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ANALYSIS

Solar cells with one-day energy payback for the factories of the future

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Scalability is a requirement before any new energy source can be expected to house a possible solution to the challenge that mankind's increasing energy demand presents. No renewable energy source is as abundant as the Sun and yet efficient and low-cost conversion of solar energy still has not been developed. We approach the challenge by firstly taking a technology that efficiently addresses the need for daily production of 1 GW_p on a global level, which does not employ elements with critically low abundance and has a low thermal budget. We then applied life cycle assessment methodologies to direct research and developed such technology in the form of a polymer solar cell that presents a significant improvement in energy payback time (EPBT) and found that very short energy payback times on the order of one day are possible, thus potentially presenting a solution to the current energy gap of >14 TW by year 2050.

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1 Introduction

The projected increase in energy demand between now (year 2011) and the year 2050 is on the order of 1 GW a day. This "one-gigawatt-a-day" challenge is equivalent to the building of

Broader context

Critical for the success of renewable energy technologes is the time it takes for a given technology to earn back the energy invested in its making and constitution. This is known as the energy payback time (EPBT) and for the most successful and mature renewable energy sources such as wind and hydro power the EPBT is in the range of 3–6 months. For the solar technologies the EPBT is significantly longer and even for mature photovoltaic (PV) technologies such as crystalline silicon the EPBT is in the range of 1–2 years and unlikely to decrease significantly due to very high processing temperatures and materials usage. The polymer solar cell is a very recent PV technology that already rivals existing PV technologies in terms of EPBT even if they currently present a shorter operational lifetime. Due to the very low processing temperature and a thin outline we demonstrate that it is possible to achieve exceptionally low EPBT values in the range of one day pending the realizations of the projected performance for the technology and through the use of solar thermal and solar electric energy for processing. We demonstrate that the technology in its current form outperforms existing PV.

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an average nuclear power plant a day for the next ~40 years (globally). There is currently no available technology that can address this while the energy sources are fortunately available. Several of the known renewable energy sources (wind, biofuel, hydro, solar) are truly renewable in the sense that their source is ultimately the sun. Except for solar energies (thermal and electric) all those sources are derived from the sun through secondary processes and thus the technical potential for extraction is inherently lower and therefore direct usage of solar energy is of great interest. The projections on world energy demand to 2050 will require an increase in energy production capacity as described above, but they also imply a growing uncertainty concerning the energy supply. World electricity demand in particular, is expected to grow more sharply than any other final form of energy. Since "renewables" are low-carbon technologies, they can tackle climate change targets and play a central role in reducing this uncertainty.

Today renewable energy sources account for almost 20% of the total electricity production. That share has changed only marginally since 2000. In 2008, 85% of the electricity from renewable sources came from hydropower, and the share of other renewable energy sources combined is around 2%.[1] Hydropower as an electricity source has been leading over the past century to electricity production, but wind and photovoltaics (PV) have been steeply increasing in the last decade. Solar cells provide just a tiny slice of the world's electricity, but thanks to technology improvements and government policies, the total PV capacity has multiplied by a factor of 27 at a rate of 40% since 2000 and now reaches a total installed capacity of 39.5 GW. Solar thermal energy conversion is presently accessible in high yield but is in efficient use in the form of heat and is not as easily distributed as solar electric energy, which thus far has been quite limited in use. There are solar electric energy conversion systems in existence that convert above 40% of the incoming solar energy into electricity and this clearly demonstrates the potential to directly convert sunlight into electrical energy with great efficiency. In the next 50 years, according to a projection by the International Energy Agency, solar generators are likely to produce the majority of the world's power by 2060.[2]

While the conversion efficiency is often used as the metric to evaluate the performance and potential usefulness of a technology or system, it does not take into account the scale of the problem at hand and the potential unavailability of elements or components, nor does it take into account if it is possible to create the required energy producing unit in a given amount of time and whether the energy producing unit is capable of delivering back the energy spent creating it in a reasonable time. To address this life cycle assessments of the different technologies is a very useful tool and ideally reveals that a given technology quickly pays back the energy spent making it and operates for significantly longer. Table 1 lists a selection of renewable energy technologies and their energy returns or energy payback times (EPBT).

When examining the above renewable energy sources, the best performers as mature technologies have energy payback times of hundreds of days to years; however the current manufacturing speed is not easily envisaged as even approaching a daily production capacity of 1 GW_p a day.

There is a general agreement that the photovoltaic (PV) technology is an ideal way to harvest solar energy as it is silent, has no moving parts and in principle requires little or no maintenance once installed. All existing PV technologies however suffer from several drawbacks: most notably their relatively high cost, large thermal budgets and relatively slow manufacture. Even if some of the PV technologies do provide competitive electricity production costs they still present a slow learning curve and energy payback times on the order of years. There are currently only two PV technologies that do not employ rare elements that technically prevents them from realistically addressing the 1-gigawatt-a-day challenge. Those are the silicon based PV and the polymer or organic solar cells.

The polymer solar cell in particular has the advantage over all PV technologies that the possible manufacturing speed is very high and the thermal budget is low as no high temperatures are needed. The polymer solar cell also has drawbacks and it can by no means be viewed as a mature technology. Especially, the operational lifetimes and power conversion efficiencies have been viewed by many as prohibitively short and low. The polymer solar cell has however consistently improved in both aspects, and now presents operational lifetimes of several years and laboratory efficiencies approaching 10%. The EC have in a report (A vision for photovoltaic technology – Report by the Photovoltaic Technology Research Advisory Council PV-TRAC, European Commission, 2005) projected operational lifetimes for more than 10 years and power conversion efficiencies of more than 10% by year 2020 (the projected attainable values are in the range of 12–17%). The most distinguishing feature of the polymer solar cell is

Table 1 Listing of different renewable energy technologies and their EPBT (in years), measured in MJ kWh_{el}^{-1}

Energy source/Technology		EPBT (years)	Source
Wind	On land	0.26	3
	Offshore	0.39	
PV technologies	Silicon mono- and polycrystalline	1.65–4.12	4,5
	Amorphous silicon	1.13	6
	GaAs PV	2.36–5	7,8
	GaInP/GaAs	2.14–4.6	
	CdTe	0.73–1.61	8,7
	CIS	2.02–2.26	5,7
	OPV	0.2–4	9–11
Hydropower		0.5	12,13
Geothermal		0.54	13
Biomass	Gasification	Below 5 years	14
	Biomass combustion	5–10	15

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the possibility to print or coat the device directly onto a thin flexible carrier substrate using roll-to-roll coating and printing methods which enable simple handling and fast processing. While the use of flexible substrates is what enables fast and low cost processing, flexibility is possibly not needed in the final product.

Justification of polymer solar cell usage in the available form (outside the laboratory) has been questioned due to the relatively low performance characteristics. It is however not uncommon for PV technologies to exhibit a lower performance in their manufactured form as compared to the laboratory records (*i.e.* mass produced silicon PV presents a power conversion efficiency of 65% compared to the laboratory record). Polymer solar cells prepared in large numbers by roll-to-roll methods thus present power conversion efficiencies in the range of 2–3% on the active area (on average). To overcome the criticism several challenging developments have been highlighted as needed before OPV can become a success and the most focus has been on improving the power conversion efficiency but also the operational lifetime. Especially the latter has recently been given much attention due to the fact that a mediocre solar cell with a long operational lifetime easily outperforms a high performing (with respect to efficiency) but short lived solar cell over time when considering the total energy that the solar cell can potentially deliver during its service life.

1.1 Organic solar cells

A wide range of materials is used in PV, from the traditional multicrystalline silicon wafers to thin-film silicon solar cells and devices composed of plastic or organic semiconductors. Both laboratory and commercial efficiencies for all kinds of solar cells have risen steadily in recent years, indicating that research efforts could further enhance the performance of solar cells. Unlike the conventional PV technologies, OPV has the lowest energy embedded in the cells, as well as the poorest performance. However, whereas the drop in efficiency when going from silicon to OPV is a factor of 10–20 (on the module level) the drop in equivalent primary energy (EPE) drops by a factor of >150.[4]

Typical OPV structures comprise a series of layers, one on top of another. As shown in the image (Fig. 1), first a transparent electrode acting as anode or cathode is set down on a transparent base. Then, a selective conductor layer can be deposited on top, followed by the active layer, where the light harvesting and photovoltaic effect take place. Another selective conductor is usually needed for guiding the holes/electrons to the opposite electrode (the last deposited layer).

There exist two basic configurations of OPV cells. The normal device geometry comprises four layers on the substrate (Fig. 1 left, without ET layer) whereas the inverted device type has five layers to enable selective transport of electrons to *i.e.* ITO and holes to the metal electrode (Fig. 1 both images). The entry of the light to the active layer depends on the position of the transparent electrode (front or back).

1.1.1 Problems with abundance of materials. There are requirements for the materials typically employed such that the possible low thermal budget is explored; better barrier materials, thinner adhesives and the development of new semitransparent electrode materials that can be printed or coated, such that vacuum steps are avoided. During the development of OPV a single transparent electrode material in the form of indium-tin-oxide (ITO) has been employed and it is only recently that significant efforts have been directed towards indium-free electrodes. The current solutions are not necessarily better or lower in cost than ITO but it can be expected that an efficient solution will be found in the near term and several approaches have already been identified.

The need for a transparent and highly conductive layer as an electrode in printed electronics was created in the mid-1970s. ITO, having both characteristics when deposited by vacuum sputtering tools first on glass and now on flexible substrates, was commercially developed and grew rapidly. But indium is a rare and scarce element. In order to remedy the problem of indium scarcity (indium is the main component of ITO) the routes towards ITO-free solar cells should include the use of a solution processable transparent polymer electrode instead of vacuum processed ITO.

A number of alternatives have been recently employed in small laboratory devices, including: carbon nanotubes,[16–18] graphene,[19–21] silver nanowires (AgNW),[22] highly conducting polymers and metal grid electrodes[23–28] as the firstly processed electrode being transparent or semi-transparent. A second approach involves a non-transparent layer as the firstly processed layer. An example of this is the use of evaporated aluminium–chromium[27,28] as the firstly processed non-transparent electrode, the semitransparent electrode having been processed lastly and being comprised of a doped polythiophene derivative (PEDOT:PSS) in combination with a metallic current collecting grid. Despite the advantage of high conductivity, the non-transparent nature of current collecting grids incurs losses in active area. None of the transparent electrode materials (alternative to ITO) available today provide both high transparency and high electrical conductivity simultaneously. More

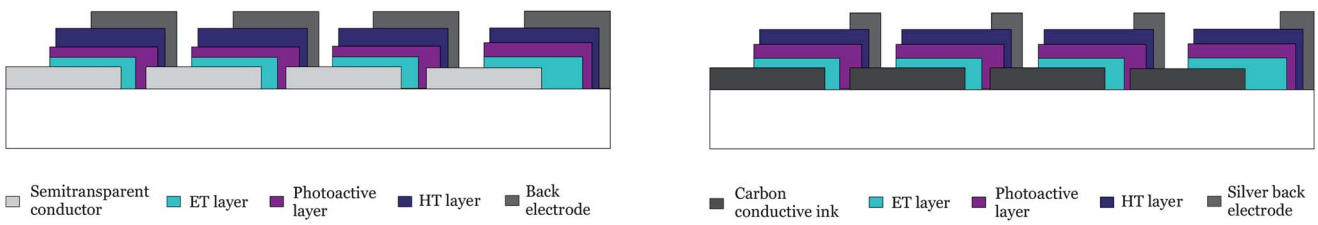

Fig. 1 Typical 5-layer OPV structure showing the front trough substrate (left image) and back illumination (right image). Note that in the latter case, the electrode size is reduced in order for the light to enter through the back. ET stands for electron transport, and HT for hole transport.

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current-collector geometries (such as hexagonal, parallel line or interdigitated) on the scale of <1 mm should be studied.

Most of the rear contacts employed today within the field of photovoltaics are based on silver while there have been efforts in replacing silver with aluminium or copper.

1.1.2 Roll-to-roll production. The manufacture of a polymer solar cell that comprises a multilayer structure is expected to be massively scaled-up by high-throughput or roll-to roll (R2R) methods. This way of production involves a series of operations for every layer: unwinding, optional surface treatment, coating/printing, annealing/drying and rewinding. In the ultimate stage the solar cells are laminated/encapsulated and tested. In Fig. 2 some of the processing steps are shown for a typical polymer solar cell where stripes, each representing a single solar cell, are coated and the back electrode is later printed on top of the device to serially connect the individual devices (stripes) into complete modules.

1.1.3 Market status. The market share for OPV, estimated to around $2 million in 2011, is projected to sky-rocket to $56 million for the next decade and reach $387 million in revenues by 2016.[29] So far, commercialization of OPV has been addressed by a few companies with a view to integration into products,[30–32] although grid-tied applications are foreseeable in a medium term, due to the fact that solar electricity costs are forecast to decrease in the near future. The cost of solar electricity is typically compared to electricity produced by traditional sources with a levelized cost of energy (LCOE) calculation. Recent estimates of electricity cost generated by OPV have shown this to be possible.[33] It is surprisingthat for these organic solar cells under an average Mediterranean solar irradiance of 1700 kWh m^{-2} per year the LCOE ranges between 0.19 and 0.50 € kWh^{-1},[33–35] while a mature 19.7% efficiency of 20 MW_p plant (depending on geography) yields LCOE values of around 0.05 to 0.07 € kWh^{-1}.

Silicon technologies could also face the "1 GW_p a day" challenge within the next decade but this would require a vast investment both in terms of energy and cost. The upscaling needed before one is able to produce 365 GW_p a year would require an energy input of around 1.37×10^{10} GJ and an investment of 641 billion euros. In comparison, 10 times less energy and 3 times lower capital investment would be needed with a similar OPV installation (Table 2).

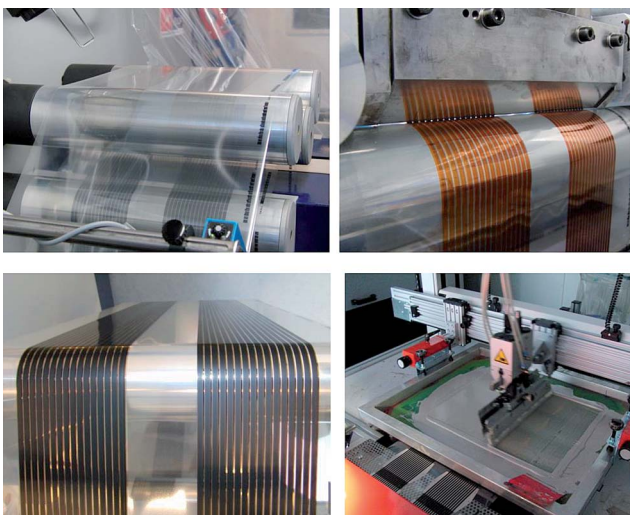

Fig. 2 Photographs of ITO-free polymer solar cells prepared entirely using roll-to-roll processing from solution. The width of the web is 305 mm and two modules comprising 16 serially connected cells are coated and printed simultaneously. The coated semi-transparent front electrode is cured (top left). The active layer is slot-die-coated on top of the transparent front electrode coated with zinc oxide, seen as the orange brown colour of the wet film (top right). The semi-transparent hole transport layer is coated and dried on top of the stack (bottom left). Finally, a silver or carbon based back electrode is screen printed to complete the modules (bottom right, here shown as silver).

1.2 The life cycle analysis as a research tool

Life cycle assessments/analyses (LCA) are an excellent tool to point out weaknesses and strengths of a process providing useful feedback for development. A LCA characterizes and quantifies the total environmental burdens of a product or system, from raw materials extraction to end-of-life management. Several LCAs of energy technologies have been carried out in the past few years as shown in Table 1, however with regard to the photovoltaic field, these studies revealed that PV technologies always consume less energy during their manufacturing than the energy generated over their entire life cycle. The EPBT has been thoroughly investigated for all PV technologies already on the market, and ranges between 4.12 and 0.73 years.[6,5,36] For mono-Si EPBTs from 4.12 to 2.68 years can be found, depending on power conversion efficiencies (11.8 or 14%), for commercial modules; polycrystalline-Si has an EPBT of 2 years for 13% efficiency;[36,37] amorphous-Si, 1.13 years for 7% efficiency;[38] CIS, 2.26–2.2 years for 8.9–11% efficiency,[5,7] and CdTe, 1.61–0.73 years for 11–13%;[7] and OPV from roll to roll processing, 0.79 to 2.02 years for 5 to 2% efficiency.[9,11] Particularly, polymer solar cells have been analysed and have proved that the embodied energy in the cells is notably lower, accounting for around 50–80% less than in silicon and thin film-based PV technologies. One of the first LCA studies on polymer solar cell preparation on a laboratory scale[10] showed that two-thirds of the energy embedded was devoted to maintaining an oxygen free nitrogen atmosphere in a glove box.

When analysing polymer solar cells prepared according to a well described process (known as ProcessOne[39]) and by using as input all the processing parameters it was found that energy payback times ranging from 1.35 to 2.02 years were possible (corresponding to 3 and 2% efficiency, respectively).[11]

Before this first thorough life cycle analysis of the roll-to-roll process for manufacturing OPV cells,[11] no precise information was available of the parameters likely to affect the future scale up of manufacturing. This first study concluded that 90% of the total energy was embedded in the transparent conductor.

The large requirements for both energy and monetary cost in the ITO sputtering were clearly identified as dominating materials cost,[35] and as an imbalance in the inventory (see ProcessOne in Fig. 11). The assumptions during computing of the embedded energy in the ITO layer were made by extrapolating the sputtering requirements on a laboratory scale—on the order of dozens of square centimetres—to a medium-scale production, in

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Table 2 Investment cost comparison for current silicon PV technologies and OPV manufactured at Risø DTU, with a current 1% and a forecasted 10% efficiency

COSTS	Unit	Silicon PV	OPV (10%)	OPV (1%)
Monetary investment				
Annual capacity production	GW_p	365	365	365
Cost per W_p produced	€	1.76	0.63	6.32
Cost 1 GW_p/day plant	Billion €	641.22	230.53	2305.34
Energy investment				
Equivalent primary energy	MJ $W^{-1}{}_p$	37.43	0.4511	4.51
EPE for production 1 GW_p/day	GJ	1.37E + 10	1.65E + 08	1.65E + 09

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the range of several square meters. And the result was around 170–235 MJ_{EPE} m^{-2} for the typical ITO thicknesses (90 to 180 nm). New sputtering tools using R2R processes are being launched on the market, and while they promise lower embedded energy in the range of 30–50 MJ_{EPE} m^{-2} this should be verified by actual manufacture while accounting for all energy inputs.

Nevertheless, this process would still represent a large share of the embedded energy. Attempts to solve this by replacing indium with abundant and less-demanding energy electrode materials have been investigated. The use of aluminium-chromium composite electrodes was analysed by the LCA methodology,[40] showing an even poorer balance in agreement with the intensive energy fabrication of the electrode. The most pertinent question that came up was what the developments should be, and we clearly showed that processes employing both vacuum and large amounts of inert gasses should be avoided.

LCA studies are founded in a standardized approach under the ISO 14040 section,[41,42] that were used to assess the overall resource consumption of a dozen of generically different polymer solar routes that have been tested at Risø DTU. The most successful route was identified by deliberate minimisation and balancing of the equivalent primary energy.

The life cycle of a process, product or service is usually divided into procurement, manufacture, use and disposal. According to the information available and following ISO standard indications, it is a matter of every LCA to draw the boundaries of the process/product; *i.e.* which processes are included into the analysis, as presented in Fig. 3. In this case, the analysis incorporates direct energy to the OPV manufacturing process and direct energy to produce materials used in the manufacturing—primary and ancillary—but does not include the energy used for manufacturing the roll-to-roll machinery. Also, in order to make fair comparisons with other OPV routes, decommissioning and/or recycling phases were included in the assumptions.

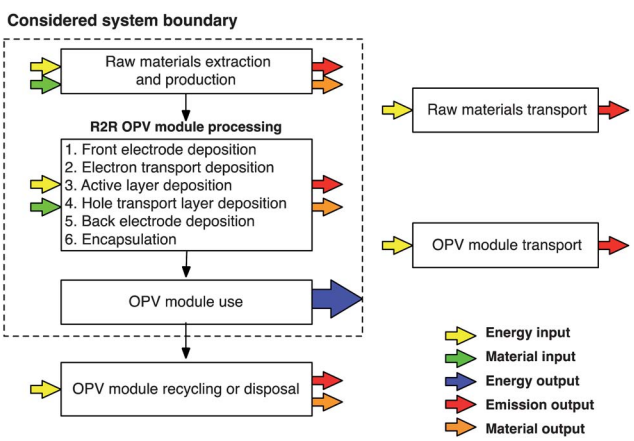

Fig. 3 Illustration of the system boundary employed in this LCA (everything inside the dashed line). All stages of the modules produced at Risø DTU are depicted as well as energy/materials inputs/outputs.

Finally, the energy requirements for transport of materials to the manufacturing plant and for transporting the finished solar cell product to the place of use, have been neglected, thus enabling direct comparison with other OPV LCAs that did not include energy requirements for transport. We view it as a safe assumption that it is of low significance per module. Typical values, as given in databases (Ecoinvent 2.1[43]), range from 0.23 MJ kg^{-1}, for road transport to 0.47 MJ kg^{-1} when it is rail freight. One module weighs much less than 1 kg m^{-2} and the required chemicals including bottles *etc.* also weigh less than 1 kg, so the total contribution that transport would account for is maximally on the order of 1 MJ m^{-2}.

1.2.1 Life cycle inventories. Life cycle inventories (LCI) are the core of any LCA. LCI stands for a list where inputs and outputs for the manufacturing of a product are gathered; *i.e.* all the materials and energy supplies and emissions throughout its life cycle.

Full characterisation of the processes at Risø DTU and all the assumptions are herein detailed. The functional unit for the OPV modules is commonly 1 m^2. Note that we use 1 m^2 of *processed* area (290 mm width) from where 45% is the photoactive area. This is a critical issue and has been discussed earlier.[40]

By compilation of material and energy inventories, the energy input for processing materials and the energy input during module manufacturing can be estimated. This enables a careful determination of the overall embedded energy in the modules, widely called cumulative energy demand (CED). Data related to the OPV processing were acquired *in situ* from materials consumption and equipment specifications. Energy related to these materials and upstream processes was obtained from databases such as Ecoinvent 2.1,[43] through SimaPro (a commercial software widely used for LCA studies) and from the literature. Both thermal and electrical budgets were converted to equivalent primary energy (EPE) at a later stage. The conversion efficiency depends on the technology mix of the electricity supply system and may vary considerably between different countries and also with time. An average of 35% has been considered.[6] In the case of thermal energy consumption this was given separately

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by using the European average factor of 85% to obtain the primary energy equivalent.[6]

Data used in the assessment, are experimental as already stated; they are not an estimate. Therefore, their quality is considered good. However, a weakness of this approach is that it is based on some assumptions, as there would otherwise be too many uncertainties to allow for a plausible assessment of the product's environmental 'goodness'. Regarding the OPV research field, on a semi-industrial scale, there is still only information available from one location in the world (Risø DTU). It should thus be recommended that the PV community starts to share information more extensively to balance the view.

Information on the uncertainty of the model outcomes provides useful information for assessing the reliability of LCA-based decisions and for guiding future research towards more accurate life cycle assessments. In order to accommodate this, we have tried to give a qualitative indication of the energy requirements we have estimated, by introducing the following qualifications: ++ very good, + good, 0 fair, − low, −−very low; as will be shown bellow.

LCA generally encompasses an inventory with all cumulative emissions; from the electricity mix and those released to raw materials production/extraction. The emissions inventory presented here is only focused on emissions of CO_2 equivalents related to the energy consumption during the manufacture. For the electricity mix of Denmark these emissions amount to 493.80 g-eqCO_2 kW_{el}^{-1} in 2010.[44]

1.2.2 Life cycle indicators: *EPBT*, *GPBT and environmental categories*. The time required to pay back the energy invested is a key parameter for evaluating the sustainability of an energy-production technology, and is of particular interest in renewable energy technologies like PV systems. The energy payback time (EPBT) is the time required for the solar PV system to generate the equivalent amount of energy consumed in the construction and decommissioning phases.

For its calculation, the required assumptions are a constant OPV module efficiency in the range of 1 to 15%—in agreement with attained performances of up to 3% at Risø DTU for flexible roll-to-roll processed solar cell modules—an insolation level of 1700kWh m^{-2} per year, typical of southern European countries, and a service lifetime of 15 years. This lifetime can be low as compared to other PV technologies, which easily reach service lifetimes of 20–30 years, but is consistent with the aims of the European Photovoltaic Technology Platform for organic photovoltaics by 2013.[45]

Through life cycle impact assessment (LCIA) the emissions, consumed resources and other items associated with a system are translated into environmental impacts. There are different methods that can be used to perform a LCIA. Two main approaches are used to classify and characterize environmental impacts: the problem-oriented approach (mid-point) and the damage-oriented approach (end-point). The former can be classified into environmental themes (*e.g.* greenhouse effect, eutrophication or acidification) whereas the latter also starts by classifying how a system flows into various environmental themes, and model each environmental theme's damage to human health, ecosystem health or damage to resources.

EcoIndicator 99 (end-point)[46] is the chosen damage-oriented method that includes the central elements of the LCIA methodology: characterisation, normalisation and weighting, which provide information on the most environmentally damaging materials. For assessing the environmental impacts, SimaPro, with the methods implemented, allowed us to identify the materials which have a detrimental effect in different environmental categories such as Carcinogens, Fossil Fuel depletion, Ecotoxicity and Climate Change.

2 Experimental

2.1 The new polymer solar cell routes

2.1.1 Materials. The substrate was a 45 μm thick packaging barrier (from Amcor) with a UV filter. For the module encapsulation the barrier foil was pre-laminated with a pressure sensitive adhesive (467MPF, 3M). The active layer was a mixture of poly(3-hexylthiophene) (P3HT) (Sepiolid P200, BASF) and phenyl-C_{61}-butyric acid methylester (PCBM) (99%, Solenne B. V.) dissolved in chlorobenzene. The P3HT : PCBM weight ratio was 30 : 30 mg ml^{-1}. Poly(3,4-ethylenedioxythiophene) : poly (styrenesulfonate) PEDOT : PSS was purchased from Agfa (Orgacon EL-P 5010) and diluted with isopropanol in a ratio of 10 : 5 by weight. The ZnO nanoparticle based solution has already been described.[47] The aqueous zinc oxide solution was prepared as described earlier[48] and was comprised of Zn $(OAc)_2 \cdot 2H_2O$ (100 mg mL^{-1}), $Al(OH)(OAc)_2$ (2 mg mL^{-1}) and FS-100 (2 mg ml^{-1}) in water. Three silver inks were used: commercial silver ink, silver ink formulated at Risø, and PV 410 silver ink (Dupont®). The ink for semitransparent silver electrodes was prepared from mixing in a 1 : 5 ratio, 18% Ag content ink produced by a method similar to that reported by Hwang *et al.*[49] and butanol. For silver ink nanoparticles (100 ± 15 nm) were dispersed in a 1 : 1 mixture of triethyleneglycolmonomethylether and *ortho*-xylene as described earlier.[23] The final concentration of silver nanoparticles was 25% (w/v).

2.1.2 Module design. The structure of the modules is illustrated in Fig. 4, comprising barrier/electrode/ZnO/P3HT : PCBM/PEDOT : PSS/electrode/barrier. Each module had 16 serially interconnected cells, where the serial connection was achieved upon screen-printing of the metal stripe back-electrode. The narrow silver stripes only served as a serial connection, whereby the thick PEDOT : PSS served as current collector.

2.1.3 Manufacturing stages. A number of consecutive steps are involved in the production of OPV modules, as shown in Table 3. The semi-transparent front electrode is deposited by slot-die coating under ambient conditions on an inline R2R printing and coating machine from Grafisk Maskinfabrik A/S (Fig. 5); as well as active ink, ETL and HTL layers. This coating machine also includes an in-line rotary screen printer (from Stork) for graphite or silver electrode printing. The temperature of the hot air convection ovens was set to 140 °C. The silver back-electrode was screen printed on a flat-bed R2R screen printer from Alraun Technik and the encapsulation was performed using a R2R laminator from Grafisk Maskinfabrik A/S.

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Process Name	Layer 1	2	3	4	5	6
A	SD PEDOT:PSS Agfa	SD ZnO np			RSP C GRAPHITE F	
B		SD ZnO np			SP Ag PV 410 F	
C	SD Ag in nBuOH				SP C GRAPHITE F	
D					SP Ag PV 410 F	
E	SD Ag nanoparticles		SD ACTIVE LAYER	SD PEDOT:PSS	SD Ag Risø's I	
F		SD ZnO			RSP C GRAPHITE I	
G	RSP GRAPHITE				RSP C GRAPHITE I	
H					SD Ag Risø's I	R2R LAMINATION
I	SD Graphene				SP C GRAPHITE F	
J					SP Ag Risø's F	
K	ITO	SD ZnO np	SD ACTIVE LAYER aq	SD PEDOT:PSS aq	SD Ag Risø's F	
One	ITO	SD ZnO np	SD ACTIVE LAYER	SD PEDOT:PSS	SP Ag PV 410 F	
Two	SD Ag nanoparticles	SD ZnO np	SD ACTIVE LAYER	SD PEDOT:PSS	SP Ag PV 410 F	

Fig. 4 Layers of the manufacturing routes for producing ITO-free solar cells and the reference route ProcessOne. SD stands for Slot die coating, RSP for Rotary screen printing and SP for screen printing.

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Table 3 Summary of the manufacturing process stages for the new routes from Process A to J shown in Fig. 4

Stages of the manufacturing routes	Step	Operation
Electrode deposition	1.1	Slot die coating/Rotary screen Printing
	1.2	Drying
ETL coating	2.1	Ink preparation
	2.2	Slot die coating
	2.3	Drying
Active layer deposition	3.1	Ink preparation
	3.2	Slot die coating
	3.3	Drying
HTL deposition	4.1	Ink preparation
	4.2	Slot die coating
	4.3	Drying
Electrode deposition	5.1	Screen printing/Slot die coating/Rotary screen printing
	5.2	Drying
Encapsulation	6.1	R2R lamination

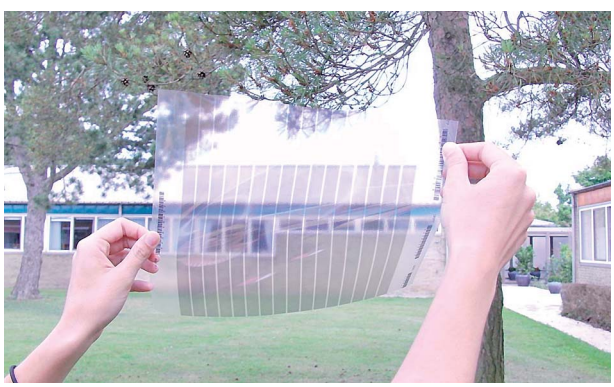

Fig. 5 Photograph showing a view through indium-free thin silver based semitransparent front electrode prepared by slot-die coating, using only low temperatures and very little material. This electrode type was explored in this work and presents a sheet resistance < 10 Ω square^{-1} and an optical transmission of > 50% in the entire visible range of wavelengths.

2.2 Processing and material inventory

Conditions for all the different coating and/or printing processes, and material inventories involved in these steps are detailed in Table 4, Table 5, Table 6 and Table 7. Further details on the equipment were already reported in ref. 30 and 50.

3 Results

3.1 Energy embedded in materials

Using the material inventory compiled in Table 4, Table 5, Table 6 and Table 7, the energy needed for manufacturing the materials was computed for Processes A to K, and for Process Two.

When the CED for a specific material was not available in the Ecoinvent database, it was necessary to calculate the energy content of a raw material or ink from scratch; *i.e.* from its basic components. Silver for the transparent electrodes is a non-particle conductive ink containing in its formula 18% silver.

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Table 4 Summary of the slot-die coating process, including drying conditions and material inventory for processing several layers of 1 m^2 OPV modules

Conditions	
Speed	0.02–2.5 m min^{-1}
Web tension	80–90N
Drying temperature	140 °C
Working corona treater power	1500 W
Working slot die power	1500 W
Working oven power	12000 W
Equipment	
Description	R2R printing and coating machine from Grafisk Maskinfabrik A/S comprising unwinder, corona treater, edge guide, Rotary screen printer, flexo unit, coating roller, oven, cooling roller and winding station.
Maximum corona treater power	1500 W
Maximum slot die coater power	1500 W
Maximum oven power	12000 W
Material inventory	
Front electrode layer	
PEDOT:PSS	39.34 g
Silver nanoparticle ink full	3.68 g
Silver ink (18% Ag)	0.26 g
Butanol	1.29 g
Graphene ink	1.97 ml
Active layer	
P3HT	0.08 g
PCBM	0.07 g
Chlorobenzene	6.19 g
ELT layer	
ZnO $(OAc)_2$	3.71 g
KOH	1.86 g
MeOH	12.24 g
Acetone	24.50 g
MEA	0.37 g
$ZnO(OAc)_2$	0.30 g
Water	2.95 g
HTL layer	
PEDOT:PSS	26.23 g
Isopropanol	38.89 g
Back electrode layer	
Silver for interconnections	0.63 g

It has been estimated to require energy for its production of as much as 275 MJ kg^{-1} and as low as 126 MJ kg^{-1}, thus resulting in 200.5 MJ kg^{-1} on average. This is very close to estimations done for PV 410 silver ink (from Dupont®) in previous studies.[11] The silver ink was then diluted in butanol 1 : 5 (v/v); the energy embedded in butanol is 81.02 MJ kg^{-1} according to the Ecoinvent database. Printable graphite conductive ink was estimated to embed 174.82 MJ kg^{-1}. Since the solid content in its formula can vary from 34 to 37%, an average of 36.5% was estimated (as shown in Table 8). With the aim to prove whether the solid content of the ink can affect the results, a sensitivity study was conducted, by varying this parameter from 0 to 100%, and no significant influence was found. Even if the embodied energy would increase by 200%, the variation of EPBT would remain below 3% of its initial value.

The graphene ink was prepared according to a method described by Hummers and Offerman[51] from natural graphite, and found to range from 200 to 400 MJ kg^{-1}. With regard to the ETL, zinc oxide ink, and HTL, PEDOT:PSS, had already been

Table 5 Summary of the flat bed screen printing process, including drying conditions and material inventory for printing the electrodes of 1 m^2 OPV modules

Conditions	
Speed	1 m min^{-1}
Residence time in oven	72 s
Drying temperature	140 °C
Working screen printer power	1500 W
Equipment	
Description	Alraun printer comprising unwinder, metering wheel, positioning camera, vacuum table, screen printer (AT701), hot air oven, transport rollers, dancing tensioning roller and rewinder.
Maximum screen printer power	38000 W
Material inventory	
Back electrode layer	
Silver ink PV410 full	19.67 g
Graphite full	19.67 g

Table 6 Summary of rotary screen printing process, including drying conditions and material inventory for printing the electrodes of 1 m^2 OPV modules

Conditions	
Speed	3 m min^{-1}
Drying temperature	140 °C
Working RS printer power	200 W
Working oven power	12000 W
Equipment	
Description	R2R printing and coating machine from Grafisk Maskinfabrik A/S comprising unwinder, corona treater, edge guide, Rotary screen printer, flexo unit, coating roller, oven, cooling roller and winding station.
Maximum RS printer power	1500 W
Maximum oven power	21000 W
Material inventory	
Front and back electrode layer	
Graphite full	4.59 g
Graphite interconnections	0.79 g

Table 7 Summary of encapsulation process, including drying conditions and material inventory for 1 m^2 OPV modules

Conditions	
Speed	120 m h^{-1}
Working laminator power	1500 W
Equipment	
Description	Laminator comprising unwinder, edge guide and cutting table, laminator, laminate unwinder, longitudinal cutting knifes and rewinder.
Maximum power	1500 W
Material inventory	
Adhesive (3M 467 MPF)	20.24 g
PET encapsulation	61.65 g

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Table 8 Detailed material inventory and CED for raw materials production in MJ_{EPE} units for Process H, with notation for certainty of data: ++ very good, + good, 0 fair,− low, −−very low

	Embedded energy (MJ_{EPE}) Per kg m^{-2}	Units Per FU	Embedded energy (MJ_{EPE}) Per FU	Certainty in energy data
PET substrate	80.75	0.0616500	4.98	++
Electrode ink (graphite)	174.82	0.0045902	0.80	0
$Zn(OAc)_2$	42.07	0.0002951	0.01	++
P3HT	1809.52	0.0000836	0.15	+
PCBM	11061.31	0.0000669	0.74	+
Chlorobenzene	61.58	0.0061869	0.38	++
Isopropanol	62.74	0.0388852	2.44	++
PEDOT:PSS	159.41	0.0262295	4.18	+
Back electrode (silver)	309.22	0.0006330	0.20	+
Adhesive	200.00	0.0202400	4.05	+
PET encapsulation	80.75	0.0616500	4.98	++
SUBTOTAL			**22.91**	

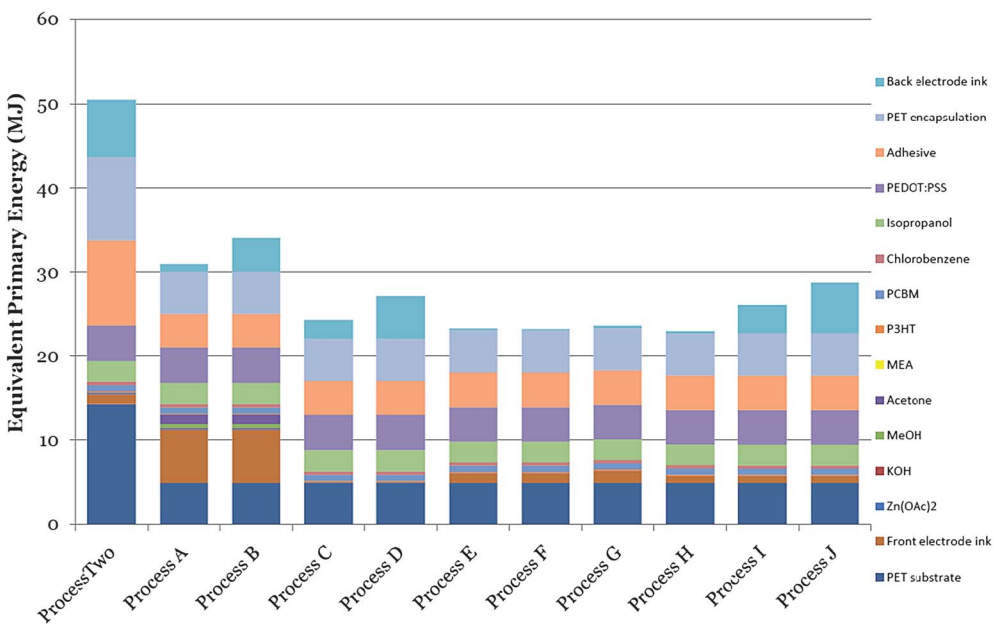

Fig. 6 Embodied energy in the materials per functional unit (m^2) in processes from A to J (MJ_{EPE}), ITO processes (ProcessOne and Process K) are not shown since they are significantly out of scale.

studied in the LCA studies for ProcessOne,[11] as well as Photoactive inks, solvents and other chemicals (potassium hydroxide, acetone, chlorobenzene, *etc.*).

The contribution to the embodied energy for each process is shown in Fig. 6. The electrode ink appears to have a greater contribution for processes A and B, which is due to the fact that PEDOT:PSS has a large embodied energy (159.41 MJ_{EPE} kg^{-1}) and is used in large amounts in those two processes.

3.2 Direct process energy

The R2R techniques that have been used for the production of solar cells are: slot-die coating, screen printing and rotary screen printing. Thus, the energy requirements at the R2R line are purely electrical. By computing the manufacturing time per functional unit (1 m^2) we can calculate the electrical consumption for all the equipment involved as shown in Table 9.

When comparing the share of the direct process energy in the manufacturing steps for processes from A to K, with ProcessOne and ProcessTwo (Fig. 7), we find that an enormous decrease is attainable, although the use of PEDOT:PSS as HTL is still appearing as the most energy demanding step. This is also observed when PEDOT:PSS acts as front electrode (*e.g.* the green coloured part of the bar chart for Processes A and B in Fig. 7) because of the low web speed employed and the consequently higher thermal input for drying.

The process with a semi-transparent silver-based electrode, named Process H, has the lowest embedded energy. The energy balance for Process H has been shown in Fig. 8.

3.3 Environmental impacts

The environmental impact of PET used as substrate and barrier encapsulation in these routes, shown in Fig. 9, is particularly high

Table 9 Electrical energy consumptions in Wh_{el} of 1 m² of OPV module processed surface in new all solution processed ITO-free routes from A to J

	A	B	C, D	E	F	G, H	I, J
S1 Front electrode processing							
Electrode coating	204.92	204.92	32.79	65.57	32.79	10.93	163.93
Drying	1844.26	1844.26	262.30	262.30	262.30	245.90	737.70
S2 ET layer deposition							
ZnO ink preparation	27.83	27.83	0	0	0	0	0
ZnO coating SD	81.97	81.97	81.97	81.97	81.97	81.97	81.97
Drying	368.85	368.85	368.85	368.85	368.85	368.85	368.85
S3 Active layer deposition							
P3HT:PCBM ink preparation	5.57	5.57	5.57	5.57	5.57	5.57	5.57
P3HT:PCBM coating SD	40.98	40.98	40.98	40.98	40.98	40.98	40.98
Drying	368.85	368.85	368.85	368.85	368.85	368.85	368.85
S4 PEDOT:PSS deposition							
PEDOT:PSS ink preparation	1.31	1.31	1.31	1.31	1.31	1.31	1.31
PEDOT:PSS slot die coating	74.52	74.52	74.52	74.52	74.52	74.52	74.52
Drying	670.64	670.64	670.64	670.64	670.64	670.64	670.64
S5 Back electrode deposition							
Slot die coating	10.93	5.46	5.46	65.57	10.93	10.93	5.46
Drying	245.90	409.84	409.84	295.08	245.90	245.90	409.84
S6 Lamination							
Encapsulation by R2R lamination	4.10	4.10	4.10	4.10	4.10	4.10	4.10
SUBTOTAL (W_{el})	3950.63	4109.10	2332.64	2305.32	2168.71	2130.46	2933.74
SUBTOTAL (MJ_{EPE})	40.64	42.27	23.94	23.71	22.31	21.91	30.18

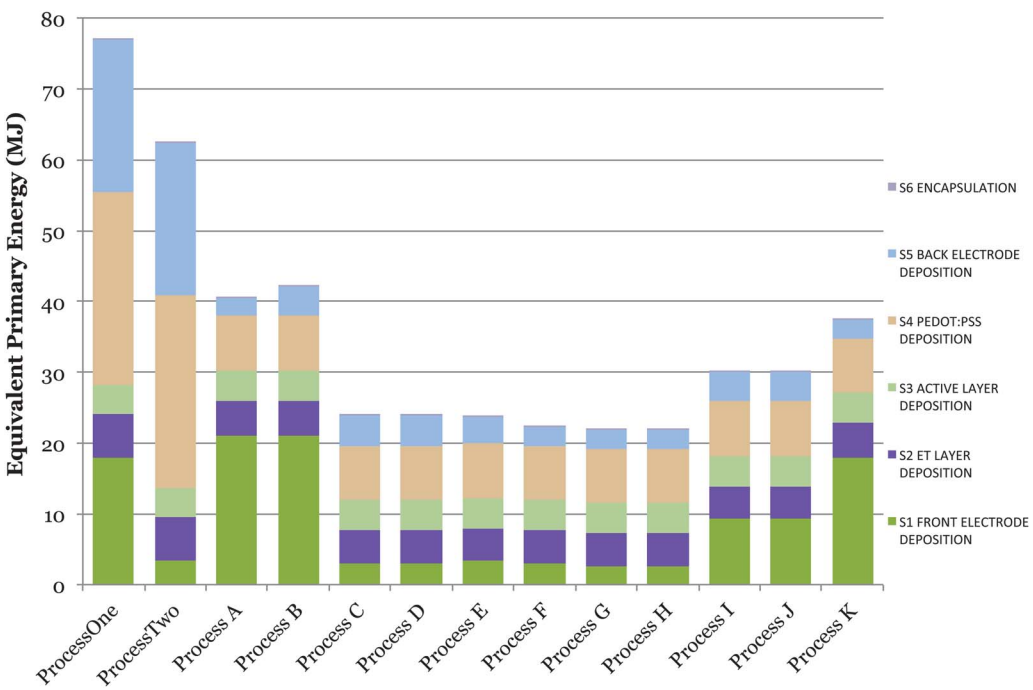

Fig. 7 Direct process energy employed for different steps in the manufacturing of OPV modules by routes A to K, and by ProcessOne and ProcessTwo.

since it is being produced by polymerization of two petroleum-based raw materials. This causes depletion of fossil fuel resources. Electricity consumed by the manufacturing equipment during manufacture also has a strong effect on fossil fuels since electrical input is produced to a large extent from fossil fuels in Denmark (80% of them in the electricity mix, 20% of the mix comes entirely from renewable sources).

Nonetheless, when comparing damaging impacts of ProcessOne, ProcessTwo and Process H (Fig. 10), there is a trend in the way of generally diminishing impacts, even in the high impacted fossil fuels category. One can thus conclude that the methodology has allowed for the successful development of a process that is more efficient at all levels.

4 Traversing the learning curve towards shorter EPBT

The processes developed have a significantly shorter energy payback time than the reference process, named ProcessOne. Among the batch of new routes, Process H, an EPBT of 1.42–0.71 years is achievable with a power conversion efficiency of respectively 0.5–1%. In comparison to ProcessOne with an energy

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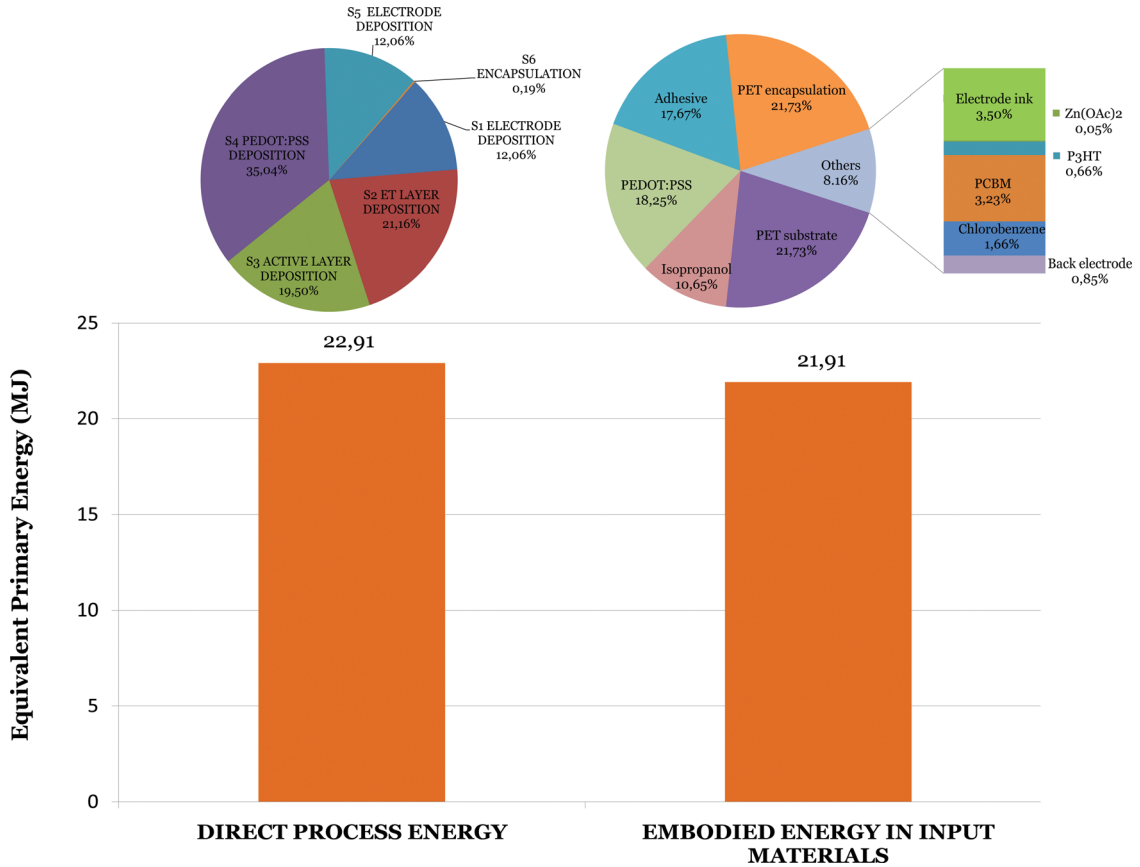

Fig. 8 Materials inventory share and direct process energy in Process H, both given in equivalent primary energy (MJ_{EPE}).

payback time of 2.02–1.35 years for respectively 2–3% efficiency this is a great leap forward and shows that it is possible to achieve a shorter energy payback time for a solar cell that at least in terms of efficiency is a poorer performer. It is also a less refined version and it can certainly be justified to assume that this can be improved even further, pending the same investment in development that *i.e.* ProcessOne has received. In Fig. 11 some of those avenues for improvement have been highlighted and they have been divided

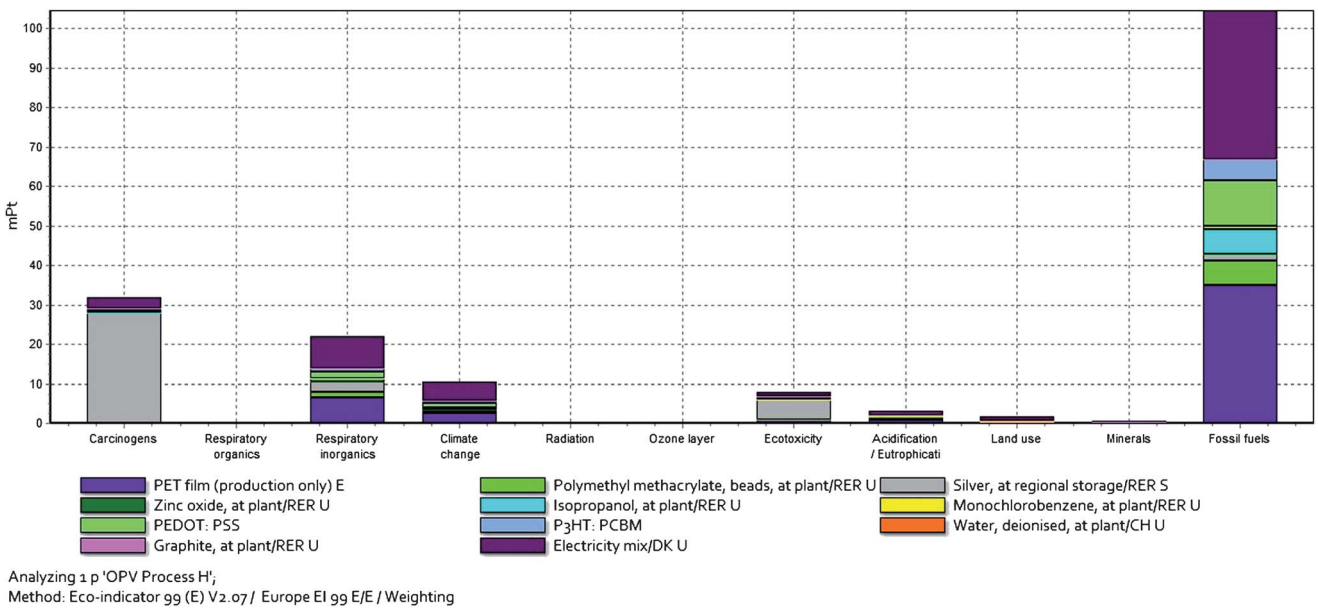

Fig. 9 Impact categories assessment weighted according to the raw materials used in the production of polymer solar cells by Process H. The methodology employed has been Eco-indicator 99, implemented in the SimaPro software, and developed under the Dutch NOH Programme by PRé Consultants.

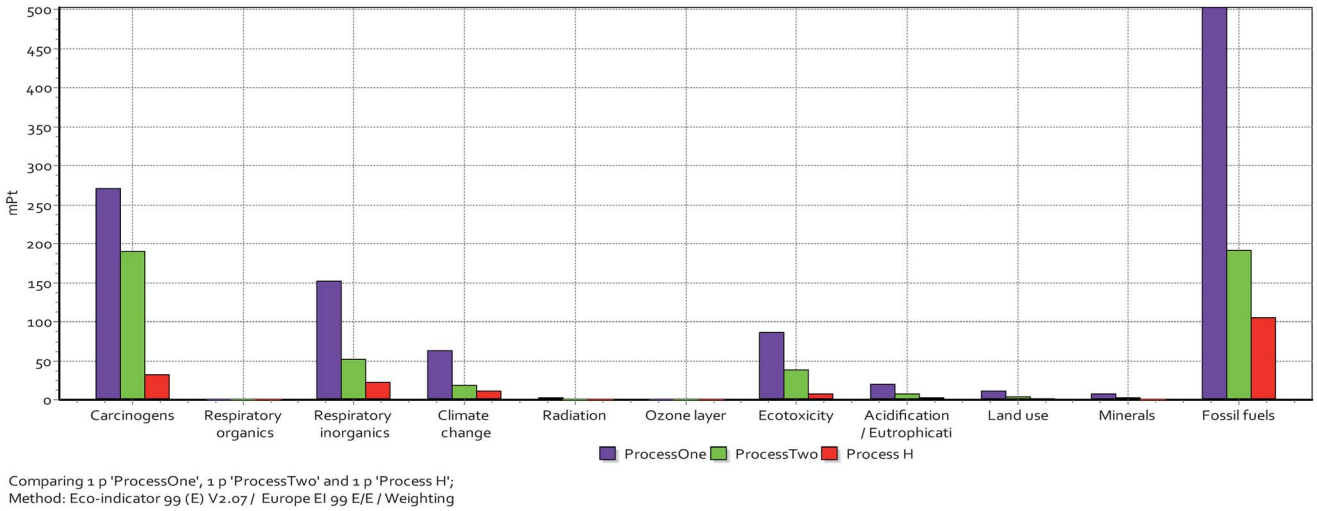

Fig. 10 Impact categories assessment corresponding to ProcessOne, ProcessTwo and Process H, weighted according Eco-indicator 99, implemented in the SimaPro software, and developed under the Dutch NOH Programme by PRé Consultants.

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into two groups according to their feasibility. One group represents how much improvement one could obtain by making very realistic developments and a second group shows how much improvement could be achieved provided that some more challenging yet realistic developments were made. Those latter assumptions will require some innovation (the former do not).

4.1 The feasible improvements

When examining Fig. 8 and Fig. 11 it is clear that the equivalent primary energy has been balanced both in terms of energy embedded in the materials and in the direct process energy. This has led to a reduction of the equivalent primary energy by a factor of ~10. Further improvements should obviously address the individual slices in the pie charts without leading to large unbalance. In terms of the direct process energy it is possible to introduce renewable energy sources very easily due to the fact that the running of a polymer solar cell printing and coating machine requires very little electricity and relatively little thermal energy. The low temperatures involved imply that a polymer solar cell production plant can be easily operated using solar electricity and solar thermal energy for drying and curing (the drying steps account for almost 90% of the total direct energy). It should be emphasized that the enabling feature is the low temperatures involved for OPV and this approach is unlikely to be efficiently applicable to PV technologies that require heating steps with temperatures much above 150 °C. Since PET is the largest contributor to the embodied energy in the module, a reduction in the thickness of the barrier—but also the adhesive—would decrease the embedded energy significantly. It should be possible to make an entirely functional encapsulated device with a thickness of 50–70 micron. Future work should also address the assessment of environmental impacts of a different range of potential plastic materials used as substrate in OPV modules: less-damaging, biodegradable or, even, recyclable plastics. Recycling organic photovoltaic scrap and re-introducing the waste as raw material in the manufacture is a cost-cutting measure but could also make the technology more sustainable: a real cradle-to-cradle technology.

Further improvements are obvious and the only reason they were not implemented in this work is due to the fact that a laboratory/pilot line was employed. Here a processing width of 305 mm with a working width of 250 mm is a constraint but a trivial one. Experiments with larger working widths have been shown to be very feasible.

Processing methods that enable full use of the processed area would be desirable. In the current version of Process H only 45% of the solar cell area is active and it has been shown that one can easily improve this to 67% and quite readily improve it to 85% and this significantly impacts the EPBT. Minor improvements would come from replacing the organic solvents employed with water as this has also been demonstrated as being possible.[48,52]

4.2 The challenging improvements

There are several achievable improvements that would require some innovation and therefore they represent valuable research targets. As shown in Fig. 11 and Table 10, these will only lead to a smaller improvement as compared to the feasible improvements described above. Of those the simultaneous formation of several layers in the solar cell stack would (provided that the technical yield is not compromised) lead to both reductions in energy usage and an improvement in overall processing speed. We also applied the projections for efficiency and lifetime set forward by the EC. Finally, recycling or end of use handling was also considered. The beneficial effects of recycling are difficult to estimate and we considered the energy gained by combustion of the solar cell and recovery of the metals (Ag and Zn) from the ashes.

The developments made from ProcessOne through Process H enable one to traverse the EPBT-efficiency plot *i.e.* to achieve significantly lower EPBT with lower efficiency (Fig. 12) and further improvements can be made through easily applicable and more challenging improvements. It is noticeable that the LCA directed improvements lead to improvements yielding better EPBT, despite the trade-off in performance; *i.e.* jumping from ProcessOne to Process H and further again—by assuming straightforward and more demanding challenges—means going down and left in the plot.

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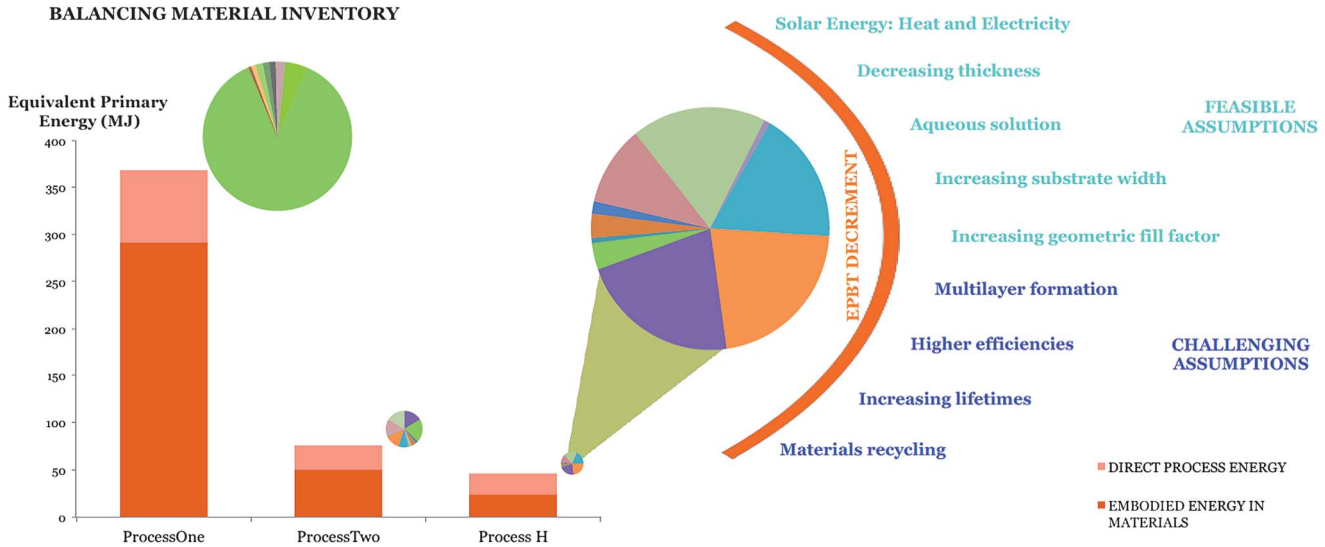

Fig. 11 Balancing the inventory of the polymer solar cells is crucial for making improvements even if this implies a sacrifice in performance. ProcessOne clearly has an unbalanced inventory with one component (the ITO-electrode) representing the majority of the equivalent primary energy (see pie-chart). By developing the polymer solar cell with the sole aim to balance the inventory and minimize the equivalent primary energy yields significantly lower values and a very balanced inventory.

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Table 10 EPBT in days *versus* real efficiencies and projections (shaded in grey colour) for Process H in its existing form and when improving following both feasible and challenging developments (shaded in grey colour). Data for ProcessOne are also listed for comparison

Efficiency	0.25%	0.5%	0.7%	1%	2%	3%	5%	10%	15%
ProcessOne	5938.7	2969.4	2120.9	1484.7	742.0	494.8	296.9	148.5	98.9
Process H	1034	517	369	259	129	86	52	26	17
Feasible assumptions	210	105	75	52	26	17	10	5	3
Challenging assumptions	82	41	29	21	10	7	4	2	1

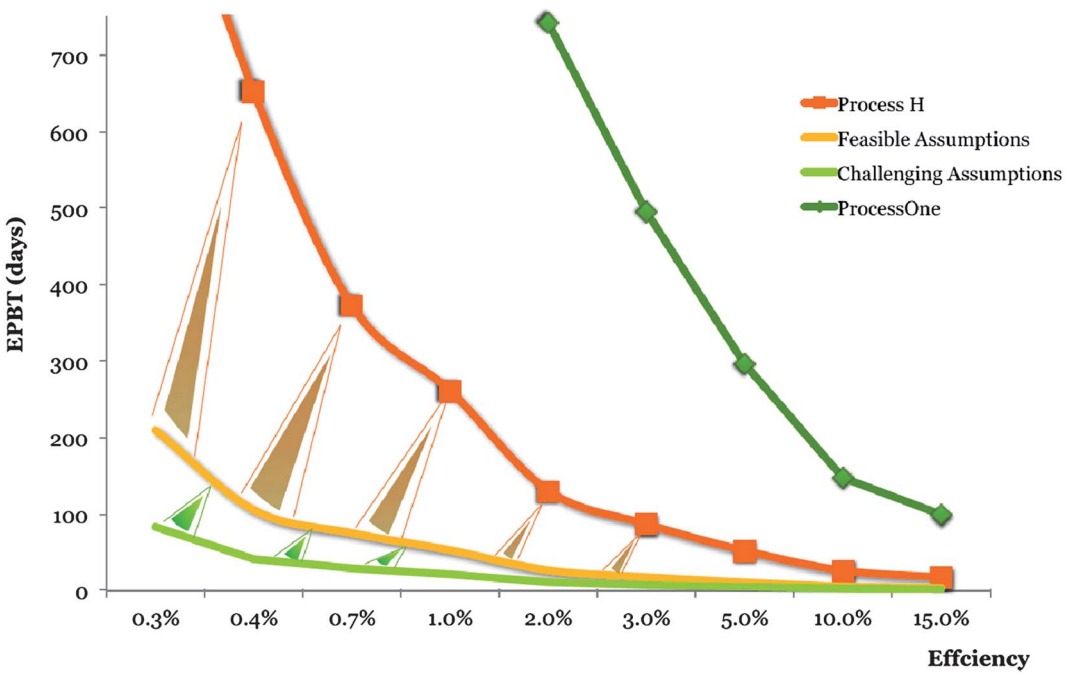

Fig. 12 Evolution of the EPBT guided by analysis (note that the scale is not linear).

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4.3 The factory

It was clear from Table 1 that the most performing renewable energy source is wind energy with energy payback times on the order of 95–140 days. It should be highlighted that this impressively fast energy payback time has been arrived at through a maturation of the technology over many years of operation and also that the technology does not scale with the 1-gigawatt-a-day challenge. This has been addressed within the wind energy sector by attempting to make larger and larger wind turbines placed in the most windy regions of the world (typically off shore). World energy demand is expected to increase by 50% each decade[13] and is estimated to reach 15 TW by 2050 as mentioned above. Global demand for each energy source will therefore increase, with fossil fuels accounting for over one-half of the increase in total primary energy demand. In one scenario where the electricity demand would be provided by PV, or in particular OPV, the manufacture of a 1 GW energy production capacity every day for satisfying this demand would be required. We envisage that the 1-gigawatt-a-day challenge can be efficiently addressed by OPV by assuming an operational lifetime of 1 year and a power conversion efficiency of 10%. This would require 1000 factories distributed around the globe near the equator in regions with high levels of insolation and in areas where land mass is accessible (*i.e.* desserts). This could also be viewed as a political instrument to lift developing regions of the world by providing them with an industry for covering the world needs. To illustrate this solution we consider a simple manufacturing installation (Fig. 13) that is 1/100 scale of the full size factory (*i.e.* 1/100.000 scale that fully addresses the 1-gigawatt-a-day challenge). We envisage the factory to be situated in southern Spain where the insolation reaches more than 1700 kWh m^{-2} per year.

It is evident that the factory is quite small both in terms of the solar electric and solar thermal installations, making the full-scale factory very realistic. The daily production capacity of only 108 m^2 also imply that the larger factory is easily envisaged. It is in fact possible to consider that significantly less and larger factories can address the problem since foil handling by printing and coating is currently achieved on the scale of 100 000–1 000 000 square metres a day. The choice of 1000 factories on a full scale was conceived to balance the system such that a very distributed manufacture across the globe is achieved. This ensures stability in terms of manufacture, electricity production and sensitivity (to insolation and politics). There are naturally challenges associated with the land use but it is noticeable that the manufacture of polymer solar cells according to Process H on a scale that fully addresses the 1-gigawatt-a-day challenge does not put any significant restraints on the current materials use. We have summarized in Table 11 the annual production of the various raw materials that enter the process and it is clear that even on a full scale (manufacture of 10^7 m^2 per day) this effort would claim very little of the currently employed materials. It is thus unlikely that an effort as outlined here would significantly affect the global raw materials cost due to speculation in raw materials as relatively little is required (significantly less than 3% of the world production for any of them).

5 Future and outlook

It should be evident that polymer solar cells house the potential to address the world's increasing energy needs. It is clear that there are several new developments in terms of system's integration that also need to be addressed. We estimate that the distribution of manufacturing facilities throughout the world will limit the extra cost associated with transport. We also find it highly likely that obvious questions of how one would distribute solar cells on this scale, connect them and replace them every year (or every few years as lifetime improves) can be solved. The most rational solution would include a structure that, depending on the location, would endure for many years thus requiring only replacement of the solar cell modules. It could be a concrete, steel or wooden structure. It is also of some importance to develop new connection methodologies and inverters that match this

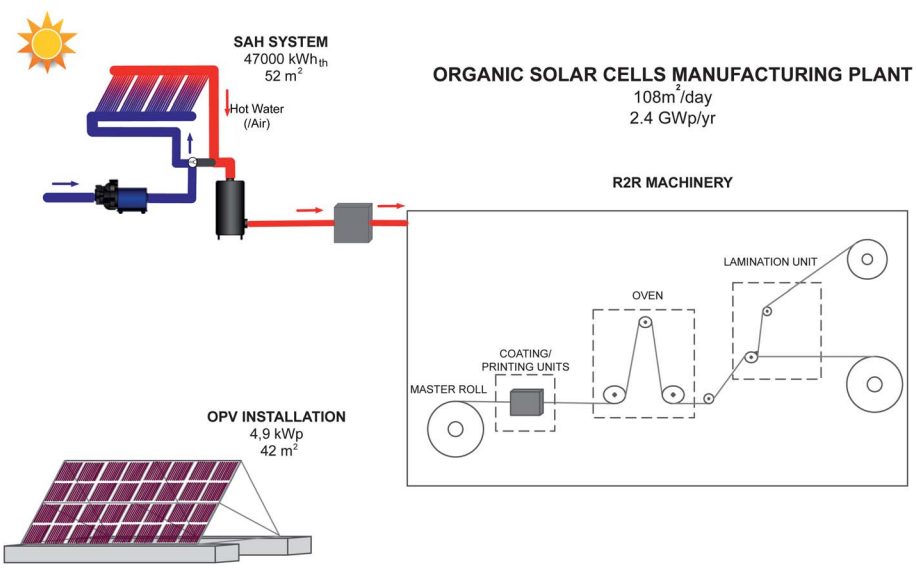

Fig. 13 A factory for manufacturing polymer solar cells using solar electric and solar thermal energy. The annual production capacity is 100 000th of what would be required to fully address the 1-gigawatt-a-day challenge.

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Table 11 Required raw materials for manufacturing 10^7 m^2 of OPV modules based on the Process H route, which would fully address the 1 gigawatt-a-day challenge as compared to the total annual production of those materials

Materials	Units per FU (g m^{-2})	Consumption (tonne/yr)	Yearly production (tonne/yr)	Share (%)
PET substrate	123.30	277425.00	4.90E + 07	0.57
Silver	0.63	42.52	19051.00	0.22
Adhesive	20.24	45540.00	2.05E + 06	2.22
Zn	0.30	663.93	1.00E + 07	0.007
Water	2.95	6639.34	—	—
P3HT	0.08	188.11	—	—
PCBM	0.07	150.49	—	—
Chlorobenzene	6.19	13905.00	—	—
Isopropanol	38.89	87491.80	—	—
PEDOT:PSS	26.23	59016.39	—	—
Graphite	4.59	10327.50	—	—

application. Operation at high voltages is likely to minimize resistive losses and will lead to lower use of conductor materials. The potential availability of surfaces on roofs and/or façades in existing and new buildings for building of integrated PV systems is also large in the developed regions of the world. We however consider the distribution in the form of solar farms in currently less developed regions as presenting a larger potential both in terms of development of poor regions and in terms of efficiency of scale.

6 Conclusions

We have demonstrated how the polymer solar cell technology can traverse the learning curve in terms of energy payback time and present shorter energy payback times through development according to directions arrived at through life cycle assessments, even when the efficiency is poorer. This is a clearly distinguishing feature of the polymer solar cell and confirms it as being a disruptive technology. We found that the use of renewable energy is particularly well suited to polymer solar cell manufacture, which is in stark contrast to all other PV technologies including other 3rd generation photovoltaics. We describe the requirements for a factory based on those observations and find that the manufacture of OPV at a rate equivalent to an electricity production capacity of 1 GW_p day^{-1} is possible while claiming very little of the available resources (significantly less than 3% at all levels). We finally observe that energy payback times as short as one day are possible when accounting for the projected performance in terms of stability and efficiency.

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