separated from each other through die cutting or slitting and cutting. After this, the cells are conveyed from the process to be packed or used. If the cells are not separated, the web material can be rewound. Converting stage might also include some cleaning, such as blowing off dust and other contaminants from the surface of the OSC.

Table 4 presents the basic properties and requirements of the plastic foil substrate. The plastic foil should be smooth and have homogeneous properties to establish a proper base for the deposition of the OSC and to maximize the cell performance. The high transparency of the plastic foil (> 80 %) ensures that light enters sufficiently into the active parts of the OSC. The surface energy is typically 20-50 mN/m but it is increased via corona/plasma treatment to allow easy deposition of the following layers. The plastic substrates withstand only temperatures that are lower than 200°C, thus this limitation needs to be taken into account in the whole OSC manufacturing process.

*Table 2. Basic properties of plastic substrates.*

Properties	Value	Target	Unit
Thickness	< 300	-	µm
Roughness, $R_a$	< 50	< 10	nm
Surface energy	20-50	> 50	mN/m
Transmission	> 80	> 90	%
Max. temperature	< 200	-	°C

The plastic foil should go through the whole OSC manufacturing process as smoothly as possible without destruction. Therefore, its runnability and stability should be maximized and static electricity minimized. Other factors affecting the R2R processability are the thickness and surface properties of the plastic foil since they influence the runnability and deposition processes, respectively.

The plastic foil forms the majority of the total thickness of the OSC. Therefore, the encapsulation quality depends significantly on the plastic foil thickness because of the fact that thicker films prevent gas permeation to a greater extent. The thickness affects also the converting processes. The high transparency of the substrate ensures maximum light entrance into the active layers of the OSC, and the high stability plays a significant role in the formation of the mechanical strength of the OSC.

Although the pre-treatment and converting stages form a base for the OSC component, they are not so important stages considering the whole manufacturing process. This results from the fact that these stages are rather optimized and highly effective. Only poor cleanliness can decrease the cell performance by hindering the homogeneous deposition of the layers of the OSC and charge generation.

## 2.2.2 Front side barrier deposition and curing

### 2.2.2.1 Layer materials and properties

The organic materials and metal electrode materials are sensitive towards oxygen and moisture, so therefore a barrier layer is needed in the front and backside of the OPV device. Two parameters are defined in order to estimate the transmission of oxygen and moisture through the protecting layer, oxygen transmission rate (OTR) and water vapour transmission rate (WVTR). In order to reach a lifetime of 10000h, it has been estimated that the WVTR should be at room temperature at  $10^{-6}$  g/m<sup>2</sup>/day and OTR  $10^{-3}$  cm<sup>3</sup>/m<sup>2</sup>/day. [28]

Front side barrier layer can be formed of single layer of oxides (SiO<sub>x</sub>, AlO<sub>x</sub>), nitrides (SiN<sub>x</sub>) or oxynitrides (AlO<sub>x</sub>N<sub>y</sub>). The main challenge in the formation of single layer barrier is that it has to be defect free in order to gain needed OTR and WVTR values. In order to decrease the paths for the moisture and oxygen to penetrate through the barrier, multilayer structures have been developed in which organic (polymer) and inorganic layers are stacked as shown in Figure 4. One option for the materials is to stack organic polyacrylate (PA) and Al or AlO<sub>x</sub>. [29] In addition, the use of inorganic and hybrid (inorganic-organic) polymer such as silicate based materials has been reported. [30] As a front side barrier, the barrier layer needs to be as transparent as possible to the light since the absorption occurs in the active layer of the OPV device. In addition, the difference in refractive index between different layers should be low in order to avoid reflection losses. The cleanliness of the processing environment is important from the viewpoint of barrier functionality, since it has been noticed that the cleanliness of the processing environment correlates to the amount of cracks in the barrier film.

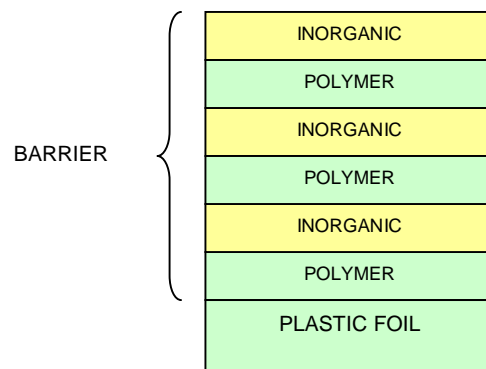


Figure 4. Schematic illustration of multilayer barrier structure composing of stacked inorganic and organic (polymer) layers.

Table 3. Basic properties and requirements for the front side barrier layer.

Properties	Value	Target	Unit
Solids content	-		wt-%
Viscosity	-		mPa·s
Surface tension	-		mN/m
Thickness	-	> 10 µm, dependent on the requirement for OTR and WVTR	nm
Roughness	-	< 10 nm	%
Transmission	-	> 90 %	
WVTR	With conventional methods: 10-6 g/m <sup>2</sup> /day with 3-5 PA-AIO <sub>x</sub> pairs (multilayer)	Depends on application Lifetime of 10000h at RT-> 10 <sup>-6</sup> g/m <sup>2</sup> /day	
OTR	-	Depends on application Lifetime of 10000h at RT-> 10 <sup>-3</sup> cm <sup>3</sup> /m <sup>2</sup> /day	

#### 2.2.2.2 Suitable deposition and drying methods

As already discussed, multilayer barrier structure consists of stacked inorganic and organic layers. Since there is no need for patterning the barrier layer, **coating methods** such as knife over coating, doctor blade coating, air knife coating etc. are suitable for deposition of the barrier layer. The coating method should be chosen considering the rheological properties of ink of the inorganic and organic materials used in the barrier layer. Coating methods can be classified to three categories which are presented in Table 6.

Table 4. Coating method classification.

Category of coating methods	Examples of the coating methods belong to the category	Characteristics
Self-metered	- Dip roll - Nip forward roll - Reverse roll	- Wet thickness is determined the conditions of the coating meniscus
Doctored	- Mayer rod - Blade/knife - Air knife - Dip & scrape	- Post applicator device determines the wet thickness
Pre-metered	- Slot die - Slide curtain - Spray	- All the ink fed into an applicator is transferred to the web

In dip coating, the web is immersed in a tank of the coating material for a certain period of time. The excess material is transferred by passing the web through nip rolls or doctor blades. In knife coating techniques, the coated ink is applied directly to the substrate and spread in a uniform manner by means of a fixed knife. Knife over coating method is especially used for high viscous inks. In air knife coating, the blast of air is used to blow off the excess ink. Thus, the viscosity is much lower than in the case of knife over coating. Knife coating methods, the properties of the knife and the support roller and the gap between the knife and the substrate play a key role in the formation of uniform barrier layer. In pre-metered methods, all the ink which is fed into an applicator is transferred to a web forming a wet coating.

Since there are a high variety of barriers materials, the certain coating methods for the barrier materials can not be defined. Therefore, in Table 7, the advantages and disadvantages of coating methods in general are summarised considering the requirements set by the process of barrier layer.

Table 5. Advantages and disadvantages of coating methods.

Printing method	Advantages		Disadvantages
Coating	<ul style="list-style-type: none"> <li>- Large uniform coated areas</li> <li>- Defect free layer</li> </ul>	<ul style="list-style-type: none"> <li>- Layer uniformity</li> <li>- Large area printing</li> <li>- High viscosity range (different methods can be used)</li> </ul>	<ul style="list-style-type: none"> <li>- Material consumption</li> <li>- No patterning possibility</li> </ul>

### 2.2.2.3 Production process challenges

The operating and shelf lifetime of OPV component are highly dependent on the properties of the barrier film. WVTR and OTR are key parameters that define the properties of the cell encapsulation. In addition, these parameters affect on the cell efficiency since the active materials and metal electrode materials are sensitive to oxygen and moisture. The barrier needs to transparent (>85 %) as possible to avoid reflections of light. Low transparency diminishes the cell efficiency of the OPV component since part of light does not exceed the active layer in which the absorption should occur.

Barrier surface properties in terms of low amount of impurities, low surface roughness affect the cell encapsulation. In addition, the thermal and mechanical properties of the barrier have an effect on encapsulation as well. The solvents for the barrier ink should be chosen in order to take into account the plastic foil beneath and the layers processed on top of the barrier. The layers and materials need to be chemically compatible i.e. the layer processed on top cannot dissolve the layer beneath.

The properties of the flexible substrate i.e. plastic foil play a key role in a formation of defect free, homogenous barrier layer. The surface roughness (rms) of the plastic foil should be less than < 1nm and free of spikes greater than tens of nanometres. [29] In addition, the surface should be free of impurities. The surface energy of the plastic foil needs to be higher than the surface tension of the inorganic barrier material ink in order to wet the ink properly on the surface. In addition, the flexible substrate needs to withstand the solvents used in barrier material inks. Furthermore, the mechanical properties of the plastic foil and thickness of the plastic foil are of importance since no cracks are allowed in the barrier layer.

Nowadays, the single layer and multilayer barrier films are deposited with low pressure gas phase technologies such as sputtering, evaporation and plasma enhanced chemical vapour deposition (PECVD). These processes require costly equipment and the used systems are very complex, so there is a need for development of low cost production methods for barrier layers. [29] The main challenge in the wet deposition methods is the R2R printing/coating of uniform layer without any cracks and pinholes which form a path to moisture and oxygen to penetrate to the OPV structure. This is one of the main challenges in the whole OPV manufacturing process, since the encapsulation is dominant factor affecting the shelf and operating lifetime of the OPV component. The coating method should be chosen according to the ink properties. Since the barrier layer consisted of several layers, different coating/printing methods can be used for organic and inorganic layers. Due to the high requirements for homogeneity of the barrier layer (OTR and WVTR), a lot of effort is needed for the optimisation process of the ink formulation and printing/coating processes. The layer thickness of the barrier layer and ink properties in terms of chemical compatibility, viscosity and surface tension correlate directly to the WVTR and OTR values. Furthermore, the layer thickness of the barrier layer should be optimised. If the barrier layer is too thick, it might form mechanical stress and thus cracks to the barrier layer. The barrier layer properties in terms of chemical compatibility, surface roughness, and surface energy affect the ink properties of the anode layer.

Hot air drying is the main method to cure the stack of inorganic/organic barrier layers. In addition, UV curing could be used for the front barrier layer since no UV sensitive active layers are present in the structure in this process phase. The drying temperatures should be chosen taken into account the thermal properties of the barrier material and the thermal stability of the plastic foil. Drying is an important processing step, since too high temperature easily causes cracks on the barrier layer. The main factor affecting the drying time is the thickness of the wet film. The layer should be dry in order to avoid oxygen and moisture to penetrate to the other layers processed as a next step. The challenges and solutions are summarised in Table 8.

Table 6. Challenges and solutions for the front side barrier layer.

Challenges	Solutions
<ul style="list-style-type: none"> <li>- Uniformity (pinhole free, no cracks) of the barrier layer</li> <li>- Suitability of coating and printing methods (non-vacuum processes) for processing barrier layers with high requirements for OTR and WVTR</li> </ul>	<ul style="list-style-type: none"> <li>- Barrier material development for non-vacuum processes</li> <li>- Ink and drying procedure optimisation</li> </ul>

## 2.2.3 Anode patterning and curing

### 2.2.3.1 Layer materials and properties

The anode layer made of indium tin oxide (ITO) is typically deposited using sputtering or evaporation followed by a separate patterning stage, such as etching or embossing. Therefore, additive patterning, such as printing, would be a more preferable deposition method in decreasing the number of process steps. ITO nanoparticles are dispersed into various alcoholic or aqueous solvents with small amounts of binders and additives. [31-33] The ITO content is 40-60 wt-%, and the drying and flow properties of the anode ink are adjusted by using at least two different solvents having different rates. In order to maximize both the printability and conductivity of the anode, the amount of binder and additives should be optimized. Due to the high cost and brittleness of the ITO layer and the poorer print quality than required, significant ink and process developments have to be performed.

The viscosity of the anode ink is 10-70 mPa·s depending on the nanoparticle content. [31-33] The ink formulation needs to be optimized to prevent both excessive spreading and poor levelling. High conductivity and transparency of the anode ensures a high OSC performance. However, the anode conductivity is often rather low (0.5-5 kΩ/sq) due to the porous nature of the nanoparticle layer although nanoparticles decrease the curing temperature and light scattering and increase the resolution. The transmission of the anode should be over 80 % to ensure proper light absorption into the photoactive layer of the OSC. By using inert atmosphere during the deposition, the conductivity improves but the R2R processability might suffer.

The anode layer should be smooth, uniform, and homogeneous to maximize the conductivity and transparency and to decrease the amount of short circuits. [31-33] The optimum layer thickness is 500-800 nm but it might be difficult to achieve at one pass when using low viscosity inks. In addition, the pattern should have excellent resolution (10-40 μm) and register as well as sharp edges. However, these requirements are not yet reached with printing since the maximum print resolution is currently around 40-100 μm. The surface energy of the ITO anode layer is approximately 40 mN/m but it is increased to 60-70 mN/m using corona/plasma treatment to enable proper deposition of the following hole injection layer. Table 9 shows the properties of the ITO anode and the future targets.

Table 7. Properties and requirements for the ITO anode ink and layer.

Properties	Value	Target	Unit
Solids content	40-60	-	wt-%
Viscosity	> 10	-	mPa·s
Surface tension	40	60-70	mN/m
Thickness	100-1000	500-800	nm
Roughness	> 100	< 5	nm
Transmission	> 80	> 90	%
Resistance	0.5-5	< 0.05	kΩ/sq
Resolution	40-100	10-40	μm

Another approach for the transparent anode is to pattern a silver metal grid and deposit a highly conductive and transparent organic polymer layer on top. [33-36] The metal grid increases the conductivity of the anode to a desired level. Silver inks are commercially available for most of the printing methods as a result of which the ink and layer properties depend on the printing method. The

solids content is typically 10-60 wt-%, resolution 40-100  $\mu\text{m}$  and viscosity over 10 mPa·s. The optimum thickness of the metal grid is 100-200 nm but currently it lies between 200 and 500 nm. The metal particles of the ink tend to agglomerate which increases the surface roughness of the grid. Optimizations and compromises are required to achieve both high conductivity and transparency. For example, the spacing between the grid lines should be ten times larger than the line width to obtain high transparency and relatively high conductivity.

The conductive organic polymer layer is typically made of PEDOT:PSS which is an aqueous solution having low solids content (1-4 wt-%) and high surface tension. [32-36] Polar co-solvents and additives are often added to increase the printability but this occurs at the expense of the solids content and conductivity of the ink. Currently, PEDOT:PSS inks are also available for careen printing with rather high solids content (4-10 wt-%) and viscosity (>8000 mPa·s). More information about PEDOT:PSS inks is presented in the Chapter 3.2.4. The organic layer should be smooth and uniform and be able to smoothen and completely cover the metal grid. The thickness should be 150-200 nm but in the case of low solids content the transfer volume needs to be high, thus making the control of the ink flow difficult. The properties and future targets of the metal grid and polymer layer are presented in Table 10.

Table 8. Properties and requirements for the metal grid and conductive organic polymer inks and layers.

	Metal grid		Conductive organic polymer		Unit
	Value	Target	Value	Target	
Solids content (wt-%)	10-60	-	1-10	> 5	wt-%
Viscosity (mPa·s)	>10	-	< 30	> 50	mPa·s
Surface tension (mN/m)	25-35	> 40	30-70	< 40	mN/m
Thickness (nm)	200-500	100-200	150-200		nm
Roughness (nm)	30-60	< 5	< 30	< 5	nm
Transparency (%)	> 80	> 90	> 90		%
Conductivity (k $\Omega$ /sq)	> 0.05	< 0.05	1-10	< 0.5	k $\Omega$ /sq
Resolution ( $\mu\text{m}$ )	40-100	10-20	-	-	$\mu\text{m}$

### 2.2.3.2 Suitable deposition and drying methods

The most suitable printing methods for the *ITO anode* are **gravure, flexography, inkjet, and screen printing** whose advantages and disadvantages are presented in Table 11. Gravure, flexography, and inkjet use low viscosity inks as a result of which thin and levelled layers are obtained. However, their excessive spreading and low thickness might decrease the resolution. Screen printing, for its part, can deposit thick layers using higher viscosity inks but the resolution is poor.

Flexography uses low nip pressure so that underlying layers are not destroyed or damaged during the anode deposition. In addition, the ink transfer amount is easily adjusted and controlled. On the other hand, the poor resolution and register decrease its suitability. Gravure printing produces high resolution at high speed and the ink transfer is adjustable and controllable. However, solid tones are reproduced via ink spreading and ink is not always transferred from all of the cells, which might lead to levelling problems and pinholes. The high nip pressure might also damage underlying layers. Inkjet is a non-contact method having a high resolution but it suffers from nozzle clogging and low printing speed. Nanoparticles agglomerate easily, thus clogging the nozzles, and the print resolution decreases with increasing speed. Screen printing uses also low printing pressure but the resolution is too poor.

Table 9. Advantages and disadvantages of the potential deposition methods for ITO anode.

Printing method	Advantages	Disadvantages
Flexography	<ul style="list-style-type: none"> <li>- Low nip pressure</li> <li>- Adjustable ink transfer</li> </ul>	<ul style="list-style-type: none"> <li>- Poor resolution and register</li> <li>- Thin layer</li> </ul>
Gravure	<ul style="list-style-type: none"> <li>- High resolution</li> <li>- Adjustable ink transfer</li> <li>- High speed</li> </ul>	<ul style="list-style-type: none"> <li>- Solid tones reproduced via ink spreading</li> <li>- Ink transfer problems</li> <li>- Thin layer</li> <li>- High nip pressure</li> </ul>
Inkjet	<ul style="list-style-type: none"> <li>- High resolution</li> <li>- Non-contact</li> </ul>	<ul style="list-style-type: none"> <li>- Low speed</li> <li>- Nozzle clogging</li> <li>- Thin layer</li> </ul>
Screen printing	<ul style="list-style-type: none"> <li>- Low pressure</li> <li>- Thick layers</li> </ul>	<ul style="list-style-type: none"> <li>- Poor resolution</li> </ul>

*Silver metal grid* has to have high resolution and low thickness. Therefore, the most suitable printing methods are **gravure, flexography, inkjet, and offset**. Gravure, flexography, and inkjet use low viscosity inks that spread easily whereas offset uses pasty high viscosity inks that spread less. On the other hand, offset produces rough ink layers due to the ink layer splitting during the ink transfer. This decreases both the transparency and conductivity of the anode. Offset also uses dampening water that might cause problems with conductive inks but waterless process is also available.

*The organic conductive layer* is thin and smooth and covers the metal grid completely. This is mainly achieved by using low viscosity inks, thus the main deposition methods are **gravure, flexography, and inkjet**. The low viscosity and solids content of the PEDOT:PSS inks require high transfer volumes which makes the control of the ink flow difficult when printing thicker layers than 100 nm. Screen printing can use high viscosity inks but the thickness is too high for this layer although thicker layers would cover the metal grid and its roughness completely and still form a smooth layer. The advantages and disadvantages of the deposition methods of the metal grid and the polymer layer are shown in Table 12.

Table 10. Advantages and disadvantages of the potential deposition methods for metal grid and conductive polymer layer.

Printing method	Advantages	Disadvantages
Flexography	<ul style="list-style-type: none"> <li>- Thin layer</li> <li>- Low viscosity inks</li> <li>- Low nip pressure</li> <li>- Adjustable ink transfer</li> </ul>	<ul style="list-style-type: none"> <li>- Poor resolution and register</li> <li>- Excessive spreading</li> </ul>
Gravure	<ul style="list-style-type: none"> <li>- Thin layer</li> <li>- Low viscosity inks</li> <li>- High resolution</li> <li>- Adjustable ink transfer</li> </ul>	<ul style="list-style-type: none"> <li>- Solid tones reproduced via ink spreading</li> <li>- Ink transfer problems</li> <li>- Excessive spreading</li> <li>- High nip pressure</li> </ul>
Offset	<ul style="list-style-type: none"> <li>- Thin layer</li> <li>- No excessive spreading</li> <li>- High resolution</li> </ul>	<ul style="list-style-type: none"> <li>- High viscosity inks</li> <li>- Ink layer roughness</li> <li>- High nip pressure</li> </ul>
Inkjet	<ul style="list-style-type: none"> <li>- Thin layer</li> <li>- Low viscosity inks</li> <li>- High resolution</li> <li>- Non-contact</li> </ul>	<ul style="list-style-type: none"> <li>- Low speed</li> <li>- Nozzle clogging</li> <li>- Excessive spreading</li> </ul>
Screen printing	<ul style="list-style-type: none"> <li>- Commercially available inks</li> <li>- Low pressure</li> <li>- Thick layers → covers roughness</li> </ul>	<ul style="list-style-type: none"> <li>- High viscosity inks</li> <li>- Thick layers</li> <li>- Poor resolution</li> </ul>

The anode layer can be dried by blowing hot air onto its surface to evaporate solvents. The drying temperature is 120-140 °C. This stage can also sinter the layer to some extent to increase its conductivity. PEDOT:PSS layer can be dried using lower temperatures (80-140 °C). The anode inks are also often UV-curable to increase their adhesion to the substrate. In this case, the ink contains binders that polymerize under UV light in the presence of photo-initiator additives.

### 2.2.3.3 Production process challenges

The anode layer should be as conductive as possible to enable efficient charge transport and generation in the cell, thus increasing the OSC efficiency and lifetime. Thus, the anode layer should be smooth, uniform, and homogeneous since unevenness and roughness decrease the conductivity and transparency. In addition, the OSC performance decreases if the resolution is poor and the edges are not sharp. Inert processing conditions can increase the conductivity by preventing the anode oxidation but this might cause problems in R2R processes. Current anodes have low conductivity and printing cannot reproduce the strict resolution requirements. Consequently, the printing process and inks require further developments.

Anode materials are highly transparent as a result of which light is conveyed reliably into the active layers of the OSC. Thus, the transparency has a smaller effect on the cell performance than the conductivity. However, some developments are still required to optimize the conductivity and transparency in the case of the metal grid based anode. To obtain high efficiency, R2R processability, and mechanical strength of the OSC, the anode should also be stable. The anode should withstand any deposition and drying stages without dimensional changes or damages and be chemically compatible with all the inks deposited after it.

The thickness of the anode affects also the OSC manufacturing and performance. Too thick anode layers can crack or increase the rigidity or brittleness of the OSC, thus decreasing runnability and changing conductivity. The smoothness and cleanness of the anode layer enable high conductivity and low light scattering as well as proper deposition of the hole injection layer (HIL). Due to the rather low viscosity of the anode ink, these properties are easily achieved, thus leading to high cell efficiency and lifetime. In addition, the surface energy of the anode layer is higher than the surface tension of the HIL ink due to the corona/plasma treatment. This ensures proper HIL ink deposition onto the anode layer.

The anode ink properties do not also affect the OSC performance. The ink viscosity is adjustable and the surface tension of the ink always lower than the surface energy of the barrier layer due to the corona/plasma treatment of the barrier layer, thus ensuring proper wetting and spreading of the anode ink. Furthermore, the plastic substrate should withstand the deposition and drying/curing of the anode layer without destruction and the barrier layer should be stable in order to achieve a good OSC performance.

The deposition and drying of the anode is extremely critical in the OSC manufacturing process since the layer conductivity and stability determine the OSC performance to a great extent. In addition, this layer should give a proper base for the following layers and be chemically compatible with them. The challenges of the anode layer as well as some possible solutions are presented in Table 13.

Table 11. Challenges of the anode layer and the solutions.

Challenges	Solutions
<ul style="list-style-type: none"> <li>- Low conductivity</li> <li>- Poor stability</li> <li>- Layer homogeneity and roughness</li> <li>- Chemical compatibility</li> <li>- Thickness and resolution requirements</li> </ul>	<ul style="list-style-type: none"> <li>- Printing process optimization</li> <li>- Material development and ink formulation</li> <li>- Inert processing</li> </ul>

## 2.2.4 Hole injection layer patterning and curing

### 2.2.4.1 Layer materials and properties

The hole injection layer (HIL) smoothenes the anode layer and enhance the OSC performance by creating a proper interfacial contact between the anode and photoactive layers, i.e., facilitating the charge transport. [32,37-42] Typically, low conductive PEDOT:PSS is used in printing applications due to its proper conductivity, transparency, and stability. PEDOT:PSS is an aqueous solution having a low solids content (1-3 wt-%) and high surface tension. Polar co-solvents (5-30 wt-%), conductivity enhancers (0-5 wt-%), and surfactants (<1 wt-%) are typically added to increase the material printability. However, this decreases the solids content (<1 wt-%) and electrical performance of the ink. The most common co-solvent is isopropanol (IPA) that decreases the surface tension of the ink to 30-40 mN/m and increases the viscosity to 10-30 mPa·s. As a result, the ink wetting, spreading, and printability improve but the ink drying characteristics change significantly. The resistivity of the HIL is 500-5000 Ωcm.

Table 12. *Properties and requirements for the hole injection layer.*

Properties	Value	Target	Unit
Solids content	< 3	> 5	wt-%
Viscosity	< 30	> 20	mPa·s
Surface tension	30-70	< 40	mN/m
Thickness	40-100	< 50	nm
Roughness	1-30	< 5	nm
Transmission	> 90		%
Resistivity	500-5000		Ωcm
Register	Depends on the module design	100-200	μm

The hole injection layer should be thin, smooth, homogeneous, and uniform. [32,37-43] The thickness ranges from 40 to 100 nm depending on the deposition method, ink properties, and ink transfer efficiency. It is preferable to have as thin layer as possible to prevent any extra charge transport losses. The HIL should also be as transparent as possible to allow large current density and maximum light absorption into the active layer. The main properties and requirements of the HIL are shown in Table 14.

### 2.2.4.2 Suitable deposition and drying methods

**Gravure, flexography, and inkjet** are the most suitable deposition methods for the HIL due to the low ink viscosity, solids content, and layer thickness. In addition, the water-based ink with polar co-solvents does not cause damages to the printing presses and materials. However, in order to prevent excessive ink spreading, the **printing speed** should be rather high and the **ink transfer volume** low. The low ink transfer volume can be obtained by using smaller cells in the printing cylinder (gravure) or in the anilox roller (flexography) or creating smaller droplets (inkjet). HIL is deposited as a solid patch on top of the anode, thus eliminating strict resolution and register demands. The advantages and disadvantages of the different printing methods are shown in Table 15.

Flexography uses low nip pressure during the ink transfer so that underlying layers are not damaged. The ink transfer is efficient since ink spreads and forms a uniform layer already onto the plate surface prior to the transfer onto the substrate. The ink transfer amount is adjusted by changing the cell volume of the anilox roller. However, the poor print resolution and register might cause problems in the deposition of HIL.

Inkjet is a non-contact method with high print quality but it suffers from nozzle clogging and low printing speed. HIL ink contains polymers that can easily agglomerate and clog the nozzles. In addition, the print resolution decreases significantly as the printing speed increases, thus creating pinholes.

Gravure has high print quality and printing speed. The amount of ink transfer can also be adjusted by changing the cell dimensions of the printing cylinder. Solid tones are reproduced via ink spreading which can lead to excessive or incomplete spreading. In addition, ink is not always transferred from all

of the cells which cause missing dots and pinholes. Gravure also has extremely expensive cylinders and the nip pressure is rather high.

*Table 13. Advantages and disadvantages of the different deposition methods for the hole injection layer.*

Printing method	Advantages	Disadvantages
Flexography	<ul style="list-style-type: none"> <li>- Thin ink layer</li> <li>- Low viscosity</li> </ul>	<ul style="list-style-type: none"> <li>- Cheap printing plates</li> <li>- Low nip pressure</li> <li>- Accurate and adjustable ink transfer</li> </ul>
Inkjet		<ul style="list-style-type: none"> <li>- Non-contact (no pressure)</li> <li>- High quality</li> </ul>
Gravure		<ul style="list-style-type: none"> <li>- High quality</li> <li>- High speed</li> <li>- Adjustable ink transfer</li> </ul>
		<ul style="list-style-type: none"> <li>- Poor resolution and register</li> <li>- Low speed</li> <li>- Nozzle clogging</li> <li>- Expensive printing cylinders</li> <li>- Solid tones reproduced via ink spreading</li> <li>- Ink transfer problems</li> <li>- High nip pressure</li> </ul>

The hole injection layer is dried by blowing hot air onto its surface to remove water and co-solvents. The drying temperature is typically 80-140 °C depending on the substrate and printing speed. The fast evaporation of IPA and slow removal of water create challenges to the drying process and make the OSC performance lower.

#### 2.2.4.3 Production process challenges

The surface properties and thickness of the HIL do not affect the OSC performance since the dilute and low viscosity water-based ink creates a thin, smooth, and uniform layer having a high surface energy. This high surface energy ensures excellent wetting, spreading, and adhesion of the low surface tension photoactive ink, thus ensuring good OSC performance. However, the control of the printing process might be difficult as a result of which the print quality and OSC performance might suffer. In addition, the ink transfer process affects the layer quality.

The high surface tension of the hole injection ink has a significant effect on the OSC efficiency. This surface tension should be lower than the surface energy of the anode to obtain good ink wetting and spreading as well as printability. Since the surface energy of the anode is often quite low, it has to be increased with corona/plasma treatment or the surface tension of the HIL ink lowered using co-solvents. The latter technique requires heavy ink formulation and optimization.

The drying of the water-based HIL ink is also difficult since it is difficult to remove water (much energy needed) from the ink layer and the polar co-solvents evaporate fast. This increases the layer unevenness and decrease the OSC performance. By using higher drying temperatures or lower printing speed the efficiency of the drying process is improved but too high temperature can damage the substrate and too low speed can lead to excessive ink spreading. The ink formulation may also improve the drying process.

The HIL conductivity and uniformity has an effect on the OSC lifetime since the charges have to flow easily between the anode and the photoactive layer. High conductivity improves the OSC performance and high uniformity the stability of the charge transfer. This layer should also withstand strong solvents used in the following deposition processes (mainly photoactive and cathode inks) since strong solvents can destroy this thin layer completely fast.

The printing of the HIL is highly important considering the whole OSC manufacturing process since it determines the charge carrying ability of the OSC. In addition, the high surface tension of the ink and difficulties in drying make the printing process control difficult and challenging on top of the anode. On the other hand, the layer properties affect the OSC performance only moderately due to its thinness, smoothness, and uniformity. Table 16 presents the main challenges of the HIL deposition and possible solutions.

Table 14. Challenges of the hole injection layer and the possible solutions to the problems.

Challenges	Solutions
<ul style="list-style-type: none"> <li>- High surface tension</li> <li>- Low viscosity and solids content → poor printability</li> <li>- Drying</li> </ul>	<ul style="list-style-type: none"> <li>- Ink formulation and optimization</li> <li>- Higher viscosity</li> <li>- Alternative co-solvents/PEDOT:PSS dispersions</li> </ul>

## 2.2.5 Photoactive layer patterning and curing

### 2.2.5.1 Layer materials and properties

The photoactive layer absorbs light and creates charges that flow through the layer to the electrodes, thus producing electricity. [11,15-18,21-25] The photoactive layer comprises of a blend of semiconductive polymers and fullerenes at the interfaces of which the charge transfer takes place. These photoactive materials are dissolved/dispersed into non-polar solvents to make them solution-processable, i.e., printable. The solvents are typically chlorinated, such as dichlorobenzene, chlorobenzene or chloroform. The solubility of the active materials is rather poor due to which the inks are extremely dilute and have low viscosity of 1-30 mPa·s. The properties and future targets of the photoactive layer are presented in Table 17.

Table 15. Properties of the photoactive layer and its requirements.

Properties	Value	Target	Unit
<b>Solids content</b>	<b>1-3</b>	<b>&gt; 5</b>	<b>wt-%</b>
<b>Viscosity</b>	<b>&lt; 30</b>	<b>&gt; 20</b>	<b>mPa·s</b>
<b>Surface tension</b>	<b>25-40</b>	-	<b>mN/m</b>
<b>Thickness</b>	<b>80-300</b>	-	<b>nm</b>
<b>Roughness</b>	<b>&lt; 30</b>	<b>&lt; 5</b>	<b>nm</b>
<b>Register</b>	<b>Depends on the module design</b>	<b>100-200</b>	<b>µm</b>

The photoactive layer should be smooth, homogeneous, and uniform and its thickness 80-300 nm. [32,37-40,43-47] The absorption maxima, i.e., the layer thicknesses where the cell efficiency reaches its maximum level, are approximately 90 nm and 250 nm. The low thickness requirement results from the poor mobility ( $\sim 0.1 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ ) of the organic semiconductors. The layer has to be smooth (< 20 nm) to allow proper light absorption without scattering.

### 2.2.5.2 Suitable deposition and drying methods

Due to the low ink viscosity, solids content, and ink layer thickness, **gravure, flexography, and inkjet** are the best alternatives for the deposition of the photoactive layer (Table 18). In order to prevent excessive spreading of the low viscosity ink, **the printing speed** should be rather high and **the ink transfer volume** small.

Flexography uses cheap printing plates that decrease the costs of the OSC manufacturing process. In addition, the low nip pressure ensures that underlying layers are not damaged during printing and the accurate, adjustable, and efficient ink transfer improves layer uniformity and control. On the other hand, the printing plates have poor solvent resistance as a result of which chlorinated solvents can damage the plate. In addition, the print quality (mainly resolution) and register are rather poor. By developing new plate materials with higher chemical resistance and resolution, optimizing the ink transfer, and using anilox rollers with small and low volume cells, flexography can become more suitable for the deposition of the photoactive layer.

Inkjet, for its part, is a non-contact method with high resolution. This eliminates the effect of the nip pressure on the OSC structure and performance and reduces the amount of waste. The final print quality is determined by the surface chemistry of the photoactive ink and the jetting parameters. Chlorinated solvents may cause damages to the print head electronics and because of the poor

solubility of the photoactive materials, the ink may contain large clusters that clog the nozzles. This drawback can be conquered by developing new photoactive materials with higher solubility. However, the print resolution and quality decreases at high speeds, thus significant process developments are still needed.

Gravure offers a high print quality and a high speed and volume production with adjustable ink transfer. The printing cylinder has superior chemical resistance also to chlorinated solvents. However, the printing cylinders are expensive and solid tones are reproduced via ink spreading. This ink spreading places great demands towards the control of ink-substrate interactions, ink transfer, ink properties, and printing conditions. Gravure printing suffers also often from incomplete ink transfer that creates pinholes. The ink spreading can be controlled by using higher printing speeds and lower cell volumes. Because of the excellent chemical resistance, high print quality, and high printing speed, gravure would be the best choice for the deposition of the photoactive layer.

Table 16. Advantages and disadvantages of the different deposition methods for the photoactive layer.

Printing method	Advantages	Disadvantages
<b>Flexography</b>	<ul style="list-style-type: none"> <li>- Thin ink layer</li> <li>- Low viscosity</li> </ul>	<ul style="list-style-type: none"> <li>- Cheap printing plates</li> <li>- Low nip pressure</li> <li>- Accurate ink transfer</li> </ul>
<b>Inkjet</b>		<ul style="list-style-type: none"> <li>- Non-contact</li> <li>- High quality</li> </ul>
<b>Gravure</b>		<ul style="list-style-type: none"> <li>- High quality</li> <li>- High speed</li> <li>- Superior chemical resistance</li> <li>- Adjustable ink transfer</li> </ul>

The photoactive layer is typically dried by blowing hot air onto its surface, which causes the ink solvent to evaporate. Typically, the drying temperature is 100-140 °C depending on the process speed, substrate, and ink solvents. During drying, the ink layer thickness decreases significantly since most of the ink volume (solvent) is removed from the ink layer.

### 2.2.5.3 Production process challenges

The photoactive layer should be thin enough to maximize the light absorption and charge generation (OSC efficiency) and to decrease process costs. However, with thin layers the printability and layer uniformity degrade because of more difficult and uncontrolled ink transfer. This decreases the OSC efficiency and lifetime. On the other hand, thick layers can crack during winding or decrease the bending capability of the OSC cell, thus causing problems in R2R processability and lifetime. The main challenge is to optimize both the printability and light absorption capacity by changing the ink properties.

The surface energy of the photoactive layer should be higher than the surface tension of the cathode ink to enable proper wetting and uniform spreading of the cathode ink. It is also advantageous to have a smooth, uniform, and clean surface and homogeneous layer structure to avoid light scattering, maximize light absorption as well as charge generation, and increase adhesion of cathode to the photoactive layer. The final layer structure is determined during drying when most of the ink volume is removed via evaporation but ink properties and composition determine mainly the surface chemistry of the photoactive layer.

The photoactive ink contains typically chlorinated solvents that are toxic and harmful to the environment. Due to the low solids content of the ink, the relative amount of solvent removed during drying is extremely large. Therefore, effective recovery systems are required. Alternative solvents and higher solids content reduce the need for the recovery and improve printability and print quality but the poor solubility of the material limits this. The increase in the solids content might also increase the layer thickness so that the OSC performance decreases. The chemical compatibility of the photoactive ink with previous layers is utmost important so that the ink does not dissolve or damage any of the underlying layers. The photoactive layer should also remain stable under any mechanical or thermal stresses encountered during the printing and drying processes.

The surface tension and viscosity of ink have only a moderate effect on the OSC performance. The surface energy of the HIL (water-based ink) is always higher than the surface tension of the photoactive ink (solvent-based), thus resulting in spontaneous wetting and spreading and good print quality. Together with the low ink viscosity, which makes the ink flow more easily, and the smooth and uniform surface of the HIL, a uniform, smooth, thin, and homogeneous photoactive layer is commonly formed. On the other hand, the lower the viscosity, the more the ink spreads and more difficult is the control of the print quality.

The plastic substrate should withstand the drying temperature of the photoactive layer to avoid register, runnability, and printability problems in the manufacturing process. In addition, the cleanliness and stability of HIL should be controlled and the cathode ink and bus bars/contact pads as solvent-based materials chosen carefully to obtain fully functional OSC and prevent print quality as well as adhesion problems.

As the whole OSC process is considered, the deposition and drying of the photoactive layer is not as significant as the deposition of the other layers. This results from the rather easy deposition and good printability of the photoactive ink on the top of the HIL. Therefore, the deposition of the other layers is more challenging and can cause bottlenecks in the manufacturing process. Table 19 presents the main process challenges and possible solutions to them.

*Table 17. Challenges of the photoactive layer and the possible solutions.*

Challenges	Solutions
<ul style="list-style-type: none"> <li>- Low solids content and solubility</li> <li>- Chlorinated solvents → recovery system</li> <li>- Low viscosity and thin layer → poor printability</li> <li>- Low surface energy</li> <li>- Control of the surface properties</li> </ul>	<ul style="list-style-type: none"> <li>- Higher solubility materials</li> <li>- Non-chlorinated solvents</li> <li>- Ink transfer control</li> <li>- Ink optimization</li> <li>- Higher surface energy (ink composition, surface treatment)</li> </ul>

## 2.2.6 Cathode patterning and curing

### 2.2.6.1 Layer materials and properties

Cathode determines the amount of electrons injected into the photoactive layer and hence the OSC efficiency. [32,34,40,45,48-52] The work function of the cathode material is typically low or matches with that of the photoactive layer to improve the OSC performance. A thin, low work function electron transport layer is often deposited between the patterned cathode and the photoactive layer to prevent interfacial mixing of these layers, improve the cathode printability, protect the photoactive layer from oxidation, and increase the OSC efficiency.

Aluminium and silver are the most commonly used cathode materials used in the printing applications due to their high conductivity. [32,34,40,45,48,52] Silver inks are commercially available but their high work function, low surface tension, and expensiveness cause severe deposition and operation problems. The work function of aluminium is low and matches perfectly with that of the P3HT:PCBM photoactive material, thus forming ohmic contact. However, aluminium inks are still under development. Typically, non-polar solvents are used which decreases the surface tension of the ink to 25-35 mN/m. Consequently, printability and wetting problems occur when printing on low surface energy photoactive layer. By increasing the amount of binders and additives, the printability improves but at the same time the conductivity decreases dramatically.

The cathode is often deposited on top of the electron transport layer. [32,34,45,48,52] The printed layer should be homogeneous to ensure excellent conductivity. The high conductivity and OSC efficiency is also obtained by using high solids content inks (10-50 wt-%) which increases the layer thickness (1-10 μm) and ink viscosity (> 200 mPa·s). However, in some printing applications, the viscosity has been 5-20 mPa·s. Properties and future targets of the cathode layer are collected into Table 20.

Table 18. Basic properties and requirements of the cathode.

Properties	Value	Target	Unit
Solids content	10-60	> 60	wt-%
Viscosity	> 5	> 200	mPa·s
Surface tension	25-35	-	mN/m
Thickness	1-10	~ 1	μm
Roughness	1	< 0.1	μm
Resistance	>10	Min.	mΩ/sq
Register	Depends on the module design	100-200	μm

Typical electron transport materials include Ca, Li, LiF, ZnO, and TiO<sub>x</sub> and some of them can be made printable. [32,34,45,48-52] These materials are often dissolved into polar solvents to improve the wetting and spreading of the cathode ink and to prevent dissolution of the photoactive layer. Electron transport materials tend to oxidise easily and they are reactive towards many materials, which limits the choice of solvents. The electron transport layer should be thin, smooth, and uniform to increase the OSC performance. Depending on the deposition method, the thickness ranges from 5 nm to 150 nm but the optimum thickness is 5-10 nm since thicker layers increase the resistance and cause losses in the cell efficiency. Therefore, the ink should have low viscosity.

#### 2.2.6.2 Suitable deposition and drying methods

The most suitable method for the *cathode* deposition is **rotary screen printing**. Screen printing produces thick ink layers with high conductivity using high solids content and viscosity inks. In addition, the low printing pressure ensures that underlying layers are not damaged during the cathode deposition. On the other hand, the print resolution is rather poor which might cause problems with small OSC sizes. If the resolution requirements are too high for screen printing, gravure, flexography, and inkjet might be used but these methods use low viscosity inks that can spread extensively and produce thin layers. Evaporation might also be used together with some patterning method, such as embossing or laser patterning but this increases significantly the complexity of the manufacturing process. Due to the fact that the inks are still under development, printing problems such as screen blocking, pinholes and too fast drying are easily formed. Table 21 shows the main advantages and disadvantages of different deposition methods.

Table 19. Advantages and disadvantages of the different deposition methods for the cathode.

Printing method	Advantages	Disadvantages
Flexography	- High resolution	- Thin layer - Low viscosity inks
Inkjet		
Gravure		
Screen printing	- High solids content - Thick layer - High viscosity - Low pressure	- Screen blocking - Low resolution
R2R evaporation	- High resolution - Homogeneous and thin layer	- No patterning/complex process with patterning

The *electron transport layer* can be deposited using **inkjet, gravure, or flexography** since the layer needs to be thin and homogeneous and the ink viscosity is low. Particle agglomeration may cause severe clogging problems in the case of inkjet. Some solvents might dissolve flexographic printing plates but the use of polar solvents eliminates this problem nearly completely.

The cathode and electron transport layer are both dried and sintered to obtain homogeneous layers. During drying, the ink solvent evaporates after which in the sintering stage the metal particles coalesce, thus forming a uniform film with high conductivity. Typical temperatures are 60-150 °C. In order to achieve complete sintering at these rather low temperatures, nanoparticles are used.

### 2.2.6.3 Production process challenges

Cathode inks use non-polar solvents that are often harmful to the environment and may dissolve underlying layers. Therefore, recovery systems are required but the relatively low amount of solvent makes the system lighter than in the case of the photoactive layer. Finding of alternative solvents and further ink development can improve the deposition process and OSC performance. The cathode ink properties have only a moderate effect on the OSC performance. The high viscosity decreases ink spreading and ensures its accurate deposition. Furthermore, the low surface tension enables excellent wetting, spreading, and adhesion of the ink. The chemical stability of the ink, for its part, affects the stability of the OSC.

The thickness of the cathode layer affects the OSC lifetime since it protect the photoactive layer from oxidation. The surface of the cathode layer oxidises rather easily and forms a rather stable, protective layer. The thicker the layer, the less oxygen gets into contact with the photoactive layer. With thinner layers, the whole cathode might oxidise, thus forcing the photoactive layer into contact with air. On the other hand, inert atmosphere is often required, thus making the construction of the printing press more complicated. The relatively high thickness of the cathode affects also the total active layer thickness, the total OSC thickness, and the R2R processability.

The conductivity of the cathode should be as high as possible to obtain long OSC lifetime and high OSC efficiency. The final structure of the cathode layer (morphology and topography) is determined in the sintering stage where individual metal particles coalesce, thus forming as homogeneous layer as possible. This can be obtained by deposition thick layers using inks with high metal content but too thick layer might also prevent complete sintering by hindering the sintering energy convection into the layer. In addition, the plastic substrate and underlying layers do not withstand high temperatures required for complete sintering. These factors increase the risk for inhomogeneous and porous cathode layer and lead to decreased efficiency and lifetime. Alternative sintering processes (e.g. laser sintering), sintering process developments, printing process optimizations, and ink formulation can improve the layer conductivity.

The cathode layer smoothness and surface energy affect the ink flow of the following layers, such as contact pads or bus bars and adhesive layer, whereas the surface cleanness ensures stable OSC operation. The mechanical and chemical stability of the cathode has significant effect on the OSC lifetime and efficiency, R2R processability, and mechanical strength. The surface chemistry as well as roughness of the photoactive layer should allow proper wetting and spreading of the cathode ink.

Considering the whole OSC manufacturing process, the deposition and drying of the cathode are highly important since they determine the conductivity, stability, and thickness of the cathode layer. These factors have extremely high influence on the OSC performance. Due to the fact that the ink development is still ongoing and the printing and drying processes require optimization, homogeneous layers are rather difficult to be produced, making this process stage a bottleneck in the OSC manufacturing. The main challenges and solutions are presented in Table 22.

Table 20. Challenges of the cathode layer and solutions.

Challenges	Solutions
<ul style="list-style-type: none"> <li>- Toxic solvents</li> <li>- Homogeneous layer</li> <li>- Conductivity, stability</li> <li>- Thick layer</li> </ul>	<ul style="list-style-type: none"> <li>- Alternative solvents</li> <li>- Ink formulation à printability vs. efficiency</li> <li>- Optimized sintering and printing process</li> <li>- Inert atmosphere</li> </ul>

## 2.2.7 Contact pads and bus bars (metallization) deposition and curing

### 2.2.7.1 Layer materials and properties

Contact pads and bus bars (metallization) are needed to prepare an anode and cathode contact from the OPV component/module and connect it to other OPV components/modules or other components such as batteries in order to store the created electricity. The main property of the materials used for metallization is high conductivity. Therefore, high conductivity metals such as silver, aluminium, copper can be used for preparing the metallization. [53] The current status in the development of high conductivity inks was already described in the previous Chapter 3.2.9, so it will be discussed only briefly in this chapter. The silver nanoparticles are most commonly used material for the metallization of components for printable electronics. The particle size is ranging from several nanometres to hundred nanometres which enable sintering in low temperatures suitable for plastic electronics. [54]

The layer thickness of metallization is normally in the range of 1-10  $\mu\text{m}$  in order to obtain adequate conductivity. Conductive pastes have high metal content (50-90 wt%) [54] and high viscosity ( $> 200$  mPas). The surface tension is required to be within 25-35 mN/m in order to wet the metallization ink on the surface of the anode, cathode and front side barrier. The target for the conductivity for printed metallization structures is to reach the same conductivity values as with the bulk metals. The metallization needs to align with the anode and cathode structures which set requirements for the registration and resolution. The basic properties and targets of the contact pads/bus bars are presented in Table 23.

Table 21. Basic properties and requirements of the contact pads/bus bars.

Properties	Value	Target	Unit
Solids content	50-90	50-70	wt-%
Viscosity	$> 5$	$> 200$	mPa·s
Surface tension	25-35		mN/m
Thickness	1-10	$\sim 1$	$\mu\text{m}$
Roughness	1	$< 0.1$	$\mu\text{m}$
Resistance	$> 2.8$ (Ag)	Min.	$\mu\Omega\text{cm}$
Register	Depends on the module design	100-200	$\mu\text{m}$

### 2.2.7.2 Suitable deposition and drying methods

Suitable processing methods for metallization layer are **rotary screen printing, ink jet printing, gravure printing and flexography printing**. A thick metallization layer is needed in order to obtain high conductivity for the printed structures. High solid content and high viscosity inks are normally used for preparing metallization. Therefore, the most suitable method for processing the metallization is rotary screen printing. However, screen printing is limited due to line resolution, speed and cost. If finer metallization structures are needed, ink jet printing, gravure printing and flexography printing can be used instead. These printing techniques require lower viscosities than screen printing, however, the solid content can be still relatively high (50-60 wt%). Therefore, several layers need to be printed on top of each other in order to obtain high conductivity.

Table 22. The advantages and disadvantages of suitable deposition methods.

Printing method	Advantages	Disadvantages
Flexography	<ul style="list-style-type: none"> <li>- High resolution</li> <li>- High printing speed</li> </ul>	<ul style="list-style-type: none"> <li>- Thin layer</li> <li>- Low viscosity</li> </ul>
Inkjet		
Gravure		
Screen printing	<ul style="list-style-type: none"> <li>- High solids content</li> <li>- Thick layer</li> <li>- High viscosity</li> <li>- Low pressure</li> </ul>	<ul style="list-style-type: none"> <li>- Screen blocking</li> <li>- Low resolution</li> </ul>

### 2.2.7.3 Production process challenges

Conductivity is one of main properties of the metallization. The thickness and conductivity of the metallization are parameters that are highly dependent of each other: For high conductivity, a rather thick metallization layer is needed. A metal layer can be oxidised easily that deteriorates the conductivity of the printed layer. Thicker metallization layer inhibits oxidization and maintains the conductivity of the metallization. However, sintering process of thick metallization layer with relatively low sintering temperatures might cause problems.

Inks for the metallization compose normally of metal particles, solvents, binders and additives. Toxic, non-polar solvents are used as a basis for the metallization ink system. Therefore, material and ink development is needed in order to replace these harmful solvents. The solvents, binders and additives should be chosen in order to take into account the layers processed beneath. The metallization should not dissolve the layers prepared earlier in the OPV manufacturing process. The ink properties in terms of viscosity, surface tension plays a key role in printing of high conductive metallization. The printing method for the metallization structure is chosen according to the requirements for conductivity, layer thickness and homogeneity. The metallization layer is processed on top of cathode, anode and front side barrier layers, so the ink of the metallization layer needs to wet on these different surfaces. The properties of the printing ink and printing method are strongly related to the morphology of the metallization layer and surface topography. Low surface roughness and cleanness are required since they have an impact on the proper adhesion of metallization ink on anode, cathode and front barrier surfaces. The stability of cathode has a moderate impact on the contact pad printing. If the surface energy of the cathode layer changes for instance due to the oxidation, it affects negatively the printing of metallization. The properties of the metallization layer in terms of surface roughness, surface energy have an effect on printing of adhesive layer.

The thermal properties of the plastic foil and organic materials limit the curing temperature of the metallization. Curing temperatures vary between 80°C to 200°C depending on the material choices. Curing temperature and time are strongly related to the conductivity of the metallization. Metal particles need to be coalesced in order to obtain homogeneous layer with high conductivity and low surface roughness. In addition, the ink properties including solvent(s), binders etc. impact on the choice of the curing temperature and time. Furthermore, too thick metallization layer prevents the complete sintering process and thus lower conductivity values are reached. A high thickness of metallization (1-10 µm) increases significantly the total active layer thickness and has a minor impact also on the total thickness of the OPV component. In addition, the thickness of the metallization has an effect on roll-to-roll processability.

The conductivity and layer thickness of the metallization are strongly related to OPV shelf lifetime and OPV operating lifetime. Other parameters such as surface topography and cleanness slightly affect the OPV operating and shelf lifetime. Instead, the impact of surface properties in terms of surface energy, surface topography and cleanness impact moderately on cell efficiency. Since metallization is used for connecting separated solar cells or solar cell modules to other components, the conductivity has a strong influence on monolithic cell integration.

Table 23. Challenges and solutions.

Challenges	Solutions
<ul style="list-style-type: none"> <li>- Low surface roughness, homogeneous layer → high conductivity</li> <li>- Toxic solvents</li> <li>- Rather thick layer</li> <li>- Sintering in rather low temperatures → high conductivity</li> </ul>	<ul style="list-style-type: none"> <li>- Optimisation of ink properties, printing and drying process</li> <li>- Material and ink development</li> <li>- Ink formulation + printing process</li> <li>- Optimized sintering and printing process, alternatives for sintering process</li> </ul>

## 2.2.8 Adhesive printing and lamination

### 2.2.8.1 Layer materials and properties

The organic solar cell needs to be protected against oxygen and moisture in order to maximize and stabilize the OSC lifetime. [28,32,55-63] This results from the easy destruction or oxidation of the cathode and active material upon exposure to these gases. The backside encapsulation is done by applying some adhesive onto the OSC and then laminating the OSC with a plastic film containing a barrier layer. Alternatively, the barrier film can be a tape-like film. The adhesive attaches the OSC to the barrier film and also acts as a sealant. Due to the strict barrier requirements, the adhesive deposition and lamination processes are often done at inert atmosphere.

The adhesive should be solvent-free and fast-curing and have low curing temperature to avoid any damages to the OSC. [28,32,55-63] Therefore, UV-curable epoxy or acrylate resins having 100 % solids content are commonly used. Epoxies have excellent barrier properties and they are commercially available for printing. The viscosity is typically above 100 mPa·s and WVTR and OTR values 0.006 g/m<sup>2</sup>/day and below 0.01 cm<sup>3</sup>/m<sup>2</sup>/day, respectively. However, the better the barrier properties, the more rigid the layer is, thus making R2R processing and adhesion problematic. Acrylates, for their part, are more easily formulated but can be too acidic to the cathode. The adhesive properties can also be changed by adding solvents (e.g. ethanol or acetates) or fillers but they can easily deteriorate the OSC performance or barrier properties. However, it should be noted that the UV-light can seriously damage the photoactive layer. Thus, other types of materials have to be developed when producing high barrier properties. The properties and future targets of the adhesive layer are presented in Table 26.

Table 24. Properties and requirements of the adhesive layer.

	Value	Target	Unit
Solids content	Depends on the adhesive		wt-%
Viscosity	> 100		mPa·s
Surface tension	Depends on the adhesive		mN/m
Thickness	2-20	> 1	µm
Roughness	< 1000	< 100	nm
Resolution	Depends on the OSC size		µm
Register	Depends on the module design	100-200	µm

The adhesive layer is typically patterned on top of the OSC but it can also be deposited as a continuous film. [28,32,55-63] The resolution depends on the active area of the OSC and the layer should be 2-20 µm thick to be able to seal the OSC and to attach the barrier film firmly over the whole OSC area. The layer should also be smooth and homogeneous to avoid any sealing problems.

The barrier film that is laminated on the top of the OSC by means of the adhesive layer is typically a plastic film coated with a thin layer of metal or barrier materials (see Chapter 3.2.2). [28,32,55-63] Similar properties are required from the barrier film as from the barrier layer deposited first in the OSC process but the film does not need to be transparent. The barrier films are laminated to the OSC in a nip under pressure. The barrier material can either face the OSC component or outside. If a metal film is used and the metal is facing the OSC, the adhesive layer should be deposited as a continuous film to avoid electric current flow from the cathode to the metal barrier. The barrier layer is typically 10-30 µm thick and the substrate roughness and pinholes play a major role in the layer performance.

### 2.2.8.2 Suitable deposition and drying methods

The adhesive layer can be patterned using rotary **screen printing, gravure, or flexography** because of the fact that the layer should be smooth, pinhole-free, and relatively thick. If the adhesive layer is deposited as a continuous film, **coating** methods would be the most appropriate since they can produce rather uniform, homogeneous, smooth, and thick layers. Table 27 compares the different deposition methods.

Gravure produces high print resolution at high printing speed. The transferred ink amount can be adjusted by changing the cell volume of the printing cylinder. The printing cylinder has also excellent

chemical resistance, thus making gravure suitable for the deposition of UV-curable materials. On the other hand, the gravure ink viscosity has to be rather low to enable ink spreading into solid areas and proper ink transfer. This can lead to too thin layers. In addition, all of the cells do not always transfer ink which forms pinholes. The nip pressure is also rather high which increases the probability for the OSC damage.

The ink transfer amount is easily and accurately adjusted and controlled in the case of flexography. The printing plates are cheap but their chemical resistance is often poor, thus making the use of UV-curable material difficult. However, the low nip pressure of flexography ensures that little damage is caused to the OSC. The resolution and register are poorer than in gravure but this is not an issue unless extremely small cells are produced. Flexography uses also low viscosity inks that produce thin layer onto the substrate.

Screen printing produces thick layer using low printing pressure and high solids content and viscosity inks. Therefore, this method would be the most suitable for the adhesive deposition. The ink layer thickness is also rather easily adjusted. On the other hand, the resolution might be too low in the case of small OSC and the chemical resistance of the screen might also cause problems.

*Table 25. Advantages and disadvantages of suitable deposition methods.*

Printing method	Advantages	Disadvantages
Flexography	<ul style="list-style-type: none"> <li>- Low nip pressure</li> <li>- Accurate and adjustable ink transfer</li> <li>- Cheap printing plates</li> </ul>	<ul style="list-style-type: none"> <li>- Poor resolution</li> <li>- Poor chemical resistance</li> <li>- Low ink viscosity and thin layer</li> </ul>
Gravure	<ul style="list-style-type: none"> <li>- High resolution</li> <li>- High printing speed</li> <li>- Superior chemical resistance</li> <li>- Adjustable ink transfer</li> </ul>	<ul style="list-style-type: none"> <li>- Low ink viscosity and thin layer</li> <li>- High nip pressure</li> <li>- Solid tones reproduced via ink spreading</li> <li>- Ink transfer problems</li> </ul>
Screen printing	<ul style="list-style-type: none"> <li>- Thick layer</li> <li>- Low printing pressure</li> <li>- High solids content and viscosity</li> </ul>	<ul style="list-style-type: none"> <li>- Poor resolution</li> <li>- Chemical resistance</li> <li>- Low speed</li> </ul>
Coating	<ul style="list-style-type: none"> <li>- Thick layer</li> <li>- Smooth and homogeneous layer</li> </ul>	<ul style="list-style-type: none"> <li>- No patterning</li> </ul>

The lamination is mainly done by laminating the OSC to barrier film in a nip between two rollers. These rollers are not heated to avoid damage to the OSC but the nip pressure as well as the roller material can be changed to ensure proper contact and adhesion between the two web materials. The UV drying of the adhesive can cause damage to the photoactive layer as a result of which drying at ambient conditions would be preferred but currently the best adhesive materials (high barrier properties) are UV-curable because of the high solids content and viscosity as well as the low spreading ability.

### 2.2.8.3 Production process challenges

The WVTR and OTR values of the adhesive and laminated barrier layer have a significant effect on the efficiency, lifetime, and encapsulation of the organic solar cell module. Moisture and oxygen can destroy or oxidise the cathode and active layers of the OSC, thus decreasing both lifetime and efficiency. The main challenge is to find appropriate materials having high moisture and oxygen barrier but still to be flexible enough to enable R2R processing.

The thickness of the adhesive layer is often high to achieve high lifetime and proper encapsulation by preventing oxygen and moisture diffusion into the OSC structure. Thick layers can, however, cause problems in the R2R processing if the adhesive material cracks or is too rigid. The relatively high thickness as compared to the active layers of the OSC affects the total thickness of the OSC.

The adhesive material should be stable and adhere properly onto the OSC to allow proper encapsulation and high lifetime. If the adhesive layer detaches from the OSC or does not withstand the mechanical, thermal, or chemical stresses encountered, oxygen and moisture get into contact with the OSC layers. The layer stability affects also the R2R processability and mechanical strength of the OSC component. The adhesive should also be chemically compatible with the underlying layers so

that they are not dissolved or damaged, thus affecting the barrier properties of the OSC. The ink viscosity is typically chosen according to the deposition method and surface tension according to the surface energy of the underlying layers.

The surface properties of the adhesive layer have a significant effect on the encapsulation. The layer should be smooth and homogeneous to achieve proper contact with the OSC and barrier film and to prevent oxygen and moisture penetration into contact with the cathode or active layers of the OSC. Pinholes are extremely detrimental to the encapsulation since they pass oxygen and moisture directly into contact with the OSC layers. Therefore, material and deposition processes require optimization and development.

The lamination process depends on the plastic foil thickness since too thick or thin materials are difficult to be attached without crack or wrinkles. In addition, the adhesive layer properties affect the quality. The surface properties of the contact pads or bus bars affect the deposition of the adhesive. The surface energy should be higher than the surface tension of the adhesive to allow proper wetting and spreading of the adhesive, and the surface should be smooth and clean to form intimate contact between the adhesive and the OSC (proper encapsulation).

The adhesive layer deposition and lamination are extremely important stages in the OSC manufacturing process since the layer properties determine the final efficiency and lifetime of the OSC by preventing oxygen and moisture transportation into the OSC structure. The most important layer properties affecting the OSC performance are the WVTR, OTR, and stability of the adhesive and barrier layer. The main challenges and their solutions are presented in Table 28.

*Table 26. Adhesive layer challenges and solutions.*

Challenges	Solutions
<ul style="list-style-type: none"> <li>- WVTR and OTR</li> <li>- Layer stability and adhesion</li> <li>- Smooth and homogeneous layer</li> </ul>	<ul style="list-style-type: none"> <li>- Material development and ink formulation</li> <li>- Process optimization</li> </ul>

### 3 Conclusions

The key requirements for the organic solar cells in order to reach entry for the energy market are sufficient lifetime, efficiency and production costs. Low production costs can be obtained by manufacturing organic solar cells (OSC) using low-cost roll-to-roll manufacturing techniques. However, the performance and stability of the OSC need to be improved before the market entry. In addition, alternative materials have to be developed in order to maximize the OSC performance.

Within this contribution, the concentration was on mapping out the requirements for the R2R production equipment and defining the key challenges of the roll-to-roll manufacturing process of organic solar cells by using Quality Function Deployment (QFD) as analysis tool. In addition, the requirements for the online measurements during the R2R manufacturing process were identified in QFD analysis that is of importance for controlling the R2R manufacturing process in order to produce high quality organic solar cell components with high yield. However, the more accurate analysis of the requirements for the measurement instruments and automation systems will be performed in Deliverable D1.4, but the key issues will also be discussed in this conclusion.

Organic solar cells have a multilayered structure where electrodes and active layers are deposited onto the substrate. Due to the high sensitivity of the layers to oxygen and moisture, the cells are encapsulated and sealed by means of barrier layers deposited onto both sides of the OSCs. There are also contact pads/bus bars to obtain contacts. The most important and challenging layers that require significant development work include barrier and electrode layers.

The barrier layers protect the OSC layers from oxygen and moisture extremely well, thus maximizing and maintaining the cell lifetime and efficiency. However, the barrier properties (WVTR, OTR) are still rather poor as a result of which further material, deposition method, and layer structure developments are required. In addition, the transparency requirement of the front side barrier layer limits the material choice and impairs the barrier properties. The barrier layer should be homogeneous since any cracks or pinholes create a path to moisture and oxygen into the OSC structure. An additional challenge arises from the fact that the substrate onto which the barrier layer is deposited should be clean and smooth so that defect-free and homogeneous barrier layer is obtained. The non-vacuum R2R deposition makes also the achievement of the barrier properties challenging.

The backside encapsulation is a simpler process since the barrier film is laminated by means of adhesive and non-transparent materials (metals) can be used. However, the adhesive layer should also be able to seal the OSC due to which the adhesive layer has to be smooth, flexible, and homogeneous and have proper adhesion.

The electrodes should be highly conductive to obtain high lifetime and efficiency of the OSC. Therefore, the electrode layers should be smooth, homogeneous, and stable. Electrodes might also require inert atmosphere processing, since they oxidise easily, thus diminishing conductivity. This places great demands for the R2R processing. The anode should also be transparent and have excellent resolution as well as sharp edges. The transparency and conductivity requirements require still material and process developments since highly conductive but non-transparent metals cannot be used. The cathode layer should be rather thick in order to protect the active layer from oxidation. Cathode inks are still under development since the lower work function of aluminium would be a more preferable choice than silver and commercial aluminium inks are not available. The final morphology of the cathode is determined in the sintering stage, thus making the sintering stage highly important. A thin electron transport layer might also be required under the cathode.

The measurement needs for the R2R manufacturing process of OSCs can be basically divided into four categories: printing environment measurements, printing machine measurements, measurements for the characteristics of printing inks, printing surfaces and print quality of single layers and OSC component/module measurements. In addition to the actual measurement processes, there is a need for a feedback loop back to the control and adjustment units in order to control the manufacturing process if deviations in the parameters defined in quality definitions are observed. Printing environment measurements compose of measurements for defining the particle level and distribution, temperature, humidity of the printing hall which are of importance in manufacturing functional organic solar cells. Printing machine measurements determine the printing parameters such as printing speed,

nip pressure, registration etc. Registration accuracy measurement and recognition is one of the key challenges in the OSC manufacturing process since the single layers need to be aligned with each other in order to produce complex solar cell structures. Misalignment due to the inaccurate registration could lead to the total module malfunction. Especially, transparent layers such as anode and hole injection layers sets challenges for the recognition and thus to the registration with the next layer. In the first phase, the development for the measurement technologies should be concentrated on the characteristics of the printing inks, surfaces and print quality in terms of viscosity, solid content, surface tension, surface energy, layer thickness, density, coverage, spreading, number of pinholes, uniformity etc. Although, these measurements are indirect, they correlate with the OSC performance, are rather readily available techniques, and are easy-to-install into R2R printing presses. In addition, there is also a need for the online conductivity measurement and measurements for front side and backside barrier properties (defects, VWTR, OTR) since these parameters determine the OSC efficiency and lifetime to a great extent. However, the development of the online measurement methods for the barrier properties is a long term objective, since accurate offline measurements are still under construction. Furthermore, the functionality of OSC component/module according to the quality requirements should be analysed and monitored before and after the lamination process.

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