

# FACESS, Flexible autonomous cost efficient energy source and storage

Final report





# **Flexible autonomous cost efficient energy source and storage**

**FACESS**

**Final report**

Kari Rönkä, Coordinator

ISBN 978-951-38-7791-0 (URL: <http://www.vtt.fi/publications/index.jsp>)  
ISSN 1455-0865 (URL: <http://www.vtt.fi/publications/index.jsp>)

Copyright © VTT 2011

JULKAISIJA – UTGIVARE – PUBLISHER

VTT, Vuorimiehentie 5, PL 1000, 02044 VTT  
puh. vaihde 020 722 111, faksi 020 722 4374

VTT, Bergsmansvägen 5, PB 1000, 02044 VTT  
tel. växel 020 722 111, fax 020 722 4374

VTT Technical Research Centre of Finland, Vuorimiehentie 5, P.O. Box 1000, FI-02044 VTT, Finland  
phone internat. +358 20 722 111, fax +358 20 722 4374

Technical editing Marika Leppilähti

Kari Rönkä. FACESS. Flexible autonomous cost efficient energy source and storage. 2011. VTT Tiedotteita – Research Notes 2602. 65 p. + app. 4 p.

**Keywords** organic solar cells, thin film batteries, roll-to-roll printing, Energy Source and Storage, chip-on-foil, flexible system integration

## Abstract

The key objective of Flexible Autonomous Cost Efficient Energy Source and Storage, FACESS project was to develop roll-to-roll printing process for organic solar cells (OSC) devices and thin film batteries (TFB). A control transistor circuitry on foil, which controls the operation voltage and charge of TFB's was also to be developed. The integration of OSC and TFB modules and control transistor circuitry into a flexible autonomous energy source and storage device was also one of the key objectives of the project. No material development was performed in the project, and thus only commercial materials were used. The technical target specification for OSC module was: 2.5% @ AM1.5, A~100 cm<sup>2</sup>, 250 mW and for the TFB module: Li-ion, 1-3 mAh/cm<sup>2</sup>, A~30 cm<sup>2</sup>.

The FACESS project (FP7 – 2007-ICT-1-215271) funded by European Commission was executed between January 1<sup>st</sup> 2008 and April 31<sup>st</sup> 2011. The developments were carried out by the following consortium: VTT Technical Research Centre of Finland (coordinator), IMEC Interuniversity Micro-Electronics Centre (Belgium), CEA Commissariat à l'Energie Atomique (France), WUT Warsaw Technical University (Poland), Umicore S.A.(Belgium), Coatema Coating Machinery GmbH (Germany), Suntrica Oy (Finland) and Coatema Maschinenbau GmbH (Germany). In this final report the key achievements, problems faced, lessons learned, as well as the vision and exploitation towards the large-scale production of flexible OSCs and TFBs, have been presented.

## Preface

The FACESS project (FP7 – 2007-ICT-1-215271) funded by European Commission was executed between January 1<sup>st</sup> 2008 and April 31<sup>st</sup> 2011. In this final report the key achievements, problems faced, lessons learned, as well as the vision and exploitation towards the large-scale production of flexible OSCs and TFBS, have been presented. The key contributors for the developments and authors for this final report have been listed below:

- VTT Technical Research Centre of Finland acted as coordinator of the project by Dr. Jukka Hast and Dr. Kari Rönkä. The main development activities were related to printing on organic solar cells, integration of Si-transistor circuitry on flexible foil and system integration. Key contributors on this work were Markus Tuomikoski, Marja Välimäki, Mari Ylikunnari, Päivi Kopola, Jaakko Lenkkeri ja Mikko Koutonen.
- IMEC Interuniversity Micro-Electronics Centre (Belgium) main development activities were related to organic solar cells and Si-transistor circuitry design, processing and integration on flexible foil. Key contributors on this work were Jan Genoe, Tom Aernouts and Bjorn Vandecasteele.
- CEA Commissariat à l’Energie Atomique (France) main development activities were related to thin Li-ion battery manufacturing and testing as well as organic solar cell and system testing. Key contributors on this work were Helene Rouault, Noella Lemaitre and Stephane Guillerez.
- WUT Warsaw Technical University (Poland) main development activities were related to electrolyte additives for thin Li-ion batteries. Key contributors on this work were Maciej Siekierski, Anna Marczevska and Maciej Marczewski.
- Umicore S.A.(Belgium) main development activities were related to battery materials and recyclability analysis. Key contributors on this work were Mieke Campforts, Eric Robert, Hossein Aminian and Michele Vanthournout.
- Coatema Coating Machinery GmbH (Germany) and Coatema Maschinenbau GmbH (Germany) main activities were related to design and manufacturing of the state-of-art inert atmosphere roll-to-roll printing machine and dissemination. Key contributors on this work were Thomas Kolbusch and Regina Reuscher.
- Suntrica Oy (Finland) main activities were related to system specification and design. Key contributors on this work were Mikko Kohvakka and Antti Backman.

# Contents

Abstract.....	3
Preface.....	4
1. Introduction.....	7
2. Potential end applications and markets for flexible autonomous energy source and storage.....	9
3. Technology development: key achievements, problems faced and lessons learned.....	14
3.1 Printing of organic solar cells.....	14
3.1.1 Printing process development.....	14
3.1.2 Lab-scale printed OSC modules.....	18
3.1.3 Inert atmosphere printing machine.....	19
3.1.4 R2R pilot fabrication of OSC modules.....	21
3.1.5 Printed organic solar cell performance and life-time.....	22
3.1.6 Summary of other OSC structures.....	25
3.2 Design and integration of Si-transistor circuitry on the foil.....	26
3.3 R2R manufacturing of thin film batteries.....	30
3.3.1 Battery with printed electrodes.....	30
3.3.2 Battery with fully printed core.....	33
3.4 System integration, packaging and testing.....	44
3.4.1 Integration prototype.....	44
3.4.2 System testing.....	48
3.4.3 Feasibility for R2R manufactured OSCs and TFBs onto the backplane.....	53
3.4.4 Recyclability.....	55
3.4.5 Demonstrator.....	57
4. Vision towards large-scale production of flexible organic solar cells, thin film batteries and power source and storage systems.....	59
4.1 Exploitation of project results.....	59
4.2 Benefit to European industry and economy by the FACESS exploitation.....	59
4.3 From roll-to-roll to roll-to-product – Taking printed intelligence developments out of the lab into products.....	63

## Appendices:

Appendix 1: FACESS dissemination activities

Appendix 2: The FACESS presentations





# 1. Introduction

Printed and organic electronics is an extremely fast developing technology field. Continuum of new innovations in materials and processing technologies enable completely new application areas which strengthen the position of organic and printed electronics as one of the future's key technology areas. The huge market potential of organic and printed electronics is already seen in market forecasts for future technologies. For instance, Frost & Sullivan forecasts \$35 billion industry market by 2015 for organic and printable electronics, and are in excess of \$300 billion by 2025. Until now, Europe has been in the top of this growing research area and to maintain the position, continuous investments which enable new innovations must be done.

One of the key issues is how these low cost printed electronic devices are powered. This is crucial especially in packaging industry and printed media which is one of the most important market entry fields in printed electronics. Organic, printable power sources such as organic solar cells, thin film batteries and fuel cells have come into intense research during the last years. Encouraging results have been obtained with printing organic electronics in laboratory, but transferring the laboratory scale processes to high volume manufacturing technologies for commercialisation needs further research. Nevertheless, organic materials have shown a strong potential which has led to first commercial products such as OLED displays.

The general objectives of FACESS project are therefore following: manufacture efficient organic solar cells (OSCs) and a thin film battery (TFB) on flexible substrate using commercially available



materials and cost efficient roll-to-roll (R2R) mass production techniques, printing, as well as integrate a control transistor circuitry on a foil. The ultimate goal is to integrate these three structures to a single assembly resulting in a flexible, fully autonomous energy source. In this assembly organic solar cells harvest the solar energy and charge the thin film batteries which provide the electricity for an external load. The Si-based transistor circuitry integrated on the foil controls the charge operation.

The FACESS project (FP7 – 2007-ICT-1-215271) funded by European Commission was

## 1. Introduction

executed between January 1<sup>st</sup> 2008 and April 31<sup>st</sup> 2011. The developments were carried out by the following consortium: VTT Technical Research Centre of Finland (coordinator), IMEC Interuniversity Micro-Electronics Centre (Belgium), CEA Commissariat à l’Energie Atomique (France) ,WUT Warsaw Technical University (Poland), Umicore S.A.(Belgium), Coatema Coating Machinery GmbH (Germany), Suntrica Oy (Finland) and Coatema Maschinenbau GmbH (Germany). In this final report the key achievements, problems faced, lessons learned, as well as the vision and exploitation towards the large-scale production of flexible OSCs and TFBS, have been presented.

## **2. Potential end applications and markets for flexible autonomous energy source and storage**

The existing solar market broadly falls into three market sectors: solar farms, building integrated photovoltaics (BIPV) and solar charging for a wide range of individual appliances. The key performance criterion for each sector is different: solar farms need a low \$/W, in BIPV long lifetime is essential and in the appliances market the key performance factor is sufficient power per unit area (which equates to the efficiency of the solar cell). The potential application areas for flexible autonomous energy source and storage development to be done in FACESS project were identified and analysed. Based on the selected application, the system specifications were drawn to form a basis for the system design.

As a result of the analysis, wireless sensor networks (WSN) were analysed and chosen the application base for the project development and demonstrator. In this analysis of potential applications and markets for the Energy and Communication Platform (ECP) were discussed.

According to the latest estimates by a market analyst firm IDTechEx, the size of the market for wireless sensor networks will reach globally 450 M\$ during 2011. The markets are estimated to hit 2000 M\$ by 2021. According to the IDTechEx, wireless Sensor Networks will eventually enable the automatic monitoring of forest fires, avalanches, hurricanes, failure of country wide utility equipment, traffic, hospitals and much more over wide areas, something previously impossible. Lack of standardisation and system providers has been hindering the growth of the market so far.

In general WSNs are an emerging wireless technology that will enable autonomous, self-healing and long-lived data collection networks in very large geographic areas. In contrast to current wireless data networks that have been targeting mainly high data throughput and low latency performance, WSNs target very low power consumption, high scalability and autonomous network configuration that eliminates the need of time consuming and expensive network configuration and maintenance operations. WSN nodes are locally powered by small batteries or solar panels. A target is that WSN nodes will operate even multiple years without battery replacements and any maintenance. WSN technology enables also the installation of sensors to places which were previously inaccessible, and perform measurements that would have previously been too expensive to realize using hardwiring methods. In sum, WSN technology enables new sensor applications and sensing of new, for example, remote and hazardous environments.

WSN technology will enable a large amount of new applications, or improve the performance, coverage, and accuracy of current sensing networks. The most potential applications such as remote monitoring of equipment, processes, groundwater, tanks and pipelines were identified. In addition, nearly

## 2. Potential end applications and markets for flexible autonomous energy source and storage

all oil, petroleum and gas companies use wireless technologies for field monitoring and automation applications, such as pipeline operation and wellhead monitoring. Suitable application areas are also environmental and traffic monitoring including automatic monitoring and warning of forest fires, avalanches, hurricanes and also traffic jams. Finally, building and home automation is one of the key application areas for WSNs.

The main arguments for the application identification and selection were the following

- Requirements for sensing are increasing in modern intelligent society
- Plenty of sensor types and use cases
- Low cost energy source has been a challenge in the research of wireless sensor networks
- High volume expectations, when wireless sensor network technology is mature enough for large scale installations
- Energy and Communication Platform will demonstrate a low-cost and light-weight solution for energy harvesting and storage
- Energy and Communication Platform demonstrates printed electronics to be used with WSN nodes.

A survey of suitable companies providing WSN products in the market was performed. The focus was on those companies that could provide WSN nodes and end-to-end solutions, while companies providing only chips or development boards were excluded. Based on the market survey a requirement, the following specifications for the FACESS project demonstrator development were selected, presented in Table 1. The system specifications for sub-modules, system layout and interconnections and packaging were also specified and designed.

## 2. Potential end applications and markets for flexible autonomous energy source and storage

Table 1. Wireless sensor node requirements for autonomous power source and storage.

Case application requirement	Case 1: router	Case 2	Case 3: leaf node	FACESS target performance and the key challenges
<b>Application specifications</b>				
Power ave/peak	400 $\mu$ W / 75 mW	500 $\mu$ W / 60 mW	200 $\mu$ W / 165 mW	
Energy consumption/day	35 J	43 J	17 J	
Typical use /day /Autonomy / maintenance time span	Always on, fully autonomous, no maintenance	Always on, fully autonomous, no maintenance	Always on, semi-autonomous	
Voltage	2.5 V – 5 V	3 V	3 V	3.7 V
<b>Power supply requirements</b>				
Running current	Ave 120 $\mu$ A, peak 25 mA	Ave 70 $\mu$ A, peak 20 mA	Ave 70 $\mu$ A, peak 55 mA	Peak 60 mA, adequate
Max dimensions /shape	100 mm x 30 mm, rectangular	N/A	95 mm x 60 mm rectangular	A4 Not to be size optimised.
Light condition (outdoor /indoor)	Outdoor	Outdoor	Outdoor	Outdoor
Solar panel performance at 100k lux	> 20 mW	> 20 mW	> 10 mW	2.4 mW/cm <sup>2</sup> 4 – 8 cm <sup>2</sup> enough, base module size 15 cm <sup>2</sup> selected
Battery capacity	> 20 mAh	> 20 mAh	> 10 mAh	65 mAh adequate
Cost (module / W/peak	< 1 EUR	N/A	N/A	~1 € with optimised OSC size
Lifetime	> 2 years	> 2 years	> 5 years	> 1 year with commercial low-cost barriers
Substrate (Flex/semi-rigid) Thickness	Semi-rigid/rigid	N/A	Rigid	Flex/semi grid
Weight	< 15 g	< 50 g (equals to 2 x AA)	< 50 g (equals to 2 x AA)	20 g with 4 OSC modules, battery, chip and backplane
Storage temperature	-40 °C ... +50 °C	N/A	-20 °C ... +65 °C	System testing -50 °C ... +125 °C
Operating temperature	-40 °C ... +50 °C	N/A	-20 °C ... +65 °C	

The most significant competitive advantages of FACESS technology were identified to be a very thin and lightweight structure, integrated battery and power conditioning electronics on the same foil and low manufacturing cost in mass volumes. Integrated battery and electronics will save space improve reliability and reduce overall cost of the solar energy harvesting system. In addition, flexible printing process enables easier variation of solar panel shape according to application requirements compared to competitive technologies. In Tables 2–4, the FACESS concept has been compared to other competing technologies.

## 2. Potential end applications and markets for flexible autonomous energy source and storage

Table 2. Comparison of FACESS concept to other solar cells technologies.

Technology	Advantages	Challenges
FACESS concept (conventional OSC structure)	<ul style="list-style-type: none"> <li>• R2R processable</li> <li>• &lt; 300 <math>\mu\text{m}</math> thin and flexibility</li> <li>• Light weight</li> </ul>	<ul style="list-style-type: none"> <li>• Improve power conversion efficiency and lifetime</li> <li>• Avoid vacuum deposition</li> </ul>
FACESS concept (inverted OSC structure)	<ul style="list-style-type: none"> <li>• Fully R2R solution processable</li> <li>• Thin, flexible and light weight</li> <li>• Prolonged lifetime</li> </ul>	<ul style="list-style-type: none"> <li>• Novel OSC structure</li> <li>• Needs high curing temperatures</li> </ul>
Commercial organic cells	<ul style="list-style-type: none"> <li>• Thin and flexibility</li> <li>• R2R processable</li> </ul>	<ul style="list-style-type: none"> <li>• Low power efficiencies</li> </ul>
Amorphous Si cells	<ul style="list-style-type: none"> <li>• Relative good power efficiency</li> </ul>	<ul style="list-style-type: none"> <li>• Toxicity of materials</li> <li>• R2R processing</li> </ul>
Dye-Sensitized Solar Cells (DSSC)	<ul style="list-style-type: none"> <li>• Overall peak power production efficiency potential high (using liquid electrolyte)</li> <li>• Mechanical robustness at high T</li> <li>• Works at low-light conditions</li> </ul>	<ul style="list-style-type: none"> <li>• Commercially low power efficiency (solid-state electrolyte)</li> <li>• Liquid electrolyte</li> <li>• Stability</li> </ul>
Copper Indium Gallium diSelenide solar cells (CIGS)	<ul style="list-style-type: none"> <li>• Best power efficiencies both reported and in markets</li> </ul>	<ul style="list-style-type: none"> <li>• Toxicity of materials</li> </ul>

Table 3. Comparison of FACESS solution to other rechargeable thin film batteries.

Technology	Characteristics	Challenges / Drawbacks
FACESS concept	<ul style="list-style-type: none"> <li>• Thin (&lt; 500<math>\mu\text{m}</math>) and flexible, 70 mAh, 3.7 V Current, max continuous discharge current 15 mA.</li> </ul>	<ul style="list-style-type: none"> <li>• High energy density (3 mAh/cm<sup>2</sup>)</li> <li>• Completely printed (mass production)</li> </ul>
Varta Microbattery	<ul style="list-style-type: none"> <li>• Between 340 to 1260 mAh</li> <li>• 3.7 V</li> <li>• Thickness 2.3 mm</li> <li>• In soft pouch but not flexible</li> </ul>	<ul style="list-style-type: none"> <li>• Not really thin flexible device</li> <li>• Discontinuous process: electrode coating and calendaring, but then pick and place steps</li> </ul>
Samsung	<ul style="list-style-type: none"> <li>• Very similar to Varta Microbattery</li> </ul>	<ul style="list-style-type: none"> <li>• Same comments than for Varta</li> </ul>

## 2. Potential end applications and markets for flexible autonomous energy source and storage

Cymbet Corporation, Angeon, Excellatron, and Infinite Power Solutions (US companies)	<ul style="list-style-type: none"> <li>• Solid state Li battery (glass electrolyte)</li> <li>• Very thin (typically &lt; 100µm)</li> <li>• High cyclability (&gt; 10000 cycles)</li> <li>• Max a few mAh, 3.7 V, max continuous discharge current &lt; mA</li> </ul>	<ul style="list-style-type: none"> <li>• No commercial breakthrough yet</li> <li>• Limited to 200 µAh/cm<sup>2</sup> due to the PVD and/or CVD techniques of deposit used</li> <li>• Possible for increase the thickness of the electrodes (so the 200 µAh/cm<sup>2</sup>) but then the performance decreases</li> </ul>
--	--	--

Table 4. Comparison of FACESS system to other system solutions.

Technology	Advantages	Challenges
FACESS concept	<ul style="list-style-type: none"> <li>• Integrated, autonomous system</li> <li>• Thin, flexible and light weight</li> </ul>	<ul style="list-style-type: none"> <li>• Efficiency improvement</li> <li>• Lifetime improvement</li> </ul>
Konarka power Plastics KT25	<ul style="list-style-type: none"> <li>• 0.25 W – 4 V, 18 g, 117 mm x 172 mm</li> </ul>	<ul style="list-style-type: none"> <li>• No integrated battery</li> </ul>
Flexcell Sunpack 7	<ul style="list-style-type: none"> <li>• Performance 7W -15 V, 600 mA,</li> </ul>	<ul style="list-style-type: none"> <li>• “Thick” 1.2 mm, heavy 500 g and big 350 x 900 mm</li> </ul>
PowerFilm The Power-pack +plus	<ul style="list-style-type: none"> <li>• Suitable small solar cells</li> </ul>	<ul style="list-style-type: none"> <li>• Battery packs very bulky and heavy</li> </ul>
Other OSC manufacturers	<ul style="list-style-type: none"> <li>•</li> </ul>	<ul style="list-style-type: none"> <li>• No integrated battery</li> </ul>

The main conclusions drawn from this analysis were:

- Competitive solutions with comparable or better PV efficiencies and flexibility already in market.
- FACESS could provide really lightweight, cost efficiency and non-toxic solution with possibility for system integration.
- FACESS technology will propose high energy density associated to flexibility and thickness. FACESS would be unique in the market providing integrated, autonomous flexible system with rechargeable battery.

The key challenges for the FACESS project were to develop the OSC and TFB printing capabilities from laboratory to pilot scale, improve OSC power conversion efficiency and improve the system lifetime.

## **3. Technology development: key achievements, problems faced and lessons learned**

In the following chapter the key achievements, problems faced and the lessons learned during the FACESS project have been presented.

### **3.1 Printing of organic solar cells**

#### **3.1.1 Printing process development**

The objective was to develop a method to fabricate organic solar cells on flexible substrate using roll-to-roll manufacturing technologies such as gravure, flexo and screen printing considering the system specifications. The work was started by optimisation of ink composition and printing parameters using lab-scale printing machines. After successful completion of the lab-scale phase, the objective was to further to transfer the process to pilot-scale printing machines. As an outcome of this work, the total OSC manufacturing process flow was to be designed and implemented.

The development work in the FACESS project was decided to be done with commercially available materials only, in order to ensure availability of sufficient material quantities for pilot-scale printing trials. The PEDOT:PSS ink was based on Clevios P VP AI 4083 (H.C. Starck) and the photoactive layer was a formulation of P3HT (Rieke Metals) and PCBM (Nano C). The ITO coated polyethylene-terephthalate film (PET) substrates were purchased from CP Films (sheet resistivity  $50 \Omega\text{square}^{-1}$ ). The definition of ITO patterns was performed with a screen printing etching paste, supplied by Merck.

#### **OSC module structures**

In the beginning of the project the FACESS consortium decided to utilise conventional organic bulk heterojunction (BHJ) solar cell structure in order to achieve project's targets (PCE of 2.5% under AM1.5 with the module size of  $100 \text{ cm}^2$ ). In addition, an inverted OSC structure was introduced in order to present future potential of this cell fabricated by means of fully wet chemical processes.



### 3. Technology development: key achievements, problems faced and lessons learned

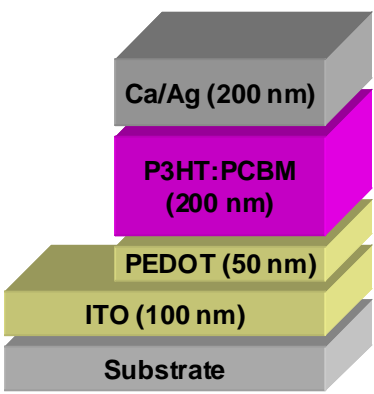
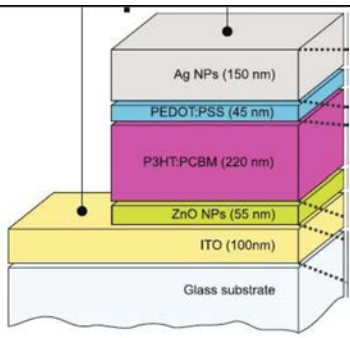
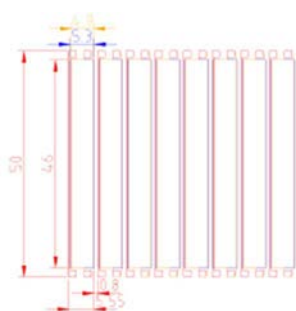
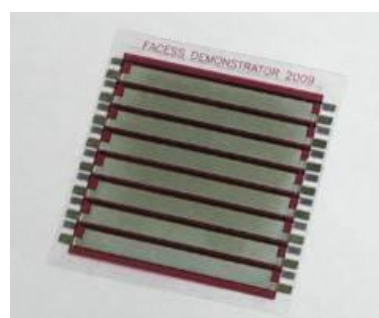
OSC type	Benefits	OSC stack	FACESS focus
Conventional BHJ	<ul style="list-style-type: none"> <li>- Well-known OSC structure and the fabrication process</li> <li>- Commercial OSC materials can be utilised</li> </ul>		Primary, R2R manufacturing
Inverted	<ul style="list-style-type: none"> <li>- Potential OSC structure for fully wet chemical processing</li> </ul>		Secondary, sheet processing

Figure 1. The summary of two OSC structure introduced in the FACESS project.

For further BHJ development two approaches was selected for the module structures: 1) OSC module with the busbars, and 2) interconnected OSC modules. In the OSC module with busbars (Layout 1), the pattern based on a multistrip structure was developed. In order to achieve high voltage required for charging the battery, 8 solar cell strips were connected in series.



(a)



(b)

Figure 2. The design (a) and the picture (b) of printed OSC modules with the busbars.

In the interconnected OSC modules (Layout 2), the extraction of the current sideways with busbars before interconnecting the cells can lead to unwanted series resistance effects which lower the overall

### 3. Technology development: key achievements, problems faced and lessons learned

performance of the module. Since a direct interconnection of neighbouring cells by overlap of the metal cathode of one with the ITO-anode of the next one can reduce such losses, this approach was chosen for further developments. A main challenge associated with converting this module structure into a R2R process, deals with the alignment, accuracy and resolution of the patterning.

The conventional module structure of bulk heterojunction (BHJ) interconnected OSC module (Layout 2 shown in Figure 3) was utilized for the system development and the prototype. Previous experiment by IMEC resulted as enhanced module PCE up to 2.5% compared Layout 1 with busbars, because lateral current collection limits cell PCE. The usage of directly interconnected individual cells in printed OSC modules was therefore predicted to achieve project's target of PCE 2.5%. In the first experiment, the manufacturing process in lab-scale was tested before implementation of OSC interconnected module processing in pilot environment, where roll-to-roll processing was done for ITO patterning and printing of active materials. Both sheet-to-sheet and R2R evaporation for the cathode electrode was utilised during the project.

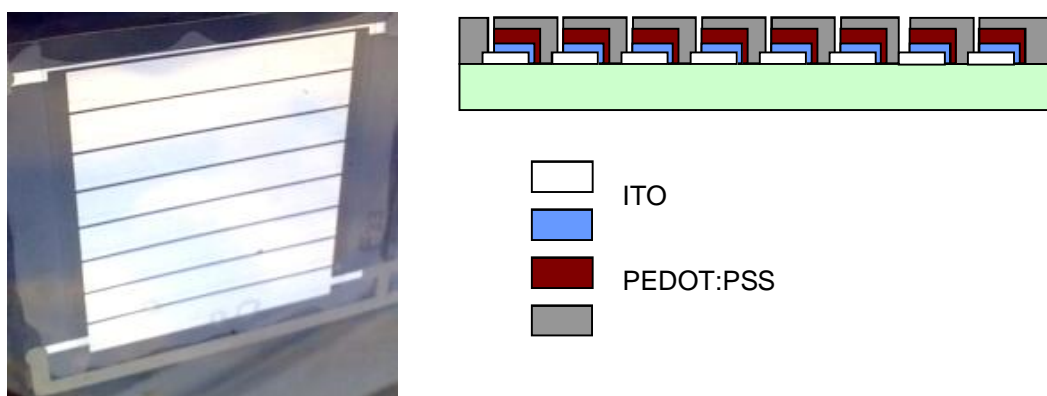


Figure 3. Printed BHJ interconnected OSC module (Layout 2).

OSC module manufacturing phases comprises of:

1. ITO-PET patterning and cleaning
2. Gravure printing of PEDOT:PSS and P3HT:PCBM photoactive layer
3. Ca/Ag metal electrode deposition.

#### **R2R ITO patterning**

Several patterning technologies and bottom electrode processing were tested during the project: laser ablation, wet chemical etching and paste etching as well as processing of conductive polymers, more specifically poly(ethylene dioxythiophene) PEDOT, doped with polystyrene sulfonate (PSS) and metal grids for improving OSC anode performance. As the final solution the R2R ITO patterning process with commercially available ITO etching paste (Isishape HiperEtch® 09S Type 40) was developed at VTT. The etching took place in a hot oven and the patterned image of ITO was revealed after paste removal in several washing steps.

Overall patterning process phases:

### 3. Technology development: key achievements, problems faced and lessons learned

1. Gel paste rotary screen printing
2. Etching
3. Gel paste washing and drying



Figure 4. The picture of patterned ITO coated plastic roll.

#### **R2R printing of PEDOT and photoactive layer**

PEDOT:PSS acts as an hole transport layer in the BHJ solar cell structure. In FACESS project, the PEDOT:PSS layer is processed with gravure printing on top of patterned and cleaned ITO. With PEDOT:PSS, it was targeted to a layer thickness of ~40 nm with variation of few nano-meters. During FACESS project, the preliminary approach taken was aimed at finding suitable chemicals which, when combined with PEDOT:PPS dispersion, lower the surface tension while maintaining good printability and electro-optical performance. The printability testing and device performance comparison by using different PEDOT:PSS ink formulations were done both in laboratory scale by using Schläfli Labratester and in R2R ROKO pilot printing machine at VTT. As a conclusion, PEDOT:PSS was found suitable for R2R processing with gravure printing.

On top of PEDOT:PSS layer, an photoactive layer of commercially available poly-3-hexylthiophene (P3HT) and [6.6]-phenyl-C61-butyric acid methyl ester (PCBM) deposited by gravure printing method was developed both in laboratory scale and R2R ROKO pilot machine. Different ink formulations, engraving parameters and printing speeds for gravure printing were developed and optimised.

#### **R2R metal electrode deposition**

The VTT vacuum R2R web coater (Figure 5) is able to handle plastic rolls with the length of 100 meters at maximum. The web width and thickness are in the range of 200–300 mm and 36–300  $\mu\text{m}$ , respectively. Two metal films can be continuously deposit in single run if needed. A shadow mask is used for the patterning of the metal films thus only a stripe pattern can be deposit on the web. This has been taken care in the designing of the OSC module layout. Since coater is brand new, optimisation of

### 3. Technology development: key achievements, problems faced and lessons learned

coating parameters is still needed. Especially, the side registration of the anode electrode and the metal film needs attention.



Figure 5. The pictures of the vacuum web coater: (a) from outside and (b) from inside.

#### OSC Encapsulation

Commercial barrier films with pressure sensitive adhesive were used in the FACESS project.. Unfortunately during the project's duration barriers were not available in rolls. Thus, A4 sized sheets were used for the encapsulation of OSC modules. As mentioned earlier, no work for specific barrier development was aimed to be done in the FACESS project.

Many companies have recently released public announcement of the availability of barrier materials in roll format. However, these materials are still in the development phase waiting for their commercial launching. It thus seems that it is a considerable challenge to manufacture high quality, pinhole free roll format barrier film since one minor defect can deteriorate device performance. At the moment, sheet format barrier films with varying performance can be provided by a number of companies on the barrier film market. It is very difficult to predict the future of barrier development. At the moment it seems that there will be only few major companies, such as 3M and Tera-Barrier Films, who may have the capability to provide high quality barrier rolls in terms of WVTR below  $10^{-6}$ . However, Amcor can already supply  $10^{-2}$  WVTR barrier rolls in high volumes which may provide a lifetime (LT80) of OSC modules up to 1000 hrs.

#### 3.1.2 Lab-scale printed OSC modules

The R2R patterning by gel-etching and cleaning process of ITO-PET substrate was confirmed first in the lab-scale printed OSC modules before the implementation in R2R pilot printing process. The PEDOT:PSS and photoactive formulas were gravure printed on patterned ITO substrate with the Labratester gravure printer. Finally the evaporation of Ca/Ag was carried out with sheet process before OSC module characterisation under AM1.5G solar illumination. The best printed module resulted PCE of 2.41%, FF 0.40, Voc 4.6V and Isc  $1.32 \text{ mA/cm}^2$  as shown in table below. The best printed OSC module with Au busbars resulted PCE of 2.3%. Overall, the performance and yield of lab-scale processed OSC modules without the busbars significantly depended on the accuracy of registration

### 3. Technology development: key achievements, problems faced and lessons learned

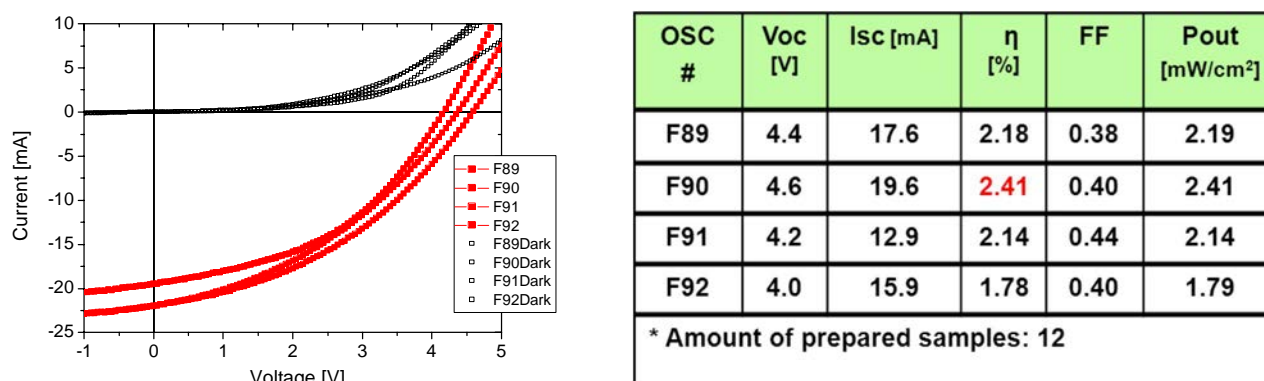


Figure 6. The IV characteristics of printed cell with the structure of **R2R patterned ITO-PET + lab-scale printed PEDOT:PSS/P3HT:PCBM + evaporated Ca/Ag**.

#### 3.1.3 Inert atmosphere printing machine

A new generation printing machines was design, manufactured and installed in FACESS project. The FACESS machine, later named as NICO, is able to realize the requested inline process determined by the complex structure of flexible polymer solar cells of the FACESS project. Because of the sensitivity of the used chemicals, the whole process must be realized inline under nitrogen atmosphere. After discussing a lot of different machine layouts the final design of the Inert Atmosphere Printing Machine was manufactured successfully.

COATEMA used all in all 10 different design approaches, specifically for the target of realizing the requested printing process as well as working in nitrogen gas atmosphere with very low amounts of oxygen and water allowed. During the design process a prototype was build and tested and the results of this testing led to the final design which is now assembled and tested at the Coatema facility and then shipped to VTT. As a end-result a unique facility for processing of oxygen and moisture sensitive components in pilot scale exists in the world.

### 3. Technology development: key achievements, problems faced and lessons learned



Figure 7. R2R Inert atmosphere printing pilot line with gravure and rotary screen printing and lamination located in VTT, Oulu.

The main characteristics of the NICO inert atmosphere pilot line are:

- 2 interchangeable printing unit slots (possibility add third later)
  - forward gravure
  - rotary silk screen
- Lamination unit
- Drying units (IR)
- Manual registration system
- Printing, curing and lamination made in nitrogen atmosphere (O<sub>2</sub> level below 1%)
- Max. web width 300 mm
- Max. web velocity 20 m/min.

The feasibility of roll-to-roll inert atmosphere printing machine developed within the FACESS project was carried out for the processing organic solar cells. The targets of the experiment were to gravure print a photo absorbing material in inert atmosphere. In overall, the photo absorbing material was successfully gravure printed on the top of PEDOT/ITO/PET samples resulting flat and pinhole-free films. Unfortunately during the FACESS project there was not enough time for broader testing for process ramp-up and optimisation as well as deeper understanding on the impact of IR heating for the performance of the solar cells.

### 3. Technology development: key achievements, problems faced and lessons learned

#### 3.1.4 R2R pilot fabrication of OSC modules

During the developments various R2R and laboratory-level combinations of ITO etching, PEDOT and photoactive printing and evaporation runs were performed in order to find the root-causes for up-scaling challenge when moving from laboratory printing to R2R pilot printing in VTT R2R ROKO pilot machine. Each single process steps were regularly found working as such. Herein, the summary of the various R2R pilot runs in total tens of kilometres has been presented carried out with VTT ROKO pilot printing machine in Figure 8.



Figure 8. ROKO pilot printing line.



Figure 9. R2R processing of OSCs.

During the R2R pilot processing it came evident that the R2R processing in pilot scale is challenging since so many issues need to be controlled at the same time. Also it needs to be notified that the used pilot environment is designed for flexible use for various developments (OFETs, OLEDs, OSCs, diagnostics, sensors etc.), not optimised for OSC processing only. The main observations are listed below:

### 3. Technology development: key achievements, problems faced and lessons learned

- A good registration of patterned ITO-PET (rotary screen printing method) and gravure printed PEDOT:PSS and P3HT:PCBM photo absorbing layers can be achieved. The gravure printing of P3HT:PCBM succeeded well, and the ink did not cause spreading over defined areas.
- Paste etching for ITO patterning in R2R pilot was developed during FACESS project. In the earlier runs etching paste residuals were observed on the ITO-PET substrate after cleaning. Cleaning steps were improved Better process control of etching and etching paste were investigated with the help of etching paste supplier Merck. Improved quality of ITO etching with better reproducibility was achieved.
- The spreading of printed PEDOT:PSS was more than expected in the earlier runs partially covering the anode contacts. Minor modifications of PEDOT:PSS formula was done for the following pilot run in order to decrease the spreading of the ink. The corrective action were successful and the spreading was now under control and did not cover anode contacts.

As a conclusion, it is evident that the scalability of OSC module printing from the laboratory processing to pilot environment introduced challenges, from which most were solved during the FACESS project. Each single process steps was found to be working repeatedly as such. The OSC module layout with evaporated metal cathode appeared to be too sensitive for mechanical conflicts and impurities of the pilot environment compared to laboratory circumstances. Further, a possible chemical reaction, such as oxidation of Ca layer might occur when the samples are transferred in air from the R2R evaporator into the glove box. The encapsulation right after the evaporation becomes crucial. Therefore, the fully R2R printed conventional bulk heterojunction (BHJ) interconnected OSC modules resulted the PCE of 0.8%, which stayed behind the project's target PCE of 2.5%. Main achievements in the R2R OSC processing were following.

- Development of OSC module processing – target of 2.5% power conversion efficiency – was achieved in the laboratory printing scale.
- Significant improvements for the R2R etching and cleaning of patterned ITO-PET substrate were managed.
- Successful development of ink formulations for the R2R gravure printing technique were achieved.
- Good R2R registration and alignment of ITO, PEDOT:PSS and photoactive layers for the interconnected OSC modules were achieved.
- R2R evaporation of Ca/Ag on the printed solar cells was proved. In future, the evaporation process will be replaced by soluble processing.

#### 3.1.5 Printed organic solar cell performance and life-time

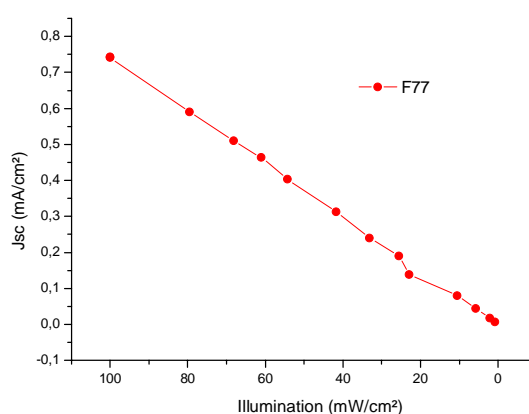
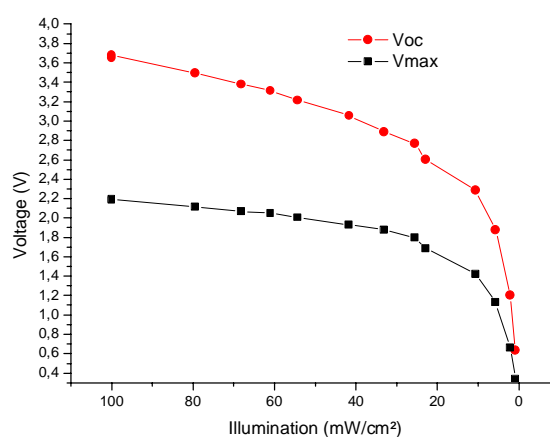
Lab-scale encapsulated 8 strips working module was tested at CEA INES in different illumination conditions, from 100 mW/cm<sup>2</sup> upto 0.83 mW/cm<sup>2</sup>. The evolution of the characteristic parameters of the modules in function of the illumination is presented in Figure 10.



### 3. Technology development: key achievements, problems faced and lessons learned

The short circuit current  $J_{sc}$  of the module varies linearly with the illumination intensity. In the contrary, the evolution of  $V_{max}$  is quite flat on a large illumination range. This is the case for  $V_{oc}$  as well, which is nearly constant ( $\approx 10\%$ ) from 100 up to 20  $mW/cm^2$ . The OSCs can therefore be used to charge the Li-ion battery on a large illumination range as the charge is done at constant voltage. The OSCs are particularly well adapted to such kind of applications.

The ageing tests for the OSC modules were performed at CEA INES with AM1.5 solar simulator where the encapsulated organic solar cells or modules can be connected so their IV curves can be monitored continuously. During the FACESS project, the lifetime of different organic solar modules were tested in different modes: a) individual modules encapsulated with different barrier materials, b) individual modules after attachment to a backplane, c) connected modules attached to a backplane. The modules were encapsulated with commercially available 10-3 barrier material. An example of ageing test results of individual modules tested one sun AM1.5 under atmospheric conditions, regulated temperature ca.  $30^\circ C$  are shown in Figure 11.



### 3. Technology development: key achievements, problems faced and lessons learned

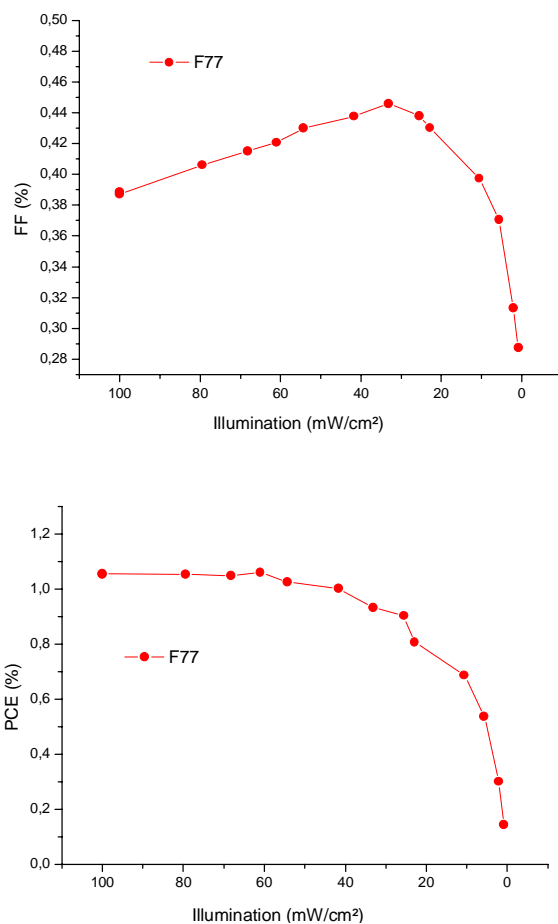


Figure 10. Evolution of the characteristic parameters ( $V_{oc}$  and  $V_{max}$ ,  $J_{sc}$ , FF and PCE) of the OSC module under different illumination conditions.

### 3. Technology development: key achievements, problems faced and lessons learned

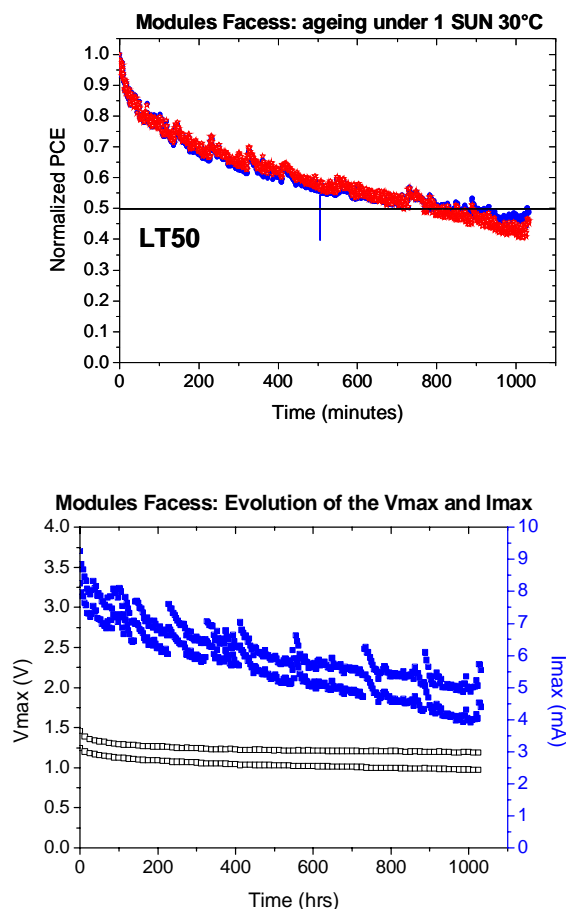


Figure 11. Evolution of the normalized PCE as well as the  $V_{max}$  and  $I_{max}$  of the different modules under accelerated ageing.

The results are really reproducible from one module to another. The drop of 50% of the initial PCE is attained after ca. 850 hours of ageing. This represents about 7 months of lifetime (1 year = 1500 hours of accelerated ageing) and is half way of the initial target of 1 year. In order to attain the 1 year target, the structure of the module could be changed for more stable structure like inverted one or a high barrier film ( $> 10^{-4}$  material) has to be employed for encapsulation. The  $V_{max}$  is relatively stable during the ageing of the module. After an initial burn-in the  $V_{max}$  value attain a plateau. This is good for the device working and the charge of the battery. Therefore, the practical life-time of the FACESS module can be longer than the (@50% of  $PCE(t_0)$ ), since the battery can be loaded as long as the  $V_{oc}$  stays over the set operational voltage of the system.

#### 3.1.6 Summary of other OSC structures

During the development for fully printed conventional bulk heterojunction (BHJ) interconnected OSC modules, also other structures and processes were done in reference. All development work was done with commercially available materials. The best results from these experiments were as following:

### 3. Technology development: key achievements, problems faced and lessons learned

#### Conventional OSC developments

- **The best gravure printed OSC module during demonstrator manufacturing**
  - $A = 15 \text{ cm}^2$ ,  $V_{oc} = 4.9 \text{ V}$ ,  $I_{sc} = 19 \text{ mA}$ ,  $FF = 0.47$ , **PCE 2.9%**
- **The best spray coated OSC module**
  - $A = 15 \text{ cm}^2$ ,  $V_{oc} = 4.7 \text{ V}$ ,  $I_{sc} = 16 \text{ mA}$ ,  $FF = 0.54$ , **PCE 2.5%**

#### Inverted OSC developments

- **The best spin coated inverted OSC**
  - $A = 15 \text{ mm}^2$ ,  $V_{oc} = 0.6 \text{ V}$ ,  $I_{sc} = 1.7 \text{ mA}$ ,  $FF = 0.57$ , **PCE 3.3%**
  - The solution processing of the  $\text{TiO}_x$  electron-transporting layer in the inverted configuration
- **The best OSC with solution processed low work function metallic ink as top electrode**
  - $A = 15 \text{ mm}^2$ ,  $V_{oc} = 0.6 \text{ V}$ ,  $I_{sc} = 2.2 \text{ mA}$ ,  $FF = 0.28$ , **PCE 2.3%**

## 3.2 Design and integration of Si-transistor circuitry on the foil

The objective was the design of transistor circuitry controlling the power supply system and the proven interconnection technology for integrating the Si-based circuitry onto flexible substrate. The interconnection technology was to be developed to be compatible with the manufacturing technologies studied for the OSC and TFB sub-modules.

The controller Si-chip was designed by IMEC and manufactured in Europractice.. Figure 12. shows the design layout and the actually produced chip.

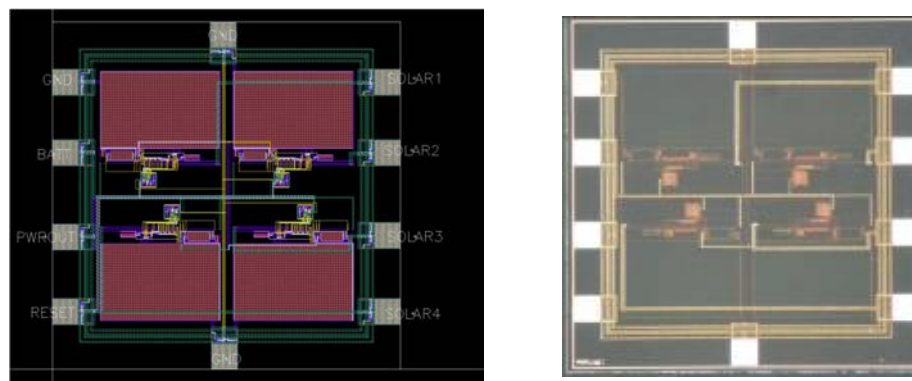


Figure 12. Left: design layout of the Si-chip; Right: Photograph of an actually produced control chip.

The necessary post processing steps to be performed on these chips are chip bumping, for flip chip assembly process, and chip thinning, necessary to make the chip flexible. Both of these processes are well established and mature on a wafer level scale, but are more difficult on a single chip base. For the FACESS chips, Electroless Nickel / Immersion Gold (ENIG) plating process was chosen as the most suitable bumping process. The advantage of this process is that it is completely maskless with a good

### 3. Technology development: key achievements, problems faced and lessons learned

quality of the bumps. The process can be divided into 3 different steps: bonding the chips on a carrier, plating the bumps and debonding the chips again.

After the bumping, the chips also needed to be thinned. This is again a standard process on a wafer level, but not on a chip scale. The different steps in this process are chip mounting, lapping and polishing. When the chip is bonded correctly, first a majority of the silicon is grinded away by lapping. This process however creates a lot of surface damage onto the chip, and therefore a polishing step is needed, to remove the damage and ensure a smooth backside of the chip.

Several potential chip on foil interconnection technologies were evaluated. Also the reliability of the manufactured chip to foil interconnections was studied. The dimensions of the controller chip and its interconnections are described in Table 5. As test methods different environmental tests, such as thermal cycling, damp heat and bending of the foils were used. Three different types of methods were studied:

1. Flip chip joining using Anisotropic Conductive Adhesive (ACA) or Non-Conductive Adhesive (NCA)
2. Flip chip joining using combination of Isotropic Conductive Adhesive (ICA) and NCA adhesives
3. Face-up bonding by direct printing of conductors over edge of thinned chip.

Table 5. Dimensions of the functional controller chip.

Component	Size mm	Thickness $\mu\text{m}$	Interconnections	Size of interconnection
Controller chip	3.5 x 3.5	40 $\mu\text{m}$ + bumps 5 $\mu\text{m}$	10 Au-plated Ni bumps	250 x 250 $\mu\text{m}$

The ACA flip-chip method was selected for more careful reliability testing because good yield and bonding quality was obtained in the interconnection process. The direct printing process was found to be a potential interconnection process for thinned chips. In addition the slightly more complicated ICA/NCA process was also tested for comparison purposes. As the substrate material the lower cost PET was selected because no considerable improvement was achieved with the higher temperatures tolerating PEN. Various adhesives were used in bonding trials. During the developments the bonding parameters, tools and methods were studied and optimized and special attention was paid to the challenges related to the handling and bonding of thinned chips. All these optimized bonding parameters resulted in a process with high yield and low contact resistances.

3. Technology development: key achievements, problems faced and lessons learned

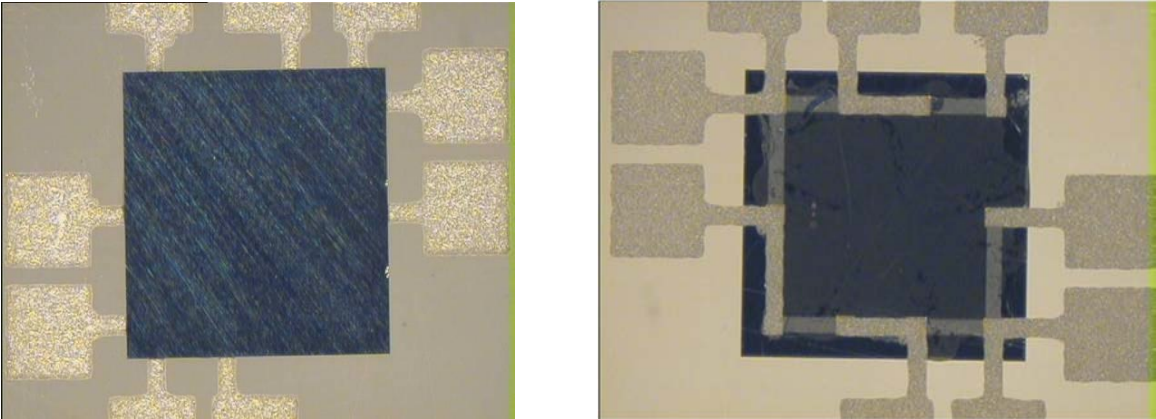


Figure 13. Top view and bottom view pictures through substrate of flip-chip bonded test structures made using DELO AC163 adhesive on PEN substrate.

The samples made by ACA flip-chip process and ICA/NCA flip-chip process survived the thermal cycling up to 1000 cycles and hot humidity testing up to 1000 hours. Their resistance stayed well below the total resistance of the whole conductive path. A slight decrease of resistance in the beginning of the tests was noticed due to the incomplete curing of the conductors, but this issue was addressed and an additional curing step was added for the following substrates. These results indicate that flip-chip structures with ACA or ICA/NCA adhesive well fulfil the reliability requirements of the consumer use category and the requirements of potential FACESS applications. Thermal cycling and hot humidity testing results for the ACA interconnections are shown below.

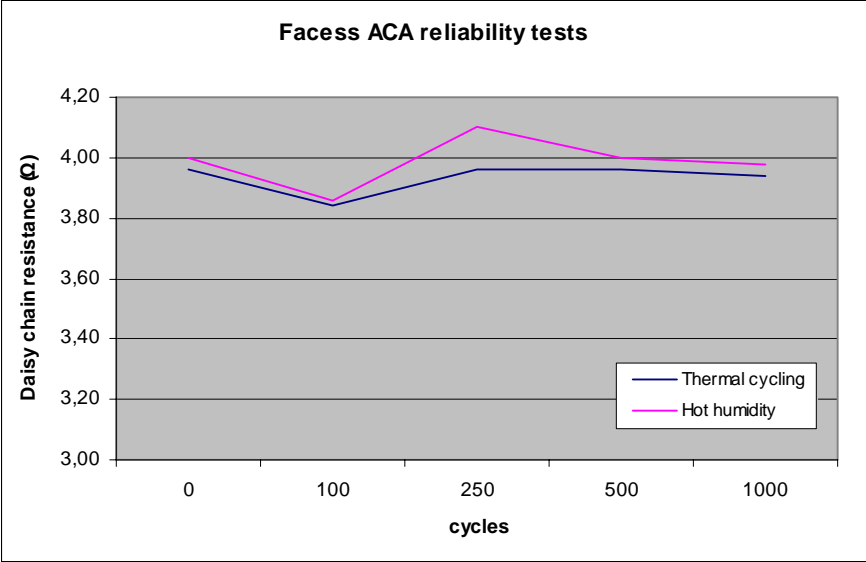


Figure 14. Daisy chain resistances of ACA interconnects during thermal cycling and hot humidity tests.

### 3. Technology development: key achievements, problems faced and lessons learned

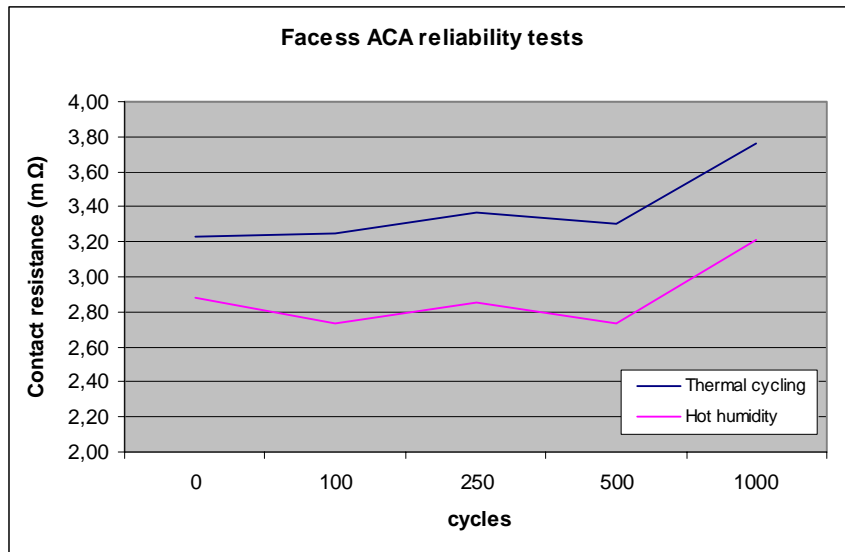


Figure 15. Contact resistances of ACA interconnects during thermal cycling and hot humidity tests.

For the further industrialisation, the flip-chip bonding process using ACA or NCA adhesive is a mature process in assembling RFID chips onto flexible antenna substrate in mass production. The process however needs a stop and go process, where the web stops for the time of the bonding process. The web must be stopped, because a force must be applied to the chip during the adhesive curing process. For many applications it would be beneficial to assemble control electronics chips onto a separate strap substrate and assemble the straps onto the web in a continuously running roll-to-roll process. Companies like Datacon or Muehlbauer have commercial equipment for such processes.

As for the final chip to foil bonding process the following specifications were chosen:

- PET substrate with thickness of 125  $\mu\text{m}$
- ACA flip-chip bonding procedure
- Flip-chip bonding directly on the back-plane substrate or onto a strap substrate of same material
- Chip thinning down to 40  $\mu\text{m}$
- Manufacturing sequence: chip bonding, assembly of OSCs, assembly of battery.

The selected process for thinned chip assembly and bonding is also suitable for R2R compatible processing in the future due minimal necessary process steps and reliable interconnection as well as due suitable snap curing adhesives and process equipments are available in the markets.

### 3. Technology development: key achievements, problems faced and lessons learned

## 3.3 R2R manufacturing of thin film batteries

The objective was to develop a method to manufacture completely a thin rechargeable Li-ion battery, in order to be connected to the printed solar cell module. Thus several investigations will have to be launched on:

- the elaboration of thick printed electrodes, essentially by developing adapted inks both transferable by printing techniques and electrochemically performing
- the implementation of an electrolytic membrane strongly linked to the electrodes and exhibiting specific characteristics to allow a good ionic conductivity.

First, the complete manufacturing protocol was to be designed and evaluated at the lab scale. Finally, the R2R printing process for thin film batteries was to be transferred to a pilot scale and tested there.

### 3.3.1 Battery with printed electrodes

The FACESS battery being a “storage” system, it has to exhibit thick electrodes, with thicknesses of about 80–100  $\mu\text{m}$ . Moreover, the electrodes have to be porous with an optimised porosity, allowing both exchanges of  $\text{Li}^+$  ions between the electrolyte and the particles of active material and relevant electronic conductivity through the printed layer. Furthermore, each electrode has to offer a smooth surface, avoiding any disturbs in the working of the battery. Inhomogeneous thicknesses of the electrodes may induce modifications of the electrical field inside the battery, and then lead to both a sub-exploitation of the nominal capacity of the battery and a reduction of its cycle life.

The FACESS Thin Film Battery was based on the Li-ion technology and the electrochemical system is composed of:

- lamellar lithiated metal oxide  $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  (or NMC) as active positive material
- graphite as negative active material.

Technology of the Li-ion Thin Film Battery selected in the FACESS project has been targeted as following:

Electrochemical couple:	$\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2/\text{Graphite}$
Nominal voltage:	3.7 V, working between 3.0 and 4.0 V
Positive electrode loading:	3.7 $\text{mAh}/\text{cm}^2$
Negative electrode loading	4.7 $\text{mAh}/\text{cm}^2$



3. Technology development: key achievements, problems faced and lessons learned

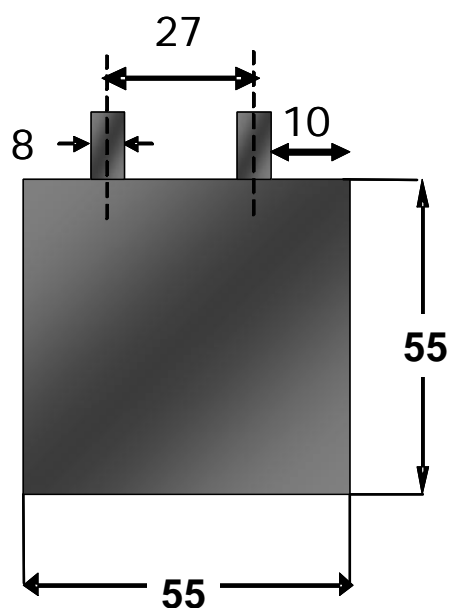


Figure 16. Dimensions of the thin film battery to be developed.

Table 6. Design and dimensions of the thin film battery to be developed.

	Version 1	Version 2
<b>Soft pouch (in mm, without the snap contacts)</b>	55 x 55 (possibly 56)	55 x 55 (possibly 56)
<b>Negative electrode (in mm)</b>	45 x 47	45 x 47
<b>Positive electrode (in mm)</b>	43 x 45	43 x 45
<b>Electrolytic component (in mm)</b>	47 x 49	47 x 49
<b>Overall area (in cm<sup>2</sup>, without the snap contacts)</b>	30.25 (possibly 30.80)	30.25 (possibly 30.80)
<b>Expected nominal capacity (in mAh, between 3.0 and 4.0V)</b>	30 mAh (if possible 70 mAh)	28mAh (if possible 65 mAh)
<b>Max continuous discharge current (in mA)</b>	150 mA (if possible 350mA)	100 mA (if possible 230mA)
<b>Max continuous charge current (in mA)</b>	15 mA (if possible 35 mA)	14 mA (32 mA)

The strategy of development of the FACESS battery was to build the TFB by stacking each component of the battery layer by layer,

### 3. Technology development: key achievements, problems faced and lessons learned

- The current collector for the first deposited electrode (negative or positive)
- The porous electrode layer (negative or positive)
- The microporous electrolyte membrane
- The second electrode layer (positive or negative)
- The current collector for the second electrode.

In the first version, a battery with printed electrodes was targeted with printed electrodes assembled with commercial microporous separator. The second phase, targeted to battery with fully printed core which would be realised by printing each components, i.e.:

- one electrode layer onto metallic foil used as current collector
- the electrolytic layer directly printed on the printed electrode
- the second electrode layer printed either onto metallic foil (version 2a) or on the electrolytic membrane (version 2b), depending on the success for printing the electrolytic membrane.

Different printing techniques were envisioned in the beginning: flexography, gravure printing and screen printing. In the beginning of the project, the developments of printed electrodes were focused on flexography technique. Together with screen printing, it allows reaching high thickness, in conditions that the ink formulation, the printing parameters and the flexography equipment are adapted. However, promising results in terms of adherence, thickness and electrical performances were achieved, the roughness of the layer was not acceptable due to a risk of short circuit when bending. Consequently, it was decided to shift the development of printed electrodes onto the screen printing technique, by using a flatbed screen printer.

For printing thick positive and negative electrode by screen printing, both printing devices as screen, squeegee and substrate holder and printing devices as speed and pressures were defined.

Printed negative (positive) electrodes were screen printed in the same shot with an average loading of 4.8 mAh/cm<sup>2</sup> (respectively 3.7mAh/cm<sup>2</sup>). The printing conditions and devices induce loading fluctuation of 0.6 mAh/cm<sup>2</sup> for the negative electrode and 0.1 mAh/cm<sup>2</sup> for the positive one between the printed patterned electrodes. The roughness obtained on each printed layer was very low. FACESS batteries built with the printed electrodes stacked with commercial separator exhibit similar electrical performances than batteries assembled with coated electrodes.

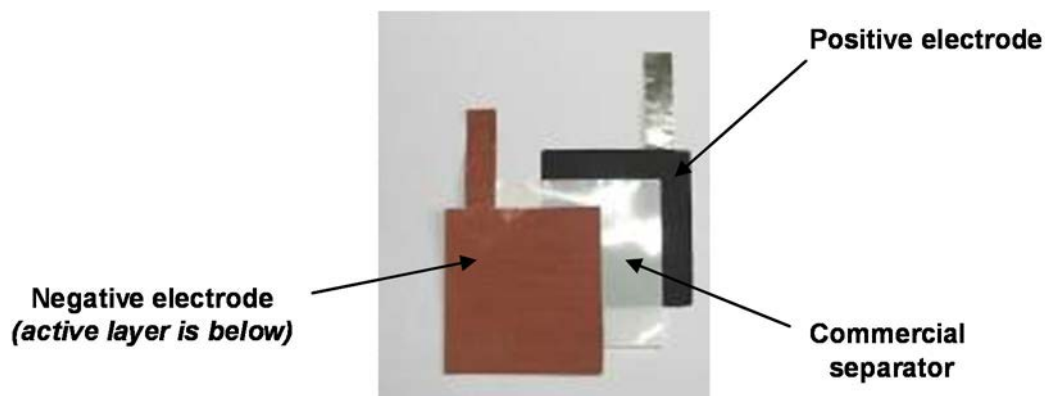


Figure 17. Core of the FACESS battery positioned as in the soft packaging. Positive and negative FACESS electrodes printed on Al and Cu foil respectively.

### 3. Technology development: key achievements, problems faced and lessons learned

#### 3.3.2 Battery with fully printed core

Printed electrodes with the well state of surface were clearly necessary for implementing the successful further step of printing. The electrolytic membrane has to ensure the physical separation between the two electrodes in order to prevent any short circuit. It has to be electronically insulating, but it has to allow the transfer of the  $\text{Li}^+$  ions from one electrode to the second one and reversely. In the case of Thin Film Battery, the membrane has to be also strongly linked with the two electrodes in order to realise a monolithic core, inducing high flexibility of the power source. In the Li technology three kinds of electrolytic component are used or under development:

- The Solid Polymer Electrolyte or SPE based on PolyEthylene Oxide (PEO) loaded with Li salt, but their working temperature (between  $60^\circ\text{C}$  and  $80^\circ\text{C}$ ) excludes the field of the portable applications.
- A separator based on Polyethylene PE or Polypropylene PP made porous by mechanical stretching and impregnated by liquid electrolyte. This solution is usually used in the Li-ion batteries and is obtained by simply stacking the microporous separator between the two electrodes.
- A Gelified Polymer Electrolyte or GPE constituted of a polymer matrix soaked with liquid electrolyte. Thus, when the membrane is gelifying, it swells and becomes conductive, thus preventing any risk of electrolyte leakage. Thus for the Li-ion technology, lots of studies are undertaken on membrane based on PolyVinylidene Fluoride (PVdF) polymer.

For the fully printed batteries, only this last concept may be applied, the step of electrolyte filling being planned either before the printing of the second electrode or as one of the finishing steps of the battery manufacturing process. For the FACESS project, this last case was so chosen, i.e. microporous membrane printed directly onto the negative electrode and impregnated with conventional liquid electrolyte.

In a complete R2R process, this choice induces to perform the electrolyte filling step at the end of the manufacturing process, that it is not so easy. But by starting from such development, the difficulties for printing a porous layer directly another porous layer and having to offer similar physico-chemical and electrochemical properties as a stand-alone membrane is very challenging. The development of printed gelled membrane (including electrolyte closely mixed with the membrane structure) will have to be developed in another framework, once each step of the complete manufacturing process will be well managed.

Besides, the printed membrane will have to exhibit a few specifications:

- The constitutive material has not to be soluble in the liquid electrolyte (based on organic solvents containing Li salts), but will have to offer very high affinity with electrolyte (good properties of wetting like the binder electrode) in order to have high capacity of retention.
- The polymeric material has to have the capability to be implemented in porous structure.
- It has to exhibit mechanical properties in order to withstand R2R process, so flexible and good mechanical resistance.

### 3. Technology development: key achievements, problems faced and lessons learned

- The polymeric material has to exhibit large electrochemical window, covering the working potential range of the full battery, typically between 0 and 4.2V versus Li<sup>+</sup>/Li
- The membrane has to be thick enough to prevent any physical contact between the electrodes (due to their roughness), but not too thick in order to reduce as much as possible the impedance of the battery.

For the printed FACESS battery, the chosen solution was so to use the principle of GPE, based on PolyVinylidene Fluoride (PVdF) polymer combined with mineral fillers, these materials inducing the microporosity inside the polymeric film, which we could name a "composite polymer membrane". Considerable interest has been shown in improving ionic conduction in nanocomposite polymer electrolytes. The best results were obtained with the use of Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, and SiO<sub>2</sub>.

In some case, it was demonstrated that the incorporation of fillers may be useful for improving the mechanical performances, increasing the cationic transference number or improving the interfacial stability. As the observed effects are strongly linked to the elaboration process used, careful attention has to be paid on the real effect of the added fillers.

Consequently, for the FACESS battery, it was decided to use a membrane ink composed of mineral fillers mixed with PVdF based polymer in a common solvent. The interest of the fillers is double:

- to have ink with adapted viscosity for the printing step
- to benefit eventually from the presence of fillers to possibly improve the mechanical strength and conductive properties of the membrane thus produced.

Three grades of PVdF were selected, evaluated and cycle tested in form of free standing membranes (without any filler). They were evaluated in full system, i.e. stacked between printed NMC positive electrode and printed graphite electrode in button cells. The cycling conditions were performed between 3.0 and 4.0V at various rates of charge-discharge, from C/20-D/20 up to C-D, then at C/10-D/10 on a long time. The electrochemical tests showed that all the PVdF grades exhibit stable electrical performances; the PVdF grade, giving the higher performance, was selected for the development of the composite polymer membrane printed onto an electrode.

Three kinds of fillers were envisioned, because often used in composites material as good compromising both to ensure good mechanical properties and offer high affinity with the electrolyte (in terms of wetting and swelling) and high electrochemical performances. Their nature and average particle size are:

- SiO<sub>2</sub> of about < 20 nm;
- TiO<sub>2</sub> of about < 20 nm;
- Filler<sub>CEA</sub> of about < 5 nm (commercial product).

As the pores of the membrane will be voluntary created by the fillers presence, the phase inversion based technique was not used for discriminating the best fillers. The technique used has consisted in the deposit of membrane inks with fillers either by simple coating onto glass but mainly by (screen) printing directly onto an electrode. The inks were formulated in Methyl Ethyl Ketone (MEK, T<sub>Boiling</sub> = 80°C), because the solvent is volatile enough to have a short curing step in the envisioned complete battery printing process.

### 3. Technology development: key achievements, problems faced and lessons learned

Thus, stand-alone membranes or printed membranes onto electrode were obtained and evaluated through cycling tests respectively in button cells or in soft pouch (FACESS batteries, version 2a).

*Comment:* the efficiency of the membrane printing process strongly depends on the ink formulation. Also the internal membrane structure, i.e. its porosity and tortuosity (the pore shape and their position) play a key role in the electrochemical performances of the component (mainly in term of ionic conductivity). So, compromises have to be established between the necessary to obtain relevant membrane morphology and to optimise the aimed electrical behaviour. Moreover, the ink has to be printable and so to exhibit the required rheological parameters as viscosity and shear thinning.

Consequently, the selection of the best fillers and their content for the different inks to be tested were undertaken in order to achieve both their printability, the porous morphology of the obtained printed layers and their electrical performances. In parallel, preliminary printing parameters have had to be developed. Thus, various ranges of compositions were tested by screen printing and visual observations were made both on the rheological quality of the inks and the morphology of the printed layers obtained.

Table 7. Range of ink compositions preliminarily tested by screen printing.

Ink composition	Fillers ratio	Printing parameters	Issues when such inks printed onto negative electrode
PVdF G1 in MEK solvent	4% to 10% SiO <sub>2</sub>	Screen 250 μm Metallic squeegee Blade pressure 3kg Speed of 30 mm/s	<ul style="list-style-type: none"> <li>• Too fluid inks for printing</li> <li>• Too much rigid membranes (of about 15 mm)</li> <li>• Occurrence of holes</li> <li>• Important deformation of Cu foils with electrode.</li> </ul>
PVdF G1 in MEK solvent	4% to 10% TiO <sub>2</sub>		
PVdF G1 in MEK solvent	4% to 10% Filler <sub>CEA</sub>		

From these first results, improvements were brought to the inks formulations (including the solid content) combined with the updating of the printing conditions, in order to obtain flexible printed membrane onto electrode without holes and with accurate outlines and no deformation of the Cu substrate.

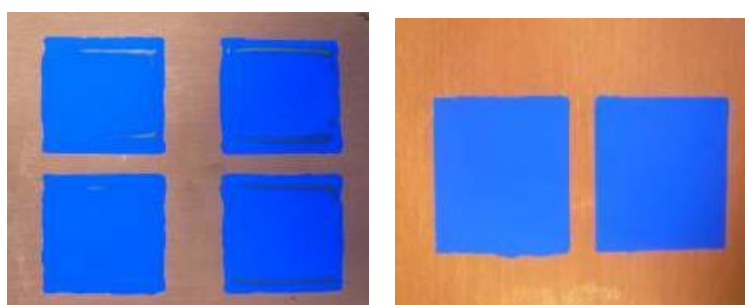


Figure 18. Improvement of the membrane directly printed onto the electrode.

Finally, four ink formulations were identified as possible solutions to be used as printed membrane, (see Table 8). The inks (F1 to F4) allowed reaching membranes with thickness comprised between 11 and 18 μm and a rate of swelling of conventional electrolyte of at least 170%. Nevertheless, note that

### 3. Technology development: key achievements, problems faced and lessons learned

the best rates were obtained with the inks loaded with the fillers<sub>SCEA</sub>. These four inks were easy to print directly onto electrode: they were not too fluid and the progressive but rapid MEK evaporation has allowed membrane elaboration on electrode with homogenous surface and without any holes.

Table 8. Range of ink compositions preliminarily tested by screen printing.

Ink composition	Fillers ratio	Thickness (μm)	Rate of electrolyte swelling (%)	Printing parameters	Issues when such inks printed onto negative electrode
F1: PVdF G1 in MEK	2% SiO <sub>2</sub>	7	168	Screen 250 μm PU squeegee Blade pressure 5kg Speed of 50 mm/s	<ul style="list-style-type: none"> <li>• Inks easier to print</li> <li>• Good porosity (60 to 75%)</li> <li>• No holes</li> <li>• Better resolution</li> <li>• More flexible membranes</li> </ul>
F2: PVdF G1 in MEK	2% TiO <sub>2</sub>	10	179		
F3: PVdF G1 in MEK	2% Filler <sub>CEA</sub>	9	187		
F4: PVdF G1 in MEK	10% Filler <sub>CEA</sub>	10	215		

In conclusion, on the printing point of view, the three kinds of fillers, i.e. SiO<sub>2</sub>, TiO<sub>2</sub> and the fillers<sub>SCEA</sub> may be used in the membrane ink formulation with the respective ratios 2%, 2% and 2% or 10%. Their definitive selection leading to the final composition of the membrane ink will be made only thanks to the cycling tests performed on Thin Film Batteries (TFB).

Thin Film Batteries (referenced FAC-PS46 to 50) were built according to the FACESS dimensions from two printed positive and negative electrodes, the membrane being (screen) printed directly onto the negative electrode. Another TFB named FAC-PS-51 was also assembled with Celgard® commercial separator to be used as reference. The batteries were so tested in cycling in the same conditions of charge and discharge at the same temperature. Their performances are presented in Table 9 and Figure 19.

Table 9. Evaluation of printed membranes onto negative electrode elaborated from different compositions.

Reference of the thin film batteries tested	Membrane characteristics		Discharge capacities recovered at various rates					
	Ink composition	Membrane thickness (μm)	C/20 (mAh)	C/10 (mAh)	C/5 (mAh)	C/2 (mAh)	C (mAh)	C/10 (mAh)
FAC-PS-48	PVdFG1-MEK-2%SiO <sub>2</sub>	12	62	51	42	12.5	0	51
FAC-PS-46	PVdFG1-MEK-10%Filler <sub>CEA</sub>	13	61	52.5	43	14	0	53
FAC-PS-47	PVdFG1-MEK-2%Filler <sub>CEA</sub>	11	60.5	53	43	13.5	0	54
FAC-PS-50	PVdFG1-MEK-2%TiO <sub>2</sub>	14	60.5	51.5	41	12.5	0	51
FAC-PS-51	Commercial separator (celgard)	25	63	53	44	15	0	56

### 3. Technology development: key achievements, problems faced and lessons learned

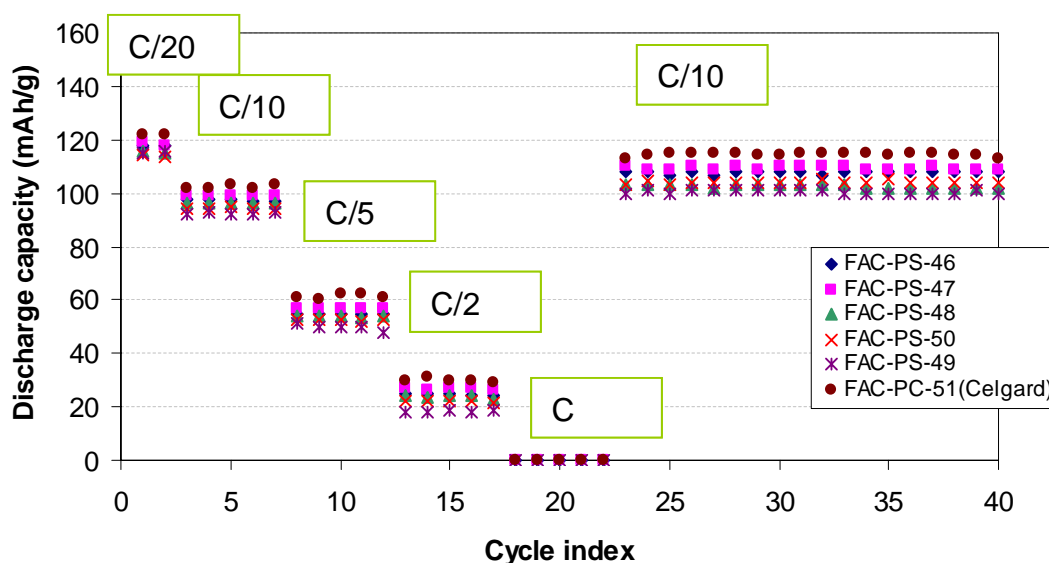


Figure 19. Cycling tests performed on various membranes directly printed onto the electrode.

In first conclusion, the four inks formulations seem to offer about the same electrical behaviour in terms of stability. Compared to the Celgard commercial separator and whatever the batteries, all the performances recovered are slightly inferior, but remain completely competitive to be used in Thin Film Batteries.

Nevertheless, the TFB referenced FAC-PS-47 seemed to exhibit the best performances. It is the reason why the formulation used for printing the corresponding membrane was selected for the printed FACESS batteries. In conclusion, the ink formulation was PVdF-G1 with 2% of Fillers<sub>CEA</sub> dissolved in Methyl Ethyl Keatone (MEK).

#### The printing process

As for the electrodes, the screen printing process was selected to realise the membrane directly onto the negative electrode. The development was undertaken on "sheet by sheet" equipment.



Figure 20. Sheet by sheet screen printer.

### 3. Technology development: key achievements, problems faced and lessons learned

Stencil metallic screen with opened patterns as for the electrode was selected in order to be sure to obtain flat surface. Indeed, preliminary printing tests have shown that marbled structure, so inhomogeneous morphology of the membrane may occur when a meshed screen is used. The thickness of the screen was designed at **250  $\mu\text{m}$**  in order to aim a membrane with only a few  $\mu\text{m}$ .

For printing the membrane onto the negative electrode, the substrate constituted of a Cu foil (about 18 $\mu\text{m}$  thick) with patterned graphite based printed electrode (about 80  $\mu\text{m}$  thick) was optimised.



Figure 21. Membrane (PVdF G1 with 2% of Fillers<sub>CEA</sub>) directly printed onto negative graphite based electrode

The batteries were so tested in cycling and then opened after about 50 cycles of complete charge and discharge. Mechanical tests were then manually performed in order to check the integrity of the batteries core by folding them in different direction and at various degrees. The results were very promising: no delamination observed, membrane remaining completely intact despite the mechanical solicitations and very flexible fully printed core.

The feasibility of the printing of membrane directly onto graphite electrode of Thin Film Lithium-ion Battery was demonstrated by screen printing by using metallic stencil screen. The membranes so obtained offer relevant characteristics as:

- regular thickness after printing, in conditions that the electrode layer below is very flat
- high adherence on the electrode and consequently flexibility of the double layer constituted by the electrolytic membrane on the negative electrode
- electrochemical performances equivalent to conventional Li-ion technology.

The next future step of this development will be concerned the print of the second electrode directly onto the electrolytic membrane. The challenge is huge, because several issues will have to be solved in parallel:

- To obtain a thick and porous electrode with a thickness of about one hundred  $\mu\text{m}$  and optimised porosity required for reaching high electrical performances, comparable to the previous versions of batteries
- To collect the current through the electrode layer by either depositing a supplementary layer or by using functionalised packaging.

In fact, the development of both the second electrode and the way for collecting the current will have to be undertaken jointly.



### 3. Technology development: key achievements, problems faced and lessons learned

Another important point to be mentioned concerns the printing techniques used. After investigating several possible techniques as flexography or screen printing with meshed screens, mitigated results were obtained. Certainly improvements may be brought but required huge investigations for solving issues of roughness or ink transfer while keeping the electrical performances of conventional Li-ion technology. In the framework of FACESS, all these developments were not possible due to complexity of the issues compared to the available time and human resources. It is why screen technique by stencil appeared the best and more efficient way to prove that manufacturing of fully printed Li-ion battery is possible. However, this technique is not possible in real Roll-to-Roll configuration. Indeed, Li-ion technology requires the print of very thick layer, what is not usual by traditional screen printing (with meshes screens). The only way to obtain both thick (of about 100 $\mu$ m) and very flat (unavoidable for having working electrochemical system) printed layers is the use of stencil screens; Consequently, the printing techniques envisioned to be online will be a Reel-to-Reel process, based on a stop-and-go movement. The band to be printed is locally stopped to perform the screen print of the layer, but at the process level, the rolls where the bands are wound or unwound turn continuously.

In conclusion, the developments of the printing process for the membrane as for the electrodes based on Sheet-by-Sheet process are transferrable on appropriated Reel-to-Reel equipment.

#### **Reel-to-Reel printing tests**

At the industrial scale, the reel-to-reel printing process is based on a stop-and-go mode and the printing through a stencil screen is performed during the stop step. However, on a large scale, the process appears continuous since the band is unwound and rewound without any stop obtained thanks to a compensative boucle.

The process works as follows:

- unwinding of the foil (metallic substrate)
- feeding through the Easycoater, i.e. a Sheet by sheet screen printer
- printing process with stencil screen
- drying process by IR and/or heated plate
- rewinding of foil.



Figure 22. Some steps of the Reel-to-Reel printing process.

Tests were performed with two Reel-to-Reel equipment and inks, designs and process parameters were optimised. During the first printing and drying tests, the substrate (Al or Cu foils) curved during the drying process. The ink moved to the edges, so that the dry sample exhibit round edges finally. Then the printing tests were switched on vacuum device the substrate being fixed during the drying process and the results were much better.

### 3. Technology development: key achievements, problems faced and lessons learned

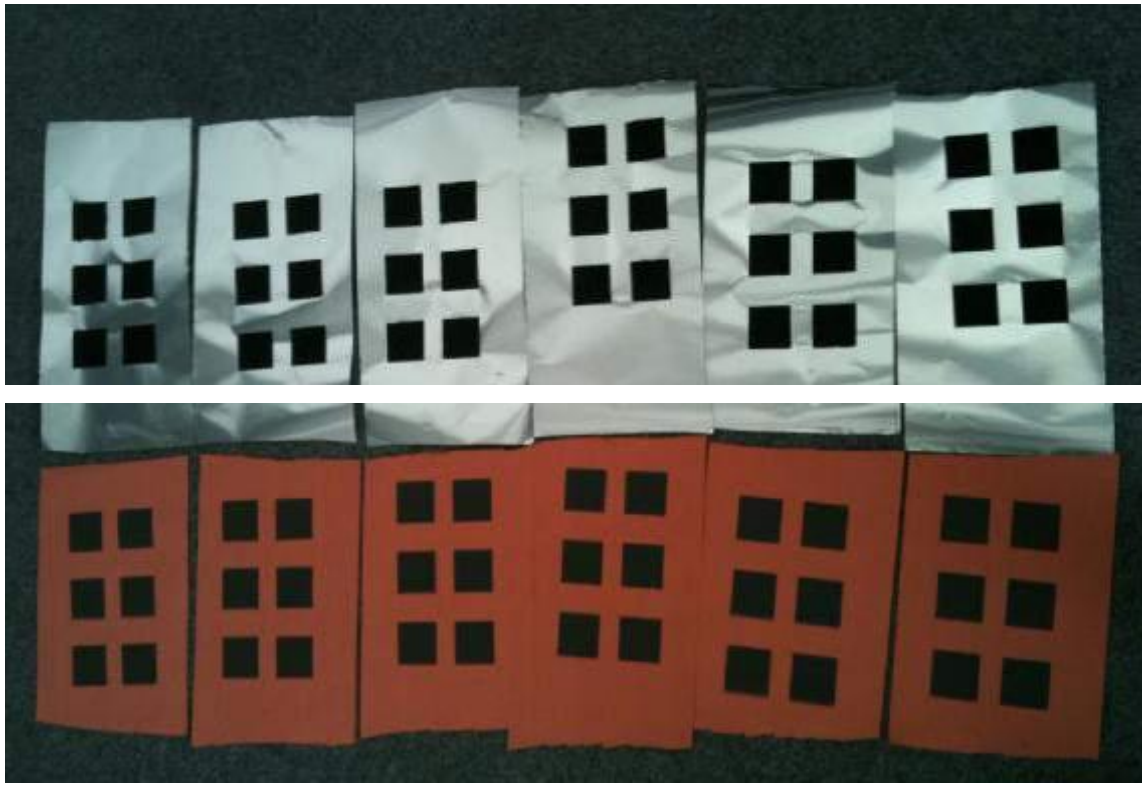


Figure 23. Negative and positive electrodes printed reel-to-reel printing trials.

The sheets of negative patterns and positive patterns have been observed accurately:

- positive patterns: depending on the printing conditions, sheets may exhibit patterns with huge spreading or well defined patterns
- negative patterns; the electrodes seem very thick, their thickness corresponding to the aimed thickness for a loading of about 4 mAh/cm<sup>2</sup>. As for the positive ones, depending on the printing and drying conditions, the outlines of the patterns are well defined or show spreading.

The outlines of the different various appears quite good and their dimensions compatible with the expected ones. In terms of roughness, for the positive electrode it was measured equal to 2 μm while it is equal to 6 μm for the negative patterns. This roughness is equivalent to the roughness obtained by traditional coating. However, for printing, these values may induce some problems when directly printing above the membrane.

In terms of loading, the negative patterns reach a loading compatible with the aimed loading of 4.8 mAh/cm<sup>2</sup> for the FACESS battery, i.e.  $4.4 \pm 0.07$  mAh/cm<sup>2</sup>. On the positive side, the loading is too low ( $1 \pm 0.1$  mAh/cm<sup>2</sup>), compared what it was expected for the FACESS battery, i.e. 3.7 mAh/cm<sup>2</sup>. Consequently, the electrode balancing always required for having relevant performances will not be able to be possible with these printed patterns. CEA has so decided to manufacture fully printed Thin Film Batteries, only with one of the reel-to-reel electrode, the negative ones.

### 3. Technology development: key achievements, problems faced and lessons learned

Fully printed batteries were built with the following core (presented in Figure 24, Figure 25 and Table 10 and tested. Comparative performance to fully sheet-to-sheet processed batteries was obtained.

- Reel-to-Reel printed negative electrode, loading  $\sim 4.5$  mAh/cm<sup>2</sup>
- Sheet-to-Sheet printed positive electrode, loading  $\sim 3.7$ – $3.8$  mAh/cm<sup>2</sup>
- Sheet-to-Sheet membrane directly printed onto printed electrode.



Figure 24. Functional fully printed batteries.

Table 10. Main characteristics of the fully printed batteries.

Prototype reference	Re2Re Negative COATEMA	Negative loading (mAh/cm <sup>2</sup> )	SbS positive CEA	Positive loading (mAh/cm <sup>2</sup> )	Positive active area (cm <sup>2</sup> )	Printed Membrane onto electrode CEA	Expected nominal capacity (mAh)
FAC-PS-33	C-AN-02	4.52	SP-NMC-5-5	3.82	19.35	SbS printed membrane	73.84
FAC-PS-34	C-AN-03	4.49	SP-NMC-5-6	3.86	19.35	SbS printed membrane	74.72
FAC-PS-35	C-AN-05	4.43	SP-NMC-6-1	3.8	19.35	SbS printed membrane	73.46
FAC-PS-36	C-AN-06	4.35	SP-NMC-6-2	3.9	19.35	SbS printed membrane	75.48
FAC-PS-38	C-AN-08	4.45	SP-NMC-7-2	3.98	19.35	SbS printed membrane	76.99
FAC-PS-42	C-AN-12	4.34	SP-NMC-8-2	4	19.35	SbS printed membrane	76.11

### 3. Technology development: key achievements, problems faced and lessons learned

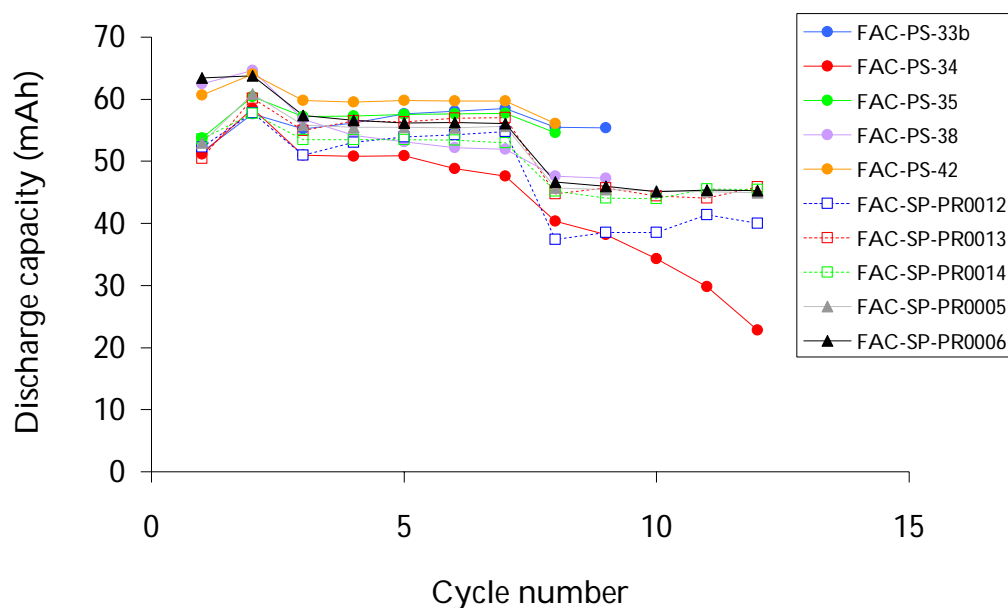


Figure 25. Performances in cycling of the fully printed batteries (33b, 34, 35, 38 and 42) compared to 5 SbS printed batteries at the lab scale.

Between 80 to 90% of the expected nominal capacities were recovered (Table 11). These results are comparable with those obtained with the SbS batteries. Indeed, the difference may be attributed to the printed membrane strongly linked to the electrode and maybe inducing internal resistance. This point will have to be studied in the future. Another reason may come from the quality of the electrode, if they loading are homogenous or not on their whole areas. This point has not been checked in details and will have to be accurately examined.

Table 11. Practical nominal capacity of the 6 fully printed batteries.

Prototype reference	Expected nominal capacity (mAh)	Practical nominal capacity (mAh)
FAC-PS-33	73.84	57.6
FAC-PS-34	74.72	58.5
FAC-PS-35	73.46	60.4
FAC-PS-36	75.48	-
FAC-PS-38	76.99	64.6
FAC-PS-42	76.11	64.0

### 3. Technology development: key achievements, problems faced and lessons learned

The present investigations have demonstrated that the fully printed batteries partially manufactured by Reel-to-Reel process exhibit comparable electrical performances as Sheet by Sheet printed batteries. The capacity recovered is high and meets the specifications. Indeed, a nominal capacity of 30 mAh was aimed, with the ambition to reach 70mAh. The trials show that the practical capacity may reach 64.0 mAh, with a very stable cyclability. However, it came clear during the system testing that more emphasis should have been put to the encapsulation, interconnections and reliability testing of the TFBs in order to better understand the life-time of the batteries from the encapsulation point of view.

Nevertheless, other investigations will be necessary. Indeed, the final objective for manufacturing fully printed TFB by R2R is to be capable to print successively each layer, i.e. one electrode onto a structured substrate, the membrane above the electrode, then the second electrode and at last the top of the packaging. As the FACESS project was the first consequent project where development of printed Lithium-ion battery was possible, the project has offered the opportunity to demonstrate that some steps of the process are possible. In parallel, the European GREENBAT project has partially used some results from FACESS to pursue now the development. These two projects will allow thus demonstrating that fully printing Li-ion battery is possible. And in the future, the work need to focus on the collective manufacturing and the implementation of flexible and structured packaging.

#### **Supramolecular additives**

Main WUT activity in FACESS project was development of the electrolyte additives being the modifiers of the charge transport process. The systems which were developed till now were tailored to the PEO solvent free polymeric matrix, and thus, must be redesigned to be optimized for the highly polar environment of the PVdF based gel type electrolyte (for both standard and printable electrolyte). On the other hand contrastively to the situation of the metallic lithium anode containing battery where the stability against reduction is most crucial in this case a high potential cathode is used enforcing the concentration on the resistivity versus oxidation. Main points of the activity were, thus, related to:

- Development of the new supramolecular anion receptor able to complex anions in highly polar environments. This compound is based on the amide derivative of already tested calyx-pyrrole based receptors. By the addition of such a compound to the electrolyte formulation two positive effects can be obtained. First is related to the improvement of the cation current fraction (increase of the lithium transference number) by the anion complexing and immobilization. Second effect is related to the stabilization of the battery internal resistivity over cycling leading to the improvement of the system life time.
- Development of the in-situ nano-filler generation in the electrolyte by the hydrolysis reaction of the organometallic precursor. The main advantage of the proposed strategy lies in the fact that typically nano filler are very easily agglomerating. In this case as the grain is created in the very viscous media at a given concentration securing the separation of the grains by the matrix the risk of agglomerates formation is depressed.
- Development of the superacidic filler. In a special process acidic groups originating for the sulfuric acid are irreversibly bound to the surface of the oxide ( $\text{Al}_2\text{O}_3$  or  $\text{TiO}_2$ ) grains. Resulting filler works not only as a mechanical modifier to the membrane structure but also can interact with anions improving the transport properties. Current development is related to the

### 3. Technology development: key achievements, problems faced and lessons learned

need of the process downscaling to grains being small enough to be incorporated in printable electrolyte formulation.

Generic conclusions were made from the studies:

- C4P represents better electrostability than CX2
- C4P also blocked autocatalytic reactions (confirmed by NMR data)
- both used supramolecular additives should be tested in PVdF based system
- functionalized alumoxane is able to bind residual water from bulk of electrolyte
- water binding proceeds through two reactions: methyl groups hydrolysis and coordination to Al atoms
- the use of ethanol, isopropanol, butanol and pentanol as a reagent doesn't provide to degradation of MAO molecule
- free coordination sites of aluminum atom enable to coordinate residual water or not evaporated alcohol
- it is possible to synthesize functionalized methylalumoxane and use it in membrane preparation
- the discharge capacities appeared stable along the cycles, whatever the rates and the additive use
- nevertheless, the performances seem to be systematically inferior to the PVdF alone
- moreover, the membrane doped with ChP4 and ChP6 exhibit the lowest performances.

From these characterizations (chemical, electrochemical and electrical by cycling tests), the additives combined with PVdF 6020 show that their effect do not modify fundamentally the PVdF performances, except the ChP4 and ChP6 additives.

## 3.4 System integration, packaging and testing

The objective was to integrate the main sub-modules (OSCs, TFBs and control circuitry) to form a functional system. The energy source and storage system was to be implemented on system substrate (or backplane), which works as physical carrier and wiring backplane for interconnection between components. The process flow of packaging, cutting and interconnections specified in the system design was to be tested and realised.

### 3.4.1 Integration prototype

Three types of components were planned for the demonstrator prototype: 4 pieces of organic solar cells sub-modules, a thin-film battery and a controller chip. The sizes of the components and types of their interconnections are described in Table 12 as well as in Figure 26, Figure 27 and Figure 28

### 3. Technology development: key achievements, problems faced and lessons learned

Table 12. The dimensions of the components used in the prototype device.

Component	Size mm	Thickness $\mu\text{m}$	Interconnections	Size of interconnection
OSC	50 x 50		4 metallic pads on PET substrate	width 1.65 mm length 4.2 mm
Battery	55 x 55		2 metal strips Al and Cu	width 8 mm length about 20 mm
Controller chip	3.5 x 3.5	30 $\mu\text{m}$ + bumps 5 $\mu\text{m}$	10 Au-plated Ni bumps	250 x 250 $\mu\text{m}$

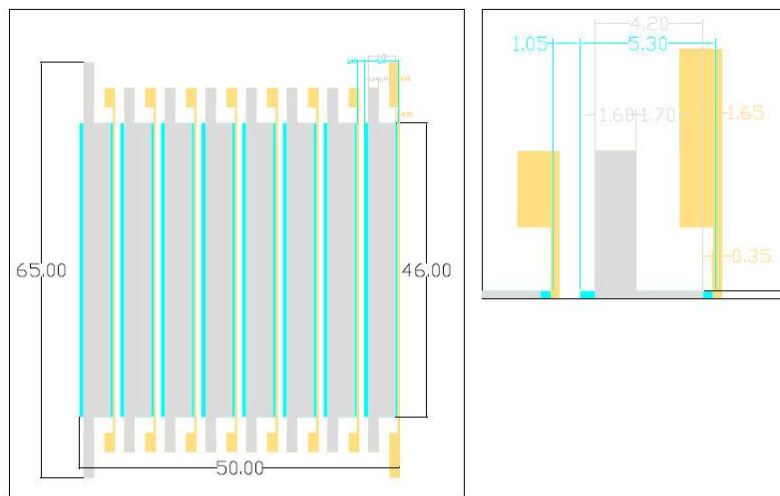


Figure 26. The layout and dimensions of the organic solar cell including detail of the pad structure at its corner.



Figure 27. A photograph of thin film battery.

### 3. Technology development: key achievements, problems faced and lessons learned

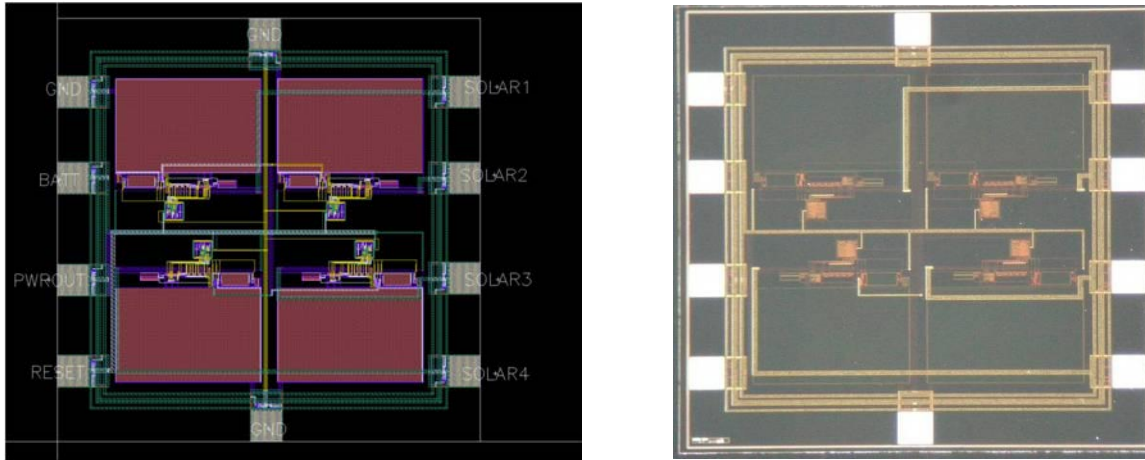


Figure 28. The layout of the controller chip (left) and a photograph (right).

The layout of system back plane was designed so that it allows flip-chip interconnection of the controller chip as shown in Figure 29. Because the pads on the chip are much smaller than those of the OSCs and battery, narrow conductors had to be used in the area of chip interconnection whereas in other parts of the backplane much wider conductors were utilized in order to lower the conductor resistances. The layout allows also an alternative way of assembly, where instead of connecting the chip directly to the backplane a separate strap substrate is utilized. In this case the ends of the thick parts of the conductors can be utilized as pads for strap substrate assembly. The strap substrates can be obtained eg. by cutting away the other parts except the inner region with chip interconnections from the backplane substrate. This strap substrate with a controller chip can then be assembled to the backplane substrate eg. using isotropically conducting adhesive. The strap substrate can be assembled either in face up position or face down position. Of course the battery has to be connected accordingly to fulfill the correct operation of the prototype device. The backplane layout was designed to be symmetrical with respect to its center line axis in the printing direction.

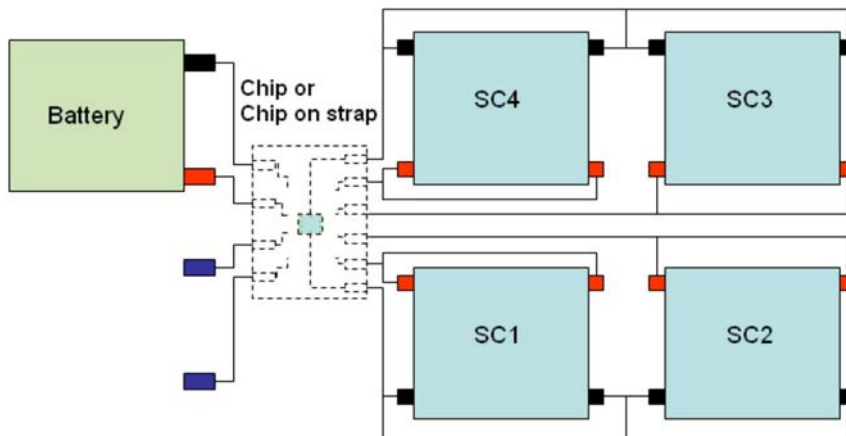


Figure 29. Principle of system integration for the demonstrator module.



### 3. Technology development: key achievements, problems faced and lessons learned

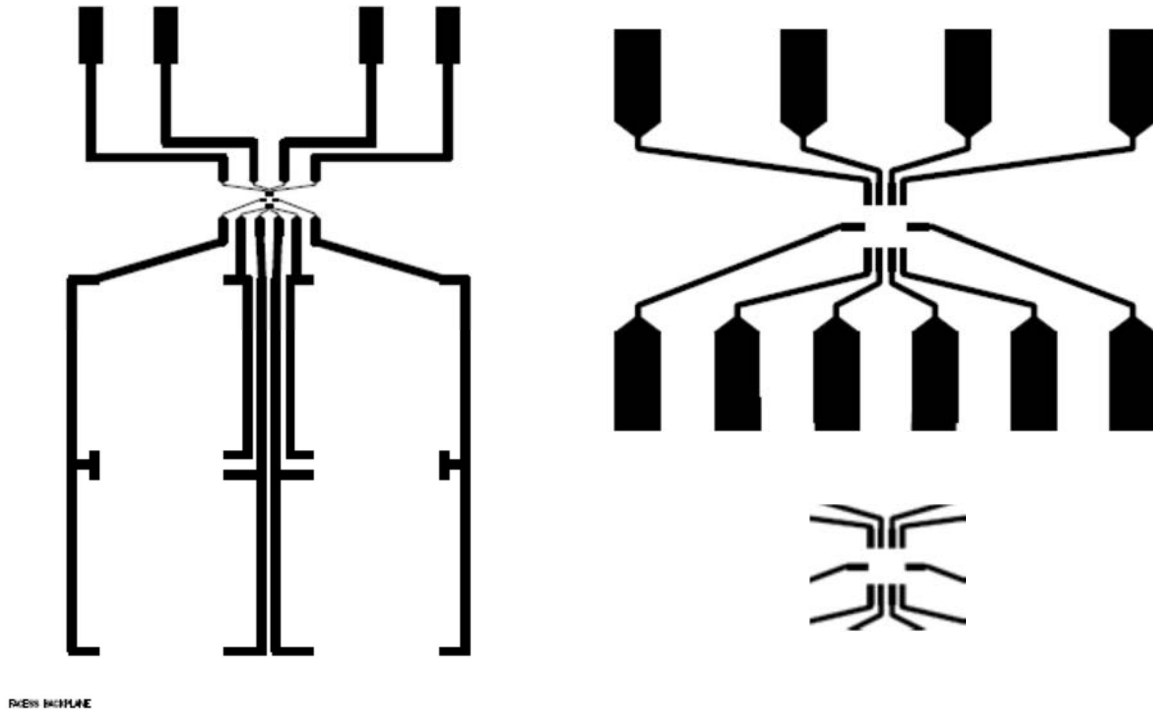


Figure 30. System backplane layout (left) and details of the pads for chip interconnection (right). The small strap structure shown in bottom right figure had to be used for assembly of controller electronics which was unintentionally assembled in a wrong orientation onto the backplane substrate.

In FACESS project, an Application Specific Integrated Circuit (ASIC) was designed and fabricated to handle the voltage conditioning requirements. The basic idea of the chip was to convert the voltage obtained by each of the solar cell sub-module so that it can charge the battery. Also, the chip needed to be thinned so that it can be embedded on a foil. Therefore, a solution based on integrated capacitors was chosen. The intended functioning of the chip was following. First, solar cell charges a capacitor up to its maximum power point. Then, the voltage of the lower voltage plate of the capacitor is increased until the voltage of the battery is obtained at the high voltage plate. And finally, charge is transferred to the battery. During the operational testing with FACESS solar cell module, battery and the designed ASIC, it was discovered that the ASIC did not work as intended. The circuitry charging the lower plate should have been powered from the solar cells, but since the selected technology does not operate under such low power voltages this could not happen and when powered from the battery, no net charging was obtained. Therefore, the end result was that the chip did not operate at voltages provided by current solar cell modules. Several alternatives were analysed. Firstly, a down-converting from a high-voltage module was analysed. The solar cell modules produce a voltage at the maximum power point (mpp) beyond the battery voltage, even under the lowest light conditions (0.001 sun). The solar cell voltage (mpp) is stored on a capacitor, the voltage is down-converted step by step until the voltage of the battery is obtained and then the obtained voltage is transferred to the battery. In this alternative, a mainstream low-cost chip manufacturing technology could be used, but a new concept

### 3. Technology development: key achievements, problems faced and lessons learned

would have needed to be elaborated and therefore, the complete system would have to be redesigned. The second alternative was to rework the present design in a low-voltage technology so that the logic part can be powered by the solar cells. In this case, a new prototype (design + production) would have taken an extra year which would have been beyond the end of the project and the cost of only the production would have been 4 times higher than budgeted for the original chip. The third alternative was to rework the present design in a low-voltage technology with external capacitors so that the chip cost could have been as low as in the original design. In this alternative, a technology for the capacitors on foil needs would have to be elaborated. In this case, smaller chips would have required completely different lamination techniques and therefore, the complete system would have been needed to be redesigned. Any of these three alternatives could have been realized in the last year of the project.

Therefore, two non-flexible backup solutions based on discrete components were implemented and analyzed. First solution was a simple comparator based switch-mode converter. Other solution was commercially available low power step-up converter with maximum power point control capability. The selection was made according to the following criteria:

- feasibility to integrate into an ASIC, including cost and footprint consideration
- efficiency in the FACESS application.

The comparator based switched mode battery charger was selected, designed and built in a circuit board sized  $24 \times 38$  mm including connectors for wires coming from the solar cell module and from the battery. Used comparators use very little power for operation and have wide operating voltage range, so they are well suited for this kind of application.

#### **Bonding process for OSC and TFB modules on the backplane**

In the FACESS project a low-temperature process for bonding the OSC modules to the backplane was developed. The adhesive paste was dispensed on the four pad areas on the backplane substrate corresponding the four contact pads in the corners of each OSC module. Then the OSC modules were assembled on the backplane and cured in the oven. The solution for the bonding process was found to be Eccobond 56C/Catalyst 9 conductive adhesive cured at  $+40$  °C for 4 hours in an inert atmosphere. Moreover, the connections were mechanically reinforced by bonding the body of the module on the backplane by UV curing adhesive Dymax 3089-GEL which was dispensed on the edges of the OSC modules and the curing was made using UV light source.

The thin film batteries can also be connected using isotropically conducting adhesive in same fashion as the OSC modules. The battery connections are made using thin copper (thickness  $12 \mu\text{m}$ ) and aluminium stripes (thickness  $40 \mu\text{m}$ ).

#### **3.4.2 System testing**

The system testing of flexible autonomous energy source was performed. The system testing included operational testing, environmental testing, lifetime testing and reliability testing. During operational testing it was discovered that the voltage conditioning chip intended for use on the backplane did not work as intended. Therefore, a backup switched mode battery charger was designed on a separate cir-

### 3. Technology development: key achievements, problems faced and lessons learned

cuit board to do the battery charging. During tests charging currents up to 530  $\mu\text{A}$  from a single solar cell module were measured in AM 1.5 conditions.

The interconnections of the OSC modules were tested in thermal humidity chamber at  $+85\text{ }^\circ\text{C} / 85\text{ \%RH}$  and in thermal cycling  $-50\text{ }^\circ\text{C} \dots +125\text{ }^\circ\text{C}$  for a time of 5 days. For thermal cycling the test included about 100 cycles. No degradation was observed in these reliability tests. Therefore it can be concluded that the interconnections between OSC modules and backplane can fulfill the set application requirements. However, it was also concluded that no bigger bending radius than 80 mm can be at the moment recommended.

For the battery interconnections reliability tests were made for the thin copper (thickness 12  $\mu\text{m}$ ) and aluminium (thickness 40  $\mu\text{m}$ ) stripes bonded to the pads of the backplane substrate using the same adhesive as used for bonding the OSCs. It can be concluded that the Cu stripes survived the test well contrary to the Al stripes. It can be recommended that thicker Cu foils should be used due difficult handling of so thin striped during assembly of the batteries on the backplane. Furthermore, the Al stripe material should be changed to nickel or at least Ni plating should be used. It came clear, that more attention should have been put earlier to the alternative thickness and metallisation of the TFB stripes.

Measurements in zero light intensity and in simulated solar light intensity were done to verify the operation and performance of the circuit. The battery charging was successfully demonstrated. Then the functionality of the system by charging an empty battery with a OSCs backplane in outdoor conditions was demonstrated successfully.

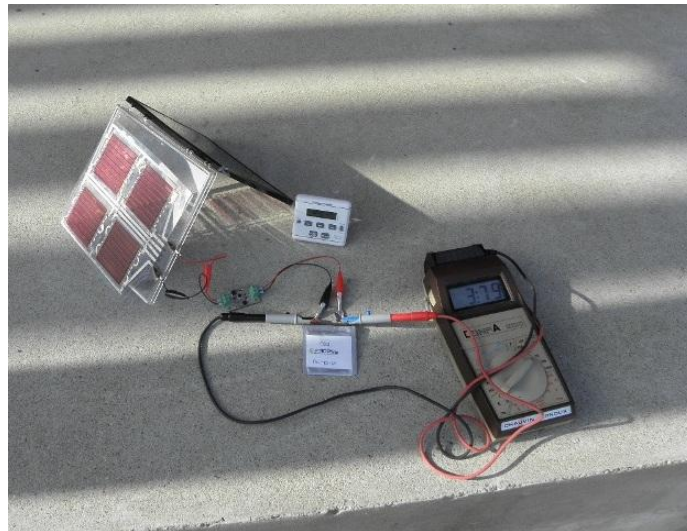


Figure 31. Protocol of charge of the battery with the OSC backplane in outdoor conditions.

### 3. Technology development: key achievements, problems faced and lessons learned

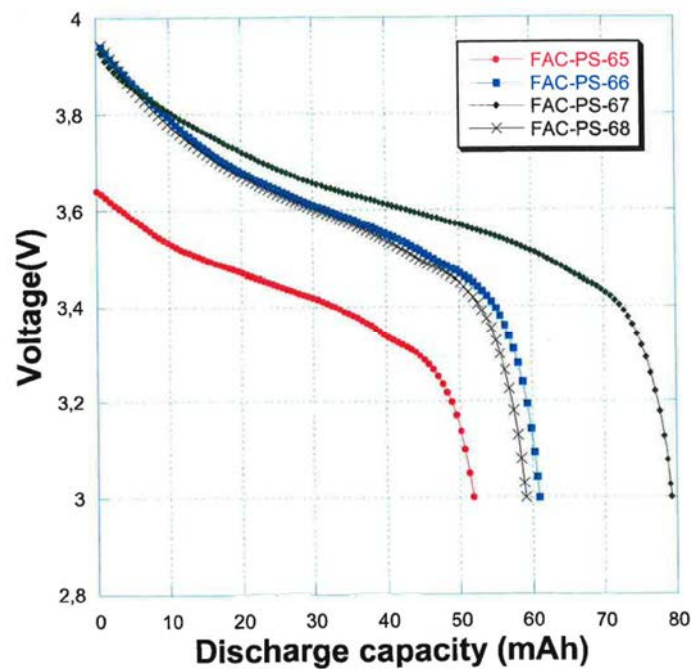


Figure 32. Discharge capacity of the battery between [3-4V].

The installed OSC backplane delivered a  $V_{max}$  of about 6.8 V in the sunny outdoor conditions. The pyranometer gave an illumination of  $93 \text{ mW/cm}^2$  when the  $I_{max}$  of the OSC backplane was 19.8 mA.

The voltage of the battery was then regularly recorded till the end of the experiment when the battery was fully charged. The curve of charge of the battery is presented in Figure 33. We can see that after 95 minutes of experiment, the charge doesn't work anymore as the illumination has dramatically dropped to  $12 \text{ mW/cm}^2$ . The voltage of the panel under this illumination is too low to ensure the charge of the battery. The experiment was pursued the day after. The illumination is of  $92 \text{ mW/cm}^2$ , the charge can re-start. When the illumination decreases to  $70 \text{ mW/cm}^2$ , the charge is slower but continues up to 3.8 V. This experiment demonstrated the functionality of the entire system in real outdoor conditions.

### 3. Technology development: key achievements, problems faced and lessons learned

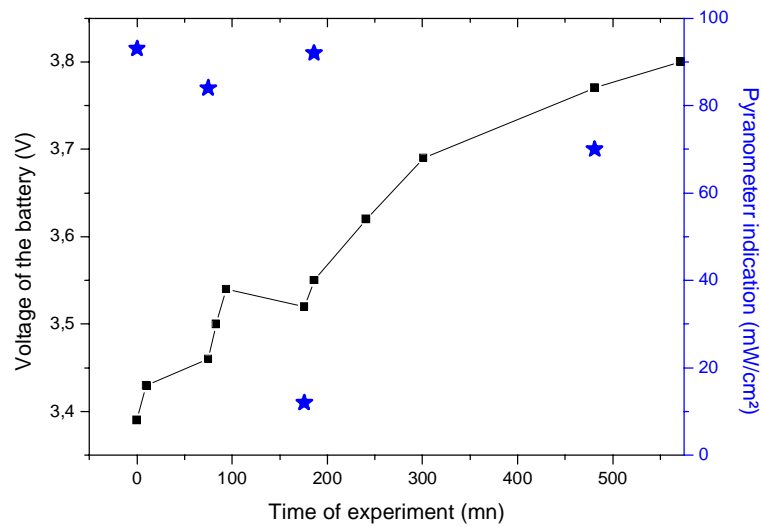


Figure 33. Curve of charge of the battery and indications of the pyranometer in function of the time of experiment in outdoor conditions.

The same experiment, charge of the battery with the OSC backplane, was performed into the solar simulator with the same complete system. Two cycles of charge were realized. The IV curves of the module was monitored and the voltage of the battery as well. At the beginning of the experiment, the charge of the battery is below 3V. When the module was connected to the charger, its voltage  $V_{max}$  is controlled at ca. 2 V by the charger but the charge of the battery can occurred. The battery was then disconnected in order to be discharged manually into a resistance. Then the voltage recorded was the one of the module and goes up to 3.7V.

### 3. Technology development: key achievements, problems faced and lessons learned

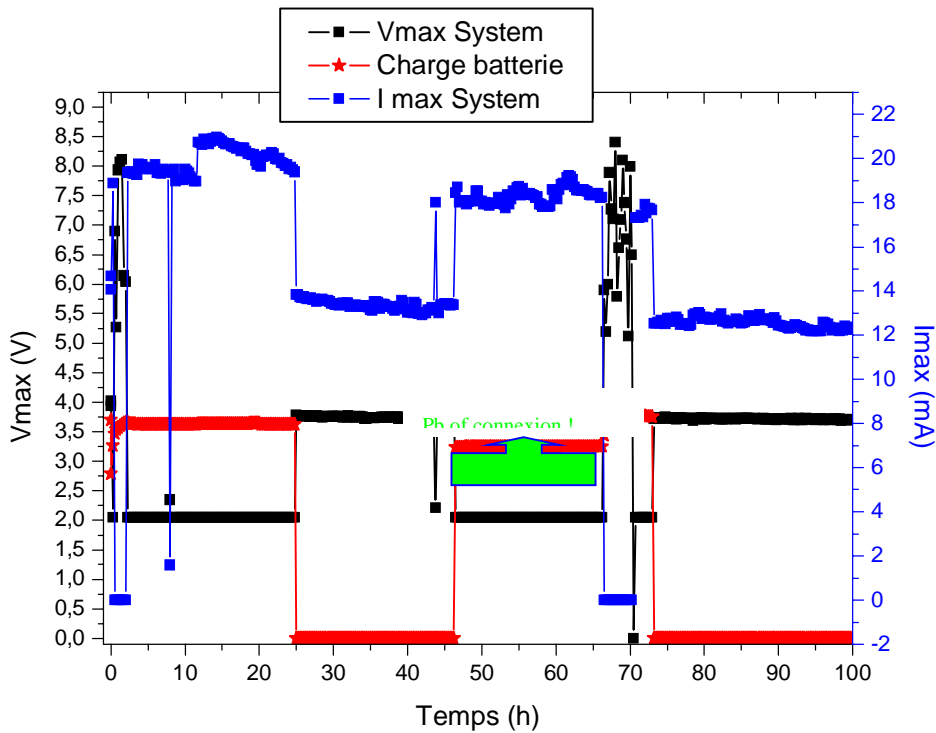


Figure 34. Evolution of the Vmax and Imax of the organic module during the charge or discharge of a battery. The same backplane was used for the following part, for the measurement of the system lifetime.

The results obtained on the backplane are presented in Figure 35. The evolution of the attached modules onto the backplane is the same than the individual modules. The lifetime of the system can be then estimated at about 7 months (@50% of PCE(t0)).

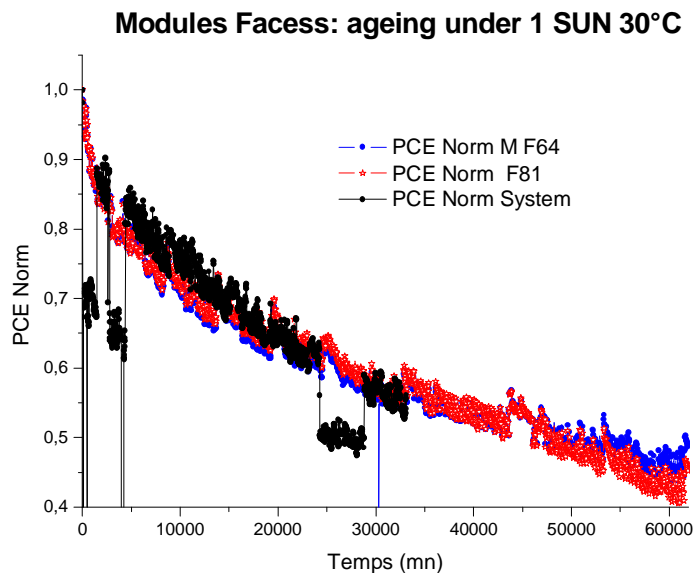


Figure 35. Evolution of the normalized PCE of the attached modules onto the backplane in comparison with the individual modules under accelerated ageing.

### 3.4.3 Feasibility for R2R manufactured OSCs and TFBs onto the backplane

There were three development stages identified for the system integration as described in Figure 36. During the project methods of integration of sub-modules was studied. It was also envisioned how the whole process flow for R2R printing of OSC and TFB would be manufactured directly onto the backplane substrate. Only the Si-chip would be then bonded afterwards.

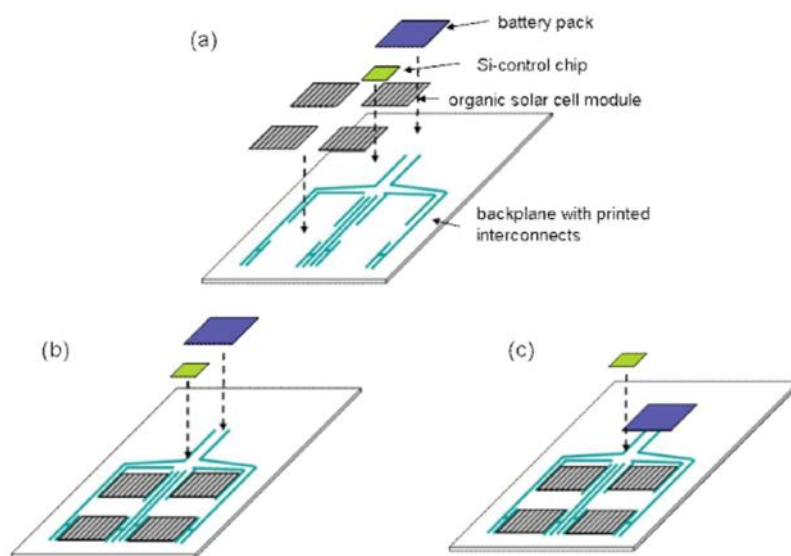


Figure 36. Three development stages for system integration in FACESS project are illustrated: (a) all modules joined to a common back plane (b) OSCs printed on back plane substrate, (c) OSCs and thin film batteries printed onto the backplane substrate. For each case the chip including control electronics shall be joined to the back plane.

As a conclusion, the potential scenario of R2R manufacturing of OSC modules, thin film batteries and chip assembly was illustrated. It shows the possibility to fabricate all the individual components on the same carrier foil in order to realise FACESS demonstrators. The processing can be started with the patterning of ITO plastic foil, which may have barrier properties to ensure long lifetime of OSC modules. The OSC modules are processed as developed in the project followed by encapsulation phase. Since evaporated metal layer is used in the OSC module structure, the TFB positive metal electrode layer can be deposited in parallel, which is clear advantage in the manufacturing cost point of view. After the encapsulation of OSC modules, the printing of positive ink can be realized with a printing tool, which only covers the width of battery since the thickness of encapsulation foil may interrupt the printability.

Since the membrane ink, composed of mineral fillers mixed with PVdF, cannot be deposited directly onto  $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  layer, it is recommended to pre-process the foil with a membrane layer and printed negative ink on a copper foil. Thereby, the foil can be laminated onto the carrier foil. As a final process phase, the chips can be bonded from the stripe onto the carrier foil.

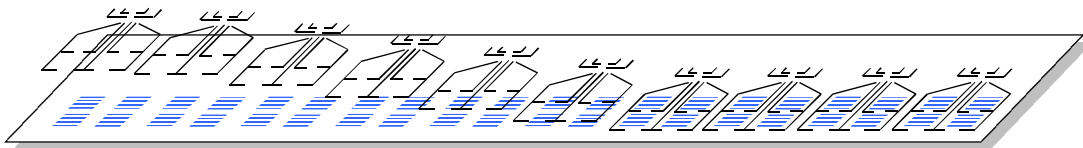
### 3. Technology development: key achievements, problems faced and lessons learned

Table 13. Potential future scenario of R2R manufacturing of OSC modules, thin film batteries and chip assembly.

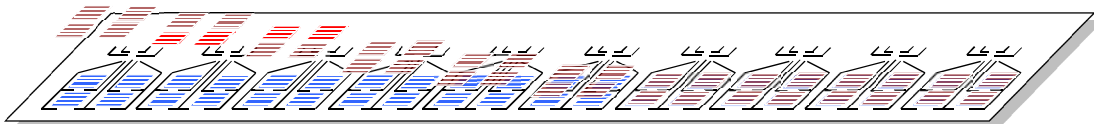
Process phase



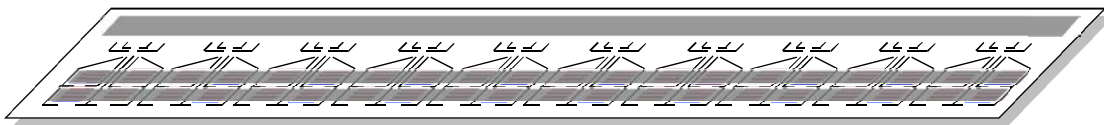
1) The patterning of ITO foil.



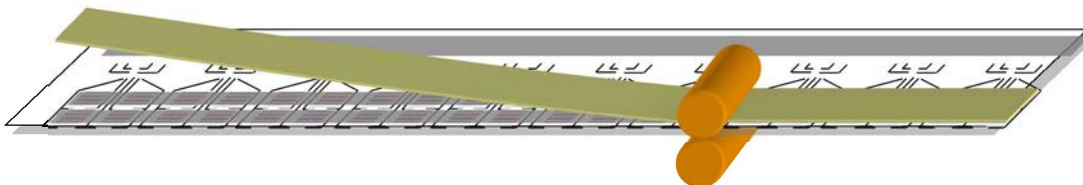
2) Rotary screen printing of backplane conductors.



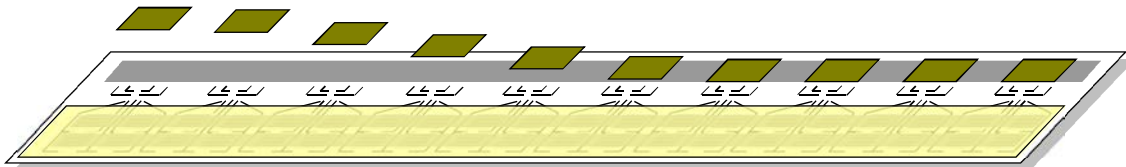
3) Gravure printing of PEDOT:PSS and P3HT:PCBM layers.



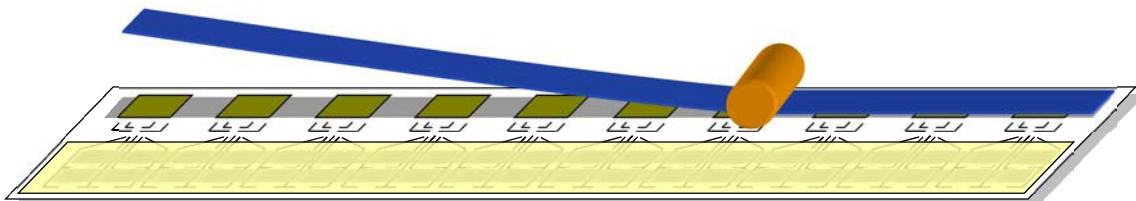
4) Vacuum deposition of aluminium metal for OSC top electrode and TFB positive electrode.



5) Encapsulation via foil lamination of OSC modules.



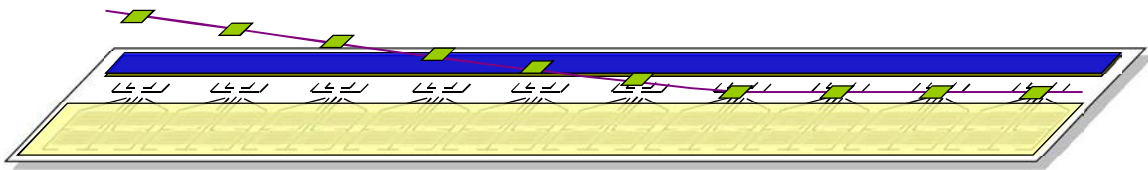
6) The printing of positive ink (lithiated metal oxide)



7) The lamination of Cu foil with printed negative ink (graphite) and the membrane ink



### 3. Technology development: key achievements, problems faced and lessons learned



8) The chip assembly using adhesive flip chip bonding.

#### 3.4.4 Recyclability

The goal of the FACESS recyclability study was to describe the recyclability of the FACESS module. Firstly recycling was discussed in general including legislation and issues hampering recycling. Secondly the recycling of WEEE, Waste Electrical and Electronic Equipment, is further described focusing on the treatment of the FACESS module with current technologies.

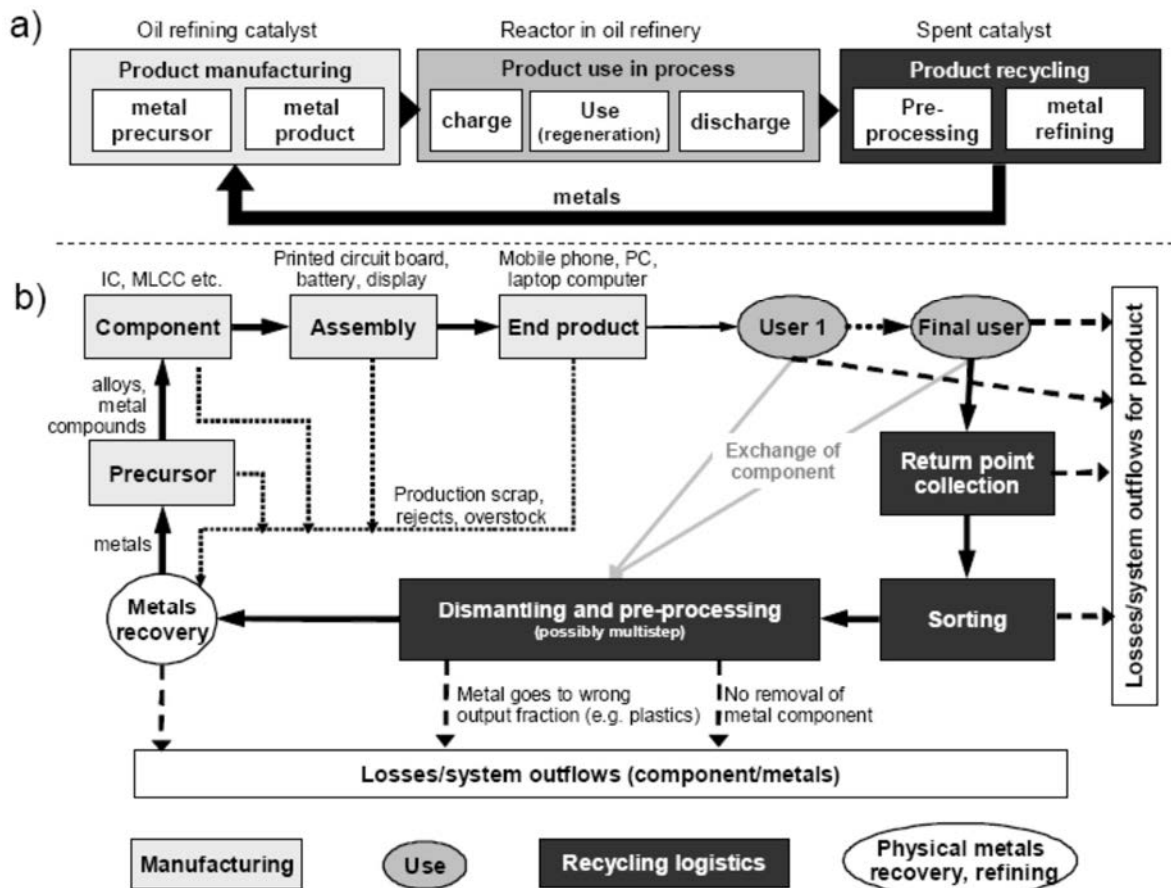


Figure 37. Closed loop systems for industrial applications (example process catalyst) versus b) Open loop systems for consumer goods (example consumer electronics).

3. Technology development: key achievements, problems faced and lessons learned

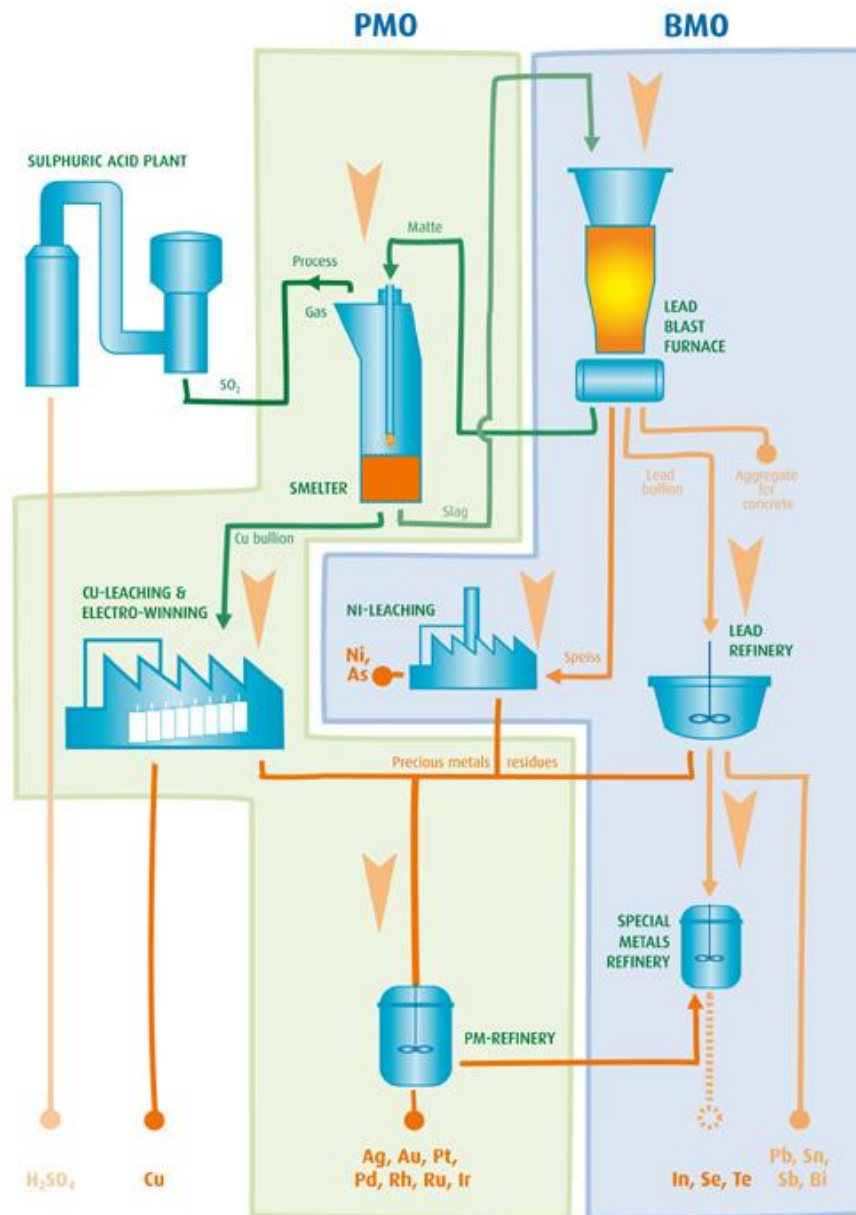


Figure 38. The Umicore precious metals recycling flow sheet.

The materials content of the FACESS module is shown in Table 14. As is typically for WEEE, the metal content of the module is low (less than 6 wt%). The remaining materials are plastics. Thus one ton of waste FACESS module contains 18.12 kg of Si, 9.7 kg Cu, 5.5 kg Al, 5.1 kg Ni, 5.0 kg Co, 4.6 kg Mn, 1.9 kg Li, 0.2 kg In, 0.07 kg Ag, 0.03 kg Ca en 0.01 kg Sn.

### 3. Technology development: key achievements, problems faced and lessons learned

Table 14. Metals content of the FACESS module.

composition module	In ppm	Sn ppm	Ca ppm	Ag ppm	Li wt%	Co wt%	Mn wt%	Ni wt%	Al wt%	Cu wt%	Si wt%	plastics wt%
with plastics	230	10	30	70	0,19	0,50	0,46	0,51	0,55	0,97	1,81	95

The FACESS module can be considered to be a complex WEEE. Such materials are typically treated in integrated smelters (such as the Umicore flowsheet, see Figure 42) in order to optimally recover a number of components. A first separation occurs by treating the material in a pyrometallurgical process in which part of the metals (“the impurities”) are put in a slag phase as oxides and the other part as metals in a metal phase. Both streams are further treated to recover the valuable metals. The division between slag and metal phase depends the process parameters such as temperature, oxygen partial pressure and charge composition. Consequently, a good understanding of the process is needed to optimally concentrate In in one of the process streams. To treat such the FACESS module, specialized technology is needed.

The FACESS module can be recycled if a good collector system is set up. Currently, mostly for consumer goods such as small WEEE, collecting is the weakest link in the recycling chain (see also figure 41). After collection, the material recovery can be done in an integrated smelter. For the FACESS module there are existing processes such as the UHT process to recycle batteries available in which In, Sn, Ag, Co, Ni and Cu metals can be recovered while the plastics are valorized as energy. Further research is going on for the UHT process to in the future also recover Li and Mn.

#### 3.4.5 Demonstrator

The aim of the demonstrator system was to prove the ability to gather energy using printed solar cells developed during FACESS project and to utilize that energy to do feasible operations, e.g. measurements as in this case. Furthermore, a flexible battery also developed during FACESS project was used as an energy storage element to store energy to be used while momentary energy consumption exceeds the energy produced by the solar cells.

The Flexible autonomous cost efficient energy source element system demonstrator was designed and manufactured. The FACESS demonstrator system consists of following physical parts:

1. Four solar cell modules attached on a flexible backplane with printed conductors
2. Flexible battery
3. Charging electronics and sensors on one circuit board
4. A separate circuit board attached on top of the first board for microcontroller and radio module
5. Mechanical enclosure connecting different parts mechanically and electrically
6. A separately powered base station for data display.

The environment monitoring electronics measures humidity, temperature and illumination values by two digital sensors that are connected to a microcontroller located in the radio module. The maximum tolerance of humidity and temperature samples are  $\pm 2\%$  RH and  $\pm 0.3^\circ\text{C}$ , respectively. The sensor operates at 2.4–5.5 V voltage range and the current consumption is 0.3  $\mu\text{A}$  on sleep mode and 0.55 mA during measurement. Illumination is measured by TAOS TSL2550D ambient light sensor. The sensor operates at 2.7– 5.5 V voltage range and the current consumption is 10  $\mu\text{A}$  at sleep mode and 0.35 mA

3. Technology development: key achievements, problems faced and lessons learned

at active mode. The base station consists of a Jennic controller board and JN5048-001-M00 wireless microcontroller module. The base station operates as the PAN coordinator of an IEEE802.15.4 star network, and receives the sensor samples from wireless sensors. Totally, four wireless sensors can be connected to the single base station.

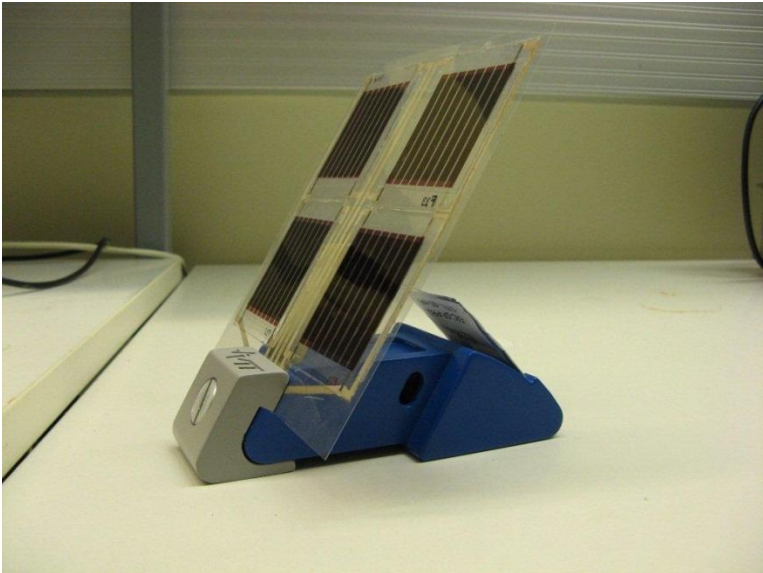


Figure 39. FACESS demonstrator system.

The key characteristics of the FACESS system demonstrator power source element are presented in Table 15.

Table 15. The key characteristics of the FACESS system demonstrator power source element.

Parameter	Typical values
Backplane size	150 mm x 150 mm
Backplane weight	13 g
Battery size	60 mm x 60 mm x 0.5 mm
Battery weight	3.4 g
Battery capacity	56 mAh
$V_{oc}$ from backplane (2P2S backplane) @ AM 1.5	8.4 V
$P_{max}$ from backplane (2P2S backplane) @ AM 1.5	150 mW
Demonstrator current consumption	0.7 mA

## **4. Vision towards large-scale production of flexible organic solar cells, thin film batteries and power source and storage systems**

### **4.1 Exploitation of project results**

During the FACESS project 9 journal articles were published and 27 papers were presented. in various conferences. The publications and venues where project results have been presented have been listed in Appendix 1 and 2. Extensive dissemination of the project results during the project have created an excellent global visibility and awareness for both the developed technology and consortium members, which gives a very good base for the future exploitation plans and actions towards development of products and large-scale production of flexible organic solar cells, thin-film batteries and related power source and storage systems.

### **4.2 Benefit to European industry and economy by the FACESS exploitation**

In the following the visions and planned actions for the further exploitation of the project results by the partners in the FACESS consortium have been discussed. The impact and potential of the technology to the European industry and economy have been also discussed.

4. Vision towards large-scale production of flexible organic solar cells, thin film batteries and power source and storage systems

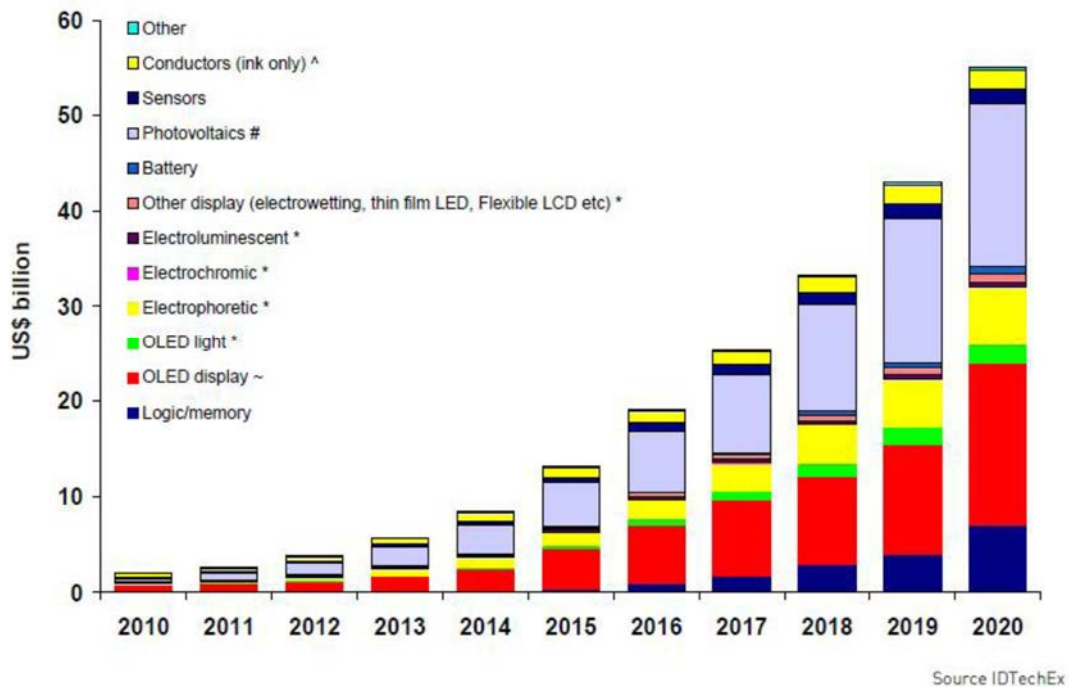


Figure 40. Global market for printed electronics (IDTechEx 2010).

According to IDTechEx 2010 estimation on the global market for printed electronics, the Solar cells market is representing the highest potential of all component types. The market entry of organic solar cells within next 3 year is possible but as has been discussed earlier, several critical obstacles with performance, lifetime and cost need to be still solved that this can be realised. For the printed batteries the market entry is estimated to following bit later.

The fastest time-to-market with FACESS developments is clearly the machinery and process know-how that can be utilised in business of Coatema. Coatema is already discussing several projects worldwide with units similar to the FACESS project since late 2010 and got already one order from Europe on a similar concept. Coatema see a significant market advantage against competitors out of Japan and Korea with this type of technology. Coatema see a opportunity as an SME to increase the market share significantly and protect against Asian competition. With the background of the FACESS project COATEMA is designing specific R2R units dedicated to large area printed electronics in different working widths from 150mm, 300 mm up to 600mm in nitrogen atmosphere. Coatema are now able to offer a complete new oven design with a minimum amount of nitrogen consumption and precise temperature control for different processes. During the 3 years a very close co-operation with VTT and CEA developed and also with industrial partners like Umicore there are regular discussions on new technologies. The estimated impact on Coatema's overall sales volume per year for the next 3 years could be around 20% in using FACESS related technologies.

For Umicore the project has shown as a proof of concept that existing cathode materials could be used in the context of the FACESS modules. The concerned material can therefore be available in our portfolio of commercial products. On the longer term, the recycling of this type of module is possible

#### 4. Vision towards large-scale production of flexible organic solar cells, thin film batteries and power source and storage systems

with the Umicore technology. Moreover, the results can be extrapolated to other types of printed functionality devices, further broadening the scope of our recycling activities.

From research organisations VTT is already signed the first commercial continuation project contracts for further development of the Organic Solar Cells technology to the commercial stage with global companies and several new deals are under negotiations. With the latest developments it has been becoming realistic to believe that the VTT OSC related knowhow and infrastructure will be one of the major offering for the VTT's global contract research business in OLAE field. The following offering for further contract research with companies developed in the FACESS project:

- OSC related research and consulting
  - OSC R2R production trials using pilot production equipment
  - laboratory scale material testing in different processing methods
  - laboratory scale material formulation for printability, printing trials with novel low band gap photo active materials
  - training of researchers and machine operators in the field of R2R fabrication
  - encapsulation and testing of barrier foils in VTT's R2R printing process
  - OSC market study and roadmapping, IP analysis
- R2R TCO patterning process
- System integration to various applications.

The VTT's near future plans for OSC offering development will be focused on

- R2R up-scaling in pilot line environment
- low band gap photo active materials
- alternative barrier material such ALD and flexible glass
- invert OSC pilot processing incl. hole blocking layer development, lamination and stability
- wet processable transparent interlayers based on ZnO/TiO
- low work function cathode ink for OSC
- ITO replacement
- submicron resolution (also for multilayer structures): active thin film thickness  $< 100 \text{ nm} \pm 5 \text{ nm}$
- special attention to system level testing and reliability.

The commercialisation of OSC developments can be also promoted via the Printocent community <http://www.printocent.net/>. PrintoCent is a VTT lead initiative to create business and production environment for companies. The operational mode is focused on manufacturing product demonstrators and based on Printed Intelligent processes. Today, PrintoCent is truly a functioning multidisciplinary business development environment with around 150 professionals working in the community. PrintoCent's Pilot Factory is a manufacturing environment designed for companies who are interested in taking developments in Printed Intelligence closer to the market place and towards commercial pro-

#### 4. Vision towards large-scale production of flexible organic solar cells, thin film batteries and power source and storage systems

duction environment. PrintoCent's pilot production reduces commercial and technical risk before fully commercial operation kick off. PrintoCent is located in Oulu, Finland.

Via Printocent large amount of companies can be contacted. As an example PRINSE'10 Industry Seminar held on 23<sup>rd</sup>-24<sup>th</sup> Nov 2010 collected 150 participants from 65 companies. The event was organised by VTT gathered together participants from different sectors of industry including brand owners, end users, OEMs and subcontractors in the frame of Printed Intelligence. Lot of demonstrators e.g. FACESS was shown to create new product ideas and discussions. Seminar presentations from industry participants introduced the different customer sector requirements and needs. Seminar presentations of researchers introduced technology capability and possibilities. Workshop discussions between industry representatives and researchers were held to find out new applications, products and possibilities for Printed Intelligence.

R2R etching and evaporation development have also been a major asset for VTT\*s OLED commercialisation efforts. Moreover, several EU and TEKES funded research projects focusing especially R2R processing of multi junction OSC devices has been started or to be started utilising the knowhow generated during the FACESS project, which means that broader amount of companies will be benefiting in long run.

The FACESS project has created for IMEC the opportunity to work on new developments in two main fields:

- OSC processing and
- System-on-foil integration aspects.

For the OSC processing, the FACESS project helped IMEC to extend its developments from the single cell level into full module processing. On the one hand, module designs and layouts have been further optimized based on modelling aspects taking e.g. series resistance effect induced by electrodes and electrode configurations into account. On the other hand, actual processing steps have been further elaborated to improve the active area coverage and the deposition of metal electrodes from solutions and inks. Participation in the FACESS project offered the opportunity to carry out these developments on larger area substrates and flexible foils.

Therefore, the FACESS project has helped IMEC to initiate new activities in the field of OSC:

- It is strongly believed that OSC device efficiency can be further improved by building multijunction structures. This has been already an activity in which IMEC has been engaged, but based on outcomes of the FACESS project it will be possible to also extend the multijunction architecture from single cell level further in full module architectures. IMEC has engaged, as coordinator, in a new European project proposal that deals with this subject in OSC.
- Additionally, IMEC has started discussions with the Dutch research centers TNO and ECN to create joined, transnational thin-film PV-platforms, including OSC, to enlarge the impact of the regional activity in this field. This collaboration will be formally started in due time under the heading of the Solliance initiative. The experience and knowledge build up in the FACESS project has allowed IMEC to take a strong position in these discussions.



#### 4. Vision towards large-scale production of flexible organic solar cells, thin film batteries and power source and storage systems

- Finally, the module developments and experiences in the FACESS project have allowed IMEC to start bilateral collaborations with industrial partners in the field of OSC. One aspect thereby is to demonstrate through a short-track project the feasibility for commercialization of new module concepts.

In the FACESS project, the work of IMEC-Ghent was mainly focused on the assembly of thin silicon chips on a plastic foil. The innovative aspect of this work for our organization was the low temperature aspect of the assembly combined with the fast processing times and the need for flexibility of the assembly due to the roll-to-roll process.

FACESS project gave IMEC the opportunity to broaden the scope of an existing patent (EP1126517 – assembly using both isotropic and non-conductive adhesive) to low temperature assembly. Next to that the work on the interconnecting using anisotropic conductive adhesive is continued in the frame of the Holst institute. Within the technical program “Integration Technologies for Flexible Systems” specific interest is shown in this technology. In the beginning of 2011 Henkel Electronic Materials (Westerlo, Belgium), a partner in this technical program, has started to develop their first anisotropic conductive paste for use in low temperature electronics

Suntrica has prepared a detailed exploitation plan with competition analysis to other solar cells technologies and identification of few concrete application cases with requirement specification for energy source and storage. As a summary, Suntrica’s exploitation plan has focused on the expected benefits of FACESS technology. Assumption has been, that the efficiency will be lower than in competitive technologies, but FACESS will benefit from integrated battery, light-weight, space saving, reliable structure, and lower manufacturing costs. The focus in further development towards products should be in the following critical success factors that have a direct and serious impact on the viability of the technology. In addition to the performance and lifetime requirements, there is an imperative need to implement easy and low cost recycling of the integrated battery in order to make the FACESS technology acceptable and competitive in the world markets. The CSFs are at least the following:

- Efficiency: minimum 50% versus competing technologies. Target is 2.5–5%.
- Cost: Better than competition. Target is 2–3 €/W. Battery may increase cost slightly.
- Lifetime: over 3 years
- High quality battery integration to the same foil will improve competitiveness.

### **4.3 From roll-to-roll to roll-to-product – Taking printed intelligence developments out of the lab into products**

In a bigger perspective, during the FACESS project developments the printed electronics and intelligence has been a technology-intensive industry in its early stages, and it is undoubtedly very excited about the future prospects of its printed components, thin film layers, etc. These developments have largely been driven by technical research institutions and R&D departments of companies. The visions of the industry have therefore also been technology heavy and the developments in a predominantly technology push mode. Product developers have been unfortunately few, and the end market needs (specification data sheets) that the industry has been trying to satisfy have therefore perhaps, in parts, been too strongly influenced by well-known and established technological component benchmarks.

4. Vision towards large-scale production of flexible organic solar cells, thin film batteries and power source and storage systems

Technological printed intelligence developments and related infrastructure has been strongly invested in Europe and serious efforts to identify factors for technology pull (market needs and opportunities) has been done in various business arenas also beyond the electronics sector. Despite such efforts, the number of end products developed for/with end customers has been rather limited. This is partly because the expectations and excitement of potential end customers – when first hearing of printed electronics – also often heighten to levels that current technological capabilities are not yet able to meet. When balancing market needs and wants with current technological capabilities, it can be said that printed intelligence technologies have the potential to disrupt and expand end markets. Ultimately, many first market executions start off as niche applications – which may still throw off many established industrial players – but these could rapidly grow to new heights. Still, strong actions are needed for commercialisation of printed intelligence. The aim of these actions is to introduce successfully technologies from the laboratory to early market trials and commercial adoption.

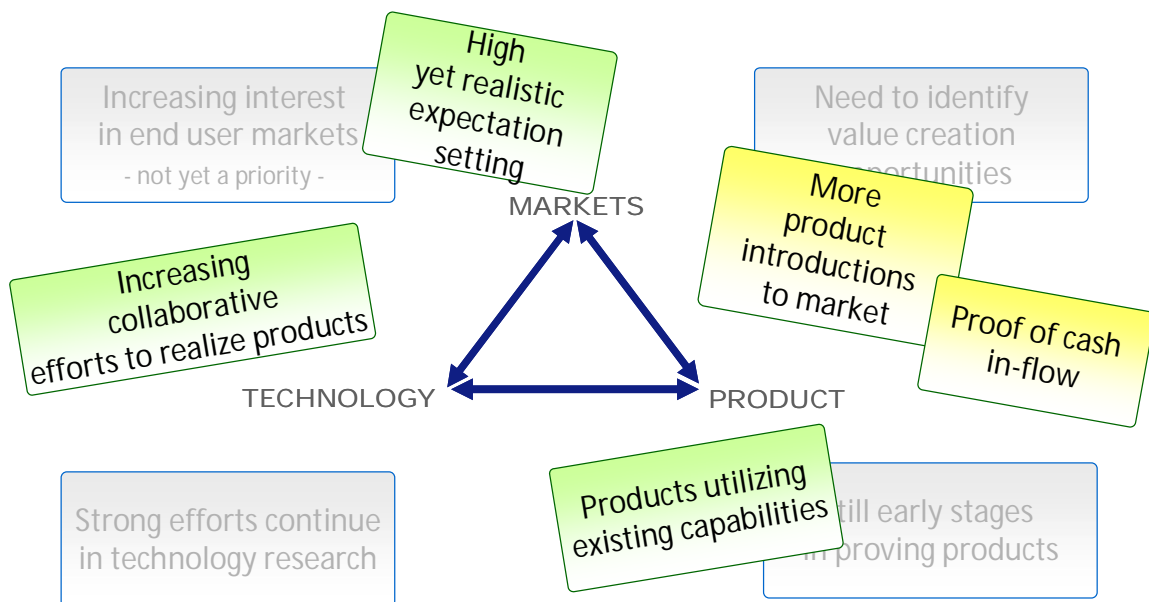


Figure 41. Trends in printed intelligence speeding up developments toward product.

The facilities in Europe are being built toward more pilot-scale production capabilities, allowing the industry to demonstrate not only proof of technology but also more proof of industrial-scale production. The end components are embedded in end devices that are tested in the markets by end customers – providing more proof of product based in part on printed intelligence. The emerging printed intelligence industry and its customer value chains require new types of solution providers and system integrators offering new business opportunities. Collaboration among globally leading companies as well as smaller companies that engage in joint demonstrator and piloting projects is needed. The printed intelligence community also needs to enforce supporting infrastructure in the areas of education, research and financing. In addition to industrial money, these efforts are strongly backed by regional and national public funding sources, including the EU.

#### 4. Vision towards large-scale production of flexible organic solar cells, thin film batteries and power source and storage systems

With piloting environments, the printed intelligence community is able to develop and manufacture prototype products on the first industrial scale. This allows companies to reduce the commercial and technical risk before investing in their dedicated commercial-scale production lines.

Materials companies have carried much of the industrial weight in developments to date in the printed electronics and printed intelligence industry. This is only natural, as materials are a key enabler. These companies are therefore in an excellent position to capture significant added value once the end product markets take off. As an industry, however, we have so far struggled to develop products that significantly satisfy existing market needs and wants. Cash inflow to companies in the field has therefore still been limited. Material cost reduction path with increasing volumes is however essential for ensuring increase of pilot-scale manufacturing trials and ultimately, improving the business case for new products. While there is continued excitement and active research efforts to improve printed component performance, more effective and environmentally friendly next-generation materials and processing techniques, ever more product and market orientation is instilled into these developments. In other words, companies and institutions are seeking more actively to apply laboratory-scale, proven technologies and capabilities to the development of new products and systems. This is also an opportune time for process equipment suppliers, system integrators, end product developers and producers to define and capture their roles in the evolving supply and customer value chains. Potential end customers for new printed technologies are also learning in practice about the status (the possibilities as well as the limitations) of new printed technologies. New innovation around near-term product possibilities is taking root. Product ideas are rapidly being demonstrated and tested in the market and efforts to scale up to production are on the way.

We hope this report encourages innovative companies and people with entrepreneurial spirit to continue to actively approach us to learn about these emerging technological possibilities and to take them toward products in commercial use.



## Appendix 1: FACESS dissemination activities

FACESS project published 9 journal articles, 27 papers in various conferences. Project results has been presented in following publications:

1. Hast, J. Jussila, S. Flexible autonomous cost efficient energy source and storage – FACESS project, ISFOE09, (invited)
2. Rouault, H. et al. Development of electrodes for printed Li-ion Thin Film Batteries, ISFOE09.
3. Tuomikoski, M. et al. Roll-to-Roll Manufacturing of Organic Photovoltaic Modules, EMPC09.
4. Jin, H., Tuomikoski, M., Hiltunen, J., Kopola, P., Maaninen, A. & Pino F. 2009. Polymer-electrode interfacial effect on photovoltaic performances in P3HT:PCBM-based solar cells, J. Phys. Chem. C 113, 16807–16810.
5. Jin, H., Olkkonen, J., Tuomikoski, M., Kopola, P., Maaninen, A. & Hast, J. 2009. Published on line: A few fabrication processes having an effect on the performance of P3HT:PCBM-based solar cells, Sol. Energ. Mater. Sol. Cel, doi:10.1016/j.solmat.2009.11.007.
6. Jin, H., Hou, Y., Teng, F., Kopola, P., Tuomikoski, M. & Maaninen, A. 2009. Effect of molecular aggregation by thermal treatment on photovoltaic properties of MEH-PPV: Fullerene-based solar cells, Sol. Energ. Mater. Sol. Cel. 93, 289–294.
7. Kopola, P. et. al., High efficient plastic solar cells fabricated with a high-throughput gravure printing method, Solar Energy Materials and Solar Cells, 10.1016/j.solmat.2010.05.027.
8. Kopola, P. et. al. Gravure printed flexible organic photovoltaic modules, Solar Energy Materials and Solar Cells, 10.1016/j.solmat.2010.12.020.
9. Dittrich, C. R2R Technologies for Flexible Electronics. LOPE-C, Frankfurt, Germany.
10. Kemppainen et al. Large area printed smart systems – from components to systems. Plastic Electronics Asia 2009. Taiwan. (invited)

## Appendix 1: FACESS dissemination activities

11. Kemppainen et al., Large area printed sensors: system and manufacturing integration, 5th Global Plastic Electronics Conference and Exhibition. 27–29 October 2009, Dresden, Germany.
12. Hast, J. et al. Roll-to-roll manufacturing of large area flexible organic photovoltaics modules. OPV Summit 09, Boston, USA (invited)
13. Hast, J. FACESS project presentation, Organic and Large Area Electronics, Clustering Meeting 13th January 2009, Brussels, Belgium.
14. Hast, J. et al. Toward roll-to-roll printed power sources and control electronics, Photonics Spectra. 2010. Kolbusch “Roll to Roll Technologies for Flexible Electronics”, SIMTech-Workshop 09, Singapore, Singapore (invited)
15. Kemppainen et al. Large area printed sensors – systems and manufacturing integration. Plastic Electronics Europe 2009. Dresden. (invited)
16. Kolbusch, T. R2R Technologies for Flexible Electronics. Fraunhofer IZM-Forum be-flexible, Munich, Germany.
17. Siekierski, M., Żukowska, G., Plewa-Marczewska, A., Piszcz, M. Makromolekularne receptory na aniony jako dodatki do elektrolitów niewodnych. VI<sup>th</sup> Congress of Chemical Technology – Warsaw 21–24 June 2009.
18. Marczewski, M., Piszcz, M., Plewa-Marczewska, A. & Siekierski, M. Charakteryzacja elektrolitów polimerowych opartych na metyloalumoksanach i glikolach polietylenowych. VI<sup>th</sup> Congress of Chemical Technology – Warsaw 21–24 June 2009.
19. Piszcz, M., Marczewska, A., Bukat, M., Kalita, M., Ryś, P., Żukowska, G. & Siekierski, M. Electrochemical investigation correlated with FT-IR and NMR spectroscopy of liquid battery electrolytes containing supramolecular additives. 216<sup>th</sup> ECS Meeting: EuroCVD 17 & SOFC-XI: Vienna Austria.
20. Marczewski, M., Piszcz, M., Plewa-Marczewska, A. & Siekierski, M. Synthesis of Water Scavenging Compounds Based on Functionalization of Methylalumoxane by Organic Compounds Containing Free Hydroxyl Group for Application in Battery Electrolytes. 216<sup>th</sup> ECS Meeting: EuroCVD 17 & SOFC-XI: Vienna Austria.
21. Siekierski, M. Hybrid Polymer Electrolytes Based on Aluminum Oxygen Compound Distributed in Matrix: Electrochemical and Rheological Characterization. 218<sup>th</sup> ECS Meeting, Las Vegas, Nevada, October 10–15, 2010.

22. Siekierski, M. Composite Polymeric Electrolytes: Strategies Towards Application in Microbateries, 218<sup>th</sup> ECS Meeting, Las Vegas, Nevada, October 10–15, 2010.
23. Aernouts, T. Strategy and recent developments of organic photovoltaics at IMEC. Presented at: Organic Semiconductor Conference. 28–30 September 2009; London, UK.
24. Aernouts, T. Research strategies in a developing OPV market. Presented at: OPV Summit. 15–16 October 2009; Boston, MA, USA.
25. Giroto, C., Rand, B., Genoe, J. & Heremans, P. Exploring spray coating as a deposition technique for the fabrication of solution-processed solar cells. In: Solar Energy Materials and Solar Cells. Vol. 93: (4) pp.454–458.
26. Giroto, C. & Genoe, J. Sprühverfahren verbilligen die Fertigung Polymer-basierter organischer Solarzellen: Solarzellen von der Rolle. In: Markt&Technik. pp.19–33.
27. Hadipour, A., Genoe, J., Heremans, P., Campo, B., Aernouts, T. & Poortmans, J. Plastic solar cells. Presented at: PV Summit. 16-19 June 2009; Shanghai, China.
28. Giroto, C., Moia, D., Rand, B., Aernouts, T. & Heremans, P. Efficient polymer solar cells via an all-spray-coated deposition. In: 35th IEEE Photovoltaic Specialists Conference – PVSC. IEEE, 2010. pp.1068–1072; (20–25 June 2010; Honolulu, Hawaii (US).
29. Giroto, C., Moia, D., Rand, B. & Heremans, P. High-performance organic solar cells with spray coated hole injection and active layers. Advanced Functional Materials. Vol. 21: (01) 64–72; 2011. (Selected for inside cover picture)
30. Rönkä, K. FACESS project presentation, Printed Electronics and Photovoltaics 2011, Dusseldorf April 5<sup>th</sup>–6<sup>th</sup>, 2011.
31. Tuomikoski, M. Painetut aurinkokennot. Älyä kuluttaja tuotteisiin – Uudet ratkaisut ja mahdollisuudet, Vantaa, Finland 5.2.2009.
32. Siekierski, M. Characterization and Optimization of the Polymeric Electrolytes Based on Methulalumoxane and Polyethylene Glycols, 217<sup>th</sup> ECS Meeting, Vancouver, Canada, April 25–30, 2010.
33. Siekierski, M. Improvement of Polymeric Electrolytes Properties: Strategies of System Modification, 217<sup>th</sup> ECS Meeting, Vancouver, Canada, April 25–30, 2010.

Appendix 1: FACESS dissemination activities

34. Marczewski, M. Synthesis of additives based on functionalisation of methylalumoxane by organic compounds containing free hydroxyl group for application in battery electrols, XII International Symposium on Polymer Electrolytes ISPE-12, 29 August – 3 September 2010, Padova, Italy.
35. Siekierski, M. Novel hybrid polymer electrolytes based on multifunctional methylalumoxane and polyethylene glycols, XII International Symposium on Polymer Electrolytes ISPE-12, 29 August – 3 September 2010, Padova, Italy.
36. Siekierski, M. Improvement of polymeric electrolytes properties – strategies of system modification, XII International Symposium on Polymer Electrolytes ISPE-12, 29 August – 3 September 2010, Padova, Italy.



## Appendix 2. The FACESS presentations

- Organic and Large Area Electronics Clustering Meeting, 19<sup>th</sup> February 2008, Brussels, Belgium
- EU-South Korea Co-operation Forum, 16<sup>th</sup> – 17<sup>th</sup> June 2008, Seoul, South Korea
- 1<sup>st</sup> International Symposium on Flexible Organic Electronics, 9<sup>th</sup> – 11<sup>th</sup> July 2008, Halikidiki, Greece
- Organic and Large Area Electronics Clustering Meeting 13<sup>th</sup> January 2009, Brussels, Belgium
- OPV Summit 09, Boston, USA
- ISFOE09
- EMPC09
- LOPE-C, Frankfurt, Germany 2009
- Plastic Electronics Asia 2009. Taiwan
- Plastic Electronics 2010 in Dresden 27<sup>th</sup> – 29<sup>th</sup> October 2009
- Kraftwerk Batterie, Mainz/Germany, 1<sup>st</sup> – 2<sup>nd</sup> February 2010
- FlexTech, Phoenix/USA, 1<sup>st</sup> – 4<sup>th</sup> February 2010
- SSI 2010, Como/Italy, 23<sup>rd</sup> – 24<sup>th</sup> March 2010
- Printed Electronics 2010, Dresden/Germany, 12<sup>th</sup> – 14<sup>th</sup> April 2010
- Pira OPv Conference, Philadelphia/USA, 28<sup>th</sup> – 30<sup>th</sup> April 2010
- DTIP 2010, Sevilla/Spain, 5<sup>th</sup> – 7<sup>th</sup> May 2010
- TPE 2010, Rudolstadt/Germany, 18<sup>th</sup> – 20<sup>th</sup> May 2010
- LOPE-C 2010, Frankfurt/Germany, 1<sup>st</sup> – 2<sup>nd</sup> June 2010
- ISFOE-2010, Chalkidiki/Greece, 7<sup>th</sup> – 9<sup>th</sup> July 2010
- IPS-18, Seoul/Korea, 25<sup>th</sup> – 30<sup>th</sup> July 2010
- ISCST-2010, St. Paul/USA, 12<sup>th</sup> – 15<sup>th</sup> September 2010
- Batteries 2010, Cannes/France, 29<sup>th</sup> September – 1<sup>st</sup> October 2010
- Battery Power 2010, Texas/USA, 19<sup>th</sup> – 20<sup>th</sup> October 2010
- 4. AchenDresdner 2010, Dresden/Germany, 25<sup>th</sup> – 26<sup>th</sup> November 2010
- Printed Electronics and Photovoltaics in Dusseldorf 5 & 6 April, 2011.

**Exhibitions:**

- ICT4EE, Brussels, 19<sup>th</sup> – 20<sup>th</sup> March 2009
- ICT2010, Brussels, 27<sup>th</sup> – 29<sup>th</sup> September 2010
- Plastic Electronics 2010, Dresden, 19<sup>th</sup> – 21<sup>st</sup> October 2010
- Printed Electronics & Photovoltaics USA 2010, Santa Clara, 30<sup>th</sup> November 2010
- ICT4EE, Brussels, 23<sup>rd</sup> – 24<sup>th</sup> February 2010
- LOPE-C 2010, Frankfurt, 1<sup>st</sup> – 2<sup>nd</sup> June 2010
- Printed Electronics and Photovoltaics, Dusseldorf 5<sup>th</sup> – 6<sup>th</sup> April 2011.



Series title, number and  
report code of publication

VTT Research Notes 2602  
VTT-TIED-2602

Author(s) Kari Rönkä		
Title <b>Flexible autonomous cost efficient energy source and storage</b>		
Abstract <p>The key objective of Flexible Autonomous Cost Efficient Energy Source and Storage, FACESS project was to develop roll-to-roll printing process for organic solar cells (OSC) devices and thin film batteries (TFB). A control transistor circuitry on foil, which controls the operation voltage and charge of TFB's was also to be developed. The integration of OSC and TFB modules and control transistor circuitry into a flexible autonomous energy source and storage device was also one of the key objectives of the project. No material development was performed in the project, and thus only commercial materials were used. The technical target specification for OSC module was: 2.5% @ AM1.5, A~100 cm<sup>2</sup>, 250 mW and for the TFB module: Li-ion, 1-3 mAh/cm<sup>2</sup>, A~30 cm<sup>2</sup>.</p> <p>The FACESS project (FP7 – 2007-ICT-1-215271) funded by European Commission was executed between January 1<sup>st</sup> 2008 and April 31<sup>st</sup> 2011. The developments were carried out by the following consortium: VTT Technical Research Centre of Finland (coordinator), IMEC Interuniversity Micro-Electronics Centre (Belgium), CEA Commissariat à l'Energie Atomique (France), WUT Warsaw Technical University (Poland), Umicore S.A.(Belgium), Coatema Coating Machinery GmbH (Germany), Suntrica Oy (Finland) and Coatema Maschinenbau GmbH (Germany). In this final report the key achievements, problems faced, lessons learned, as well as the vision and exploitation towards the large-scale production of flexible OSCs and TFBs, have been presented.</p>		
ISBN 978-951-38-7791-0 (URL: <a href="http://www.vtt.fi/publications/index.jsp">http://www.vtt.fi/publications/index.jsp</a> )		
Series title and ISSN VTT Tiedotteita – Research Notes 1455-0865 (URL: <a href="http://www.vtt.fi/publications/index.jsp">http://www.vtt.fi/publications/index.jsp</a> )	Project number	
Date December 2011	Language English	Pages 65 p. + app. 4 p.
Name of project		Commissioned by
Keywords Organic solar cells, thin film batteries, roll-to-roll printing, Energy Source and Storage, chip-on-foil, flexible system integration		Publisher VTT Technical Research Centre of Finland P.O. Box 1000, FI-02044 VTT, Finland Phone internat. +358 20 722 4520 Fax +358 20 722 4374



## VTT TIEDOTTEITA – RESEARCH NOTES

- 2587 Markus Olin, Kari Rasilainen, Aku Itälä, Veli-Matti Pulkkanen, Michal Matuszewicz, Merja Tanhua-Tyrkkö, Arto Muurinen, Lasse Ahonen, Markku Kataja, Pekka Kekäläinen, Antti Niemistö, Mika Laitinen & Janne Martikainen. Bentoniittipuskurin kytketty käyttäytyminen. Puskuri-hankkeen tuloksia. 2011. 86 s.
- 2588 Häkkinen, Kai. Alihankintayhteistyön johtamisesta metalliteollisuudessa. 2011. 71 s.
- 2589 Pasi Ahonen. Constructing network security monitoring systems (MOVERTI Deliverable V9). 2011. 52 p.
- 2590 Maija Ruska & Lassi Similä. Electricity markets in Europe. Business environment for Smart Grids. 2011. 70 p.
- 2591 Markus Jähi. Vartiointipalvelujen arvonmuodostus asiakkaan näkökulmasta. 2011. 91 s. + liitt. 6 s.
- 2592 Jari M. Ahola, Jani Hovila, Eero Karhunen, Kalervo Nevala, Timo Schäfer & Tom Nevala. Moni-teknisen piensarjatuotteen digitaalinen tuoteprosessi. 2011. 121 s. + liitt. 37 s.
- 2593 Mika Nieminen, Ville Valovirta & Antti Pelkonen. Systemiset innovaatiot ja sosiotekninen muutos. Kirjallisuuskatsaus. 2011. 80 s.
- 2594 Katri Valkokari, Tapio Koivisto, Raimo Hyötyläinen, Maarit Heikkinen, Magnus Simons, Maaria Nuutinen, Tiina Apilo & Juha Oksanen. Management of future innovative firms and networks. Espoo 2011. 179 p.
- 2595 Martti Flyktman, Janne Kärki, Markus Hurskainen, Satu Helynen & Kai Sipilä. Kivihiilen korvaaminen biomassoilla yhteistuotannon pölypolttokattiloissa. 2011. 65 s. + liitt. 33 s.
- 2596 Aki-Petteri Leinonen. Identity management for web-enabled smart card platform. 2011. 64 p. + app. 2 p.
- 2597 Markku Kiviniemi, Kristiina Sulankivi, Kalle Kähkönen, Tarja Mäkelä & Maija-Leena Merivirta. BIM-based Safety Management and Communication for Building Construction. 2011. 123 p.
- 2598 Heidi Korhonen, Tiina Valjakka & Tiina Apiolo. Asiakasymmärrys teollisuuden palveluliiketoiminnassa. Tavoitteena ostava asiakas. 2011. 111 s.
- 2599 Riikka Juvonen, Vertti Virkajärvi, Outi Priha & Arja Laitila. Microbiological spoilage and safety risks in non-beer beverages produced in a brewery environment. 2011. 107 p. + app. 3 p.
- 2600 Juha Luoma. Keski-Euroopan olosuhteisiin suunniteltujen kitkarenkaiden yleisyys Suomessa. 2011. 16 s..
- 2601 Timo Malm, Matti Vuori, Jari Rauhamäki, Timo Vepsäläinen, Johannes Koskinen, Jari Seppälä, Heikki Virtanen, Marita Hietikko & Mika Katara. Safety-critical software in machinery applications. 2011. 111 p. + app. 10 p.
- 2602 Kari Rönkä. FACESS, Flexible autonomous cost efficient energy source and storage. Final report. 2011. 64 p. + app. 4 p.