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Third Generation Photovoltaics – Early Intervention for Circular Economy and a Sustainable Future

Rhys G. Charles^{1*}, Matthew L. Davies², Peter Douglas^{3,4}

¹ COATED Engineering Doctorate, Swansea University, UK

² Materials Research Centre, College of Engineering, Swansea University, UK

³ Chemistry Group, College of Medicine, Swansea University, UK

⁴ School of Chemistry and Physics, University of KwaZulu-Natal, South Africa

* Corresponding Author, 290952@swansea.ac.uk / rhys.charles@hotmail.com, +44 7968 310 288

Abstract

Third generation photovoltaics (3GPV) which include dye-sensitised solar cells (DSSCs); organic photovoltaics (OPV); and perovskite solar cells, are promising green energy technologies in their infancy which hold the promise of low cost energy generation for the future. At this early stage in development, full lifecycle optimisation taking account of all parts of the product lifecycle is required to maximise the resource efficiency benefits associated with the use of these technologies to create a truly sustainable renewable energy technology. Here we examine the advantages of lifecycle optimisation for 3GPV technologies along with key aspects of design; materials selection; manufacturing processes; likely applications of the technologies; and potential recycling and refurbishment strategies. We identify features which are conducive to circular economy and identify barriers to resource efficiency for these technologies, and suggest some potential solutions and priority areas for future research.

1 Introduction

We are on the brink of significant climate change and face the limits of current linear economic models. Transition is necessary to a resource efficient ‘circular economy’ (CE) with widespread deployment of sustainable green energy technologies. Retention of materials within the economy through recovery and regeneration of products at the end of each service life maximises their economic productivity, offsetting demand for primary resources and decoupling growth from resource consumption. CE is regenerative by design, and replaces the concepts of ‘end-of-life’ (EoL) and ‘waste’ with ‘restoration’ and ‘resources’. Key features include elimination of waste through industrial symbiosis, superior design, appropriate business models and reverse logistics systems [1].

CE lends itself to leasing and take-back, as well as the transition to a service based economy in which consumers become product users rather than owners and manufacturers retain ownership, which facilitates effective take-back, reuse, refurbishment/remanufacturing and recycling. Resource efficiency afforded by CE yields economic, social and environmental benefits [2].

Prioritising reuse and repair > refurbishment/upgrade > remanufacturing > recycling; results in greatest resource efficiency benefits and larger savings in em-

bedded costs (economic and environmental) of products and components. Whenever costs of reverse logistics and returning products to market are lower than production costs in linear models, circular systems afford greater value than linear alternatives. Benefits are amplified by cycling resources in consecutive product lifecycles and extending the useful life of products. The economic benefits of CE are expected to become more important in the future as prices of primary raw materials rise [3].

CE presents opportunities for substitution of virgin materials by cascading products, components and materials across multiple product lifecycles. Resource efficiency benefits result from using cascading materials in new applications, since more of the embodied costs (labour, energy, materials) are retained than is achievable through traditional recycling pathways. This creates opportunities for organisations to valorise ‘waste’ through industrial symbiosis. Resource efficiency gains from use of post-consumer materials/components are enabled by design for disassembly and materials separation, which reduces costs of reverse cycles and maintains materials quality and longevity of viable use within the CE [1].

Growing recognition of these benefits, rising/volatile resource prices [3]; global resource criticality concerns [4]; and rising production costs has made CE an attractive prospect particularly for manufacturers who

rely on supplies of critical raw materials (CRMs). Materials criticality issues and environmental impacts associated with the use of toxic materials in devices can also be mitigated with appropriate circular practices. Additionally, intangible company assets such as brand value may be enhanced as consumers become increasingly environmentally aware [3]. Organisations such as First Solar who produce CdTe PV have therefore adopted business models that unlock the power of CE and generate value through development of appropriate recycling technologies; long product lifecycles; and linking value chains with other industries and supply chain partners.

Commercial viability of PV is based on the levelised cost of electricity (LCOE) generated, determined by the power conversion efficiency (PCE), cost, and lifetime, of PV products. Resource efficiency benefits afforded by CE can potentially reduce the economic and environmental costs of module production, enhancing commercial viability and increasing competitiveness with alternative renewable energy technologies. Energy payback time (EPBT), emissions associated with electricity generation ($\text{CO}_2\text{eq/kWh}$) and the cost of energy generation ($\$/\text{Wp}$) can all be reduced through adoption of circular practices. Studies have shown that EPBT for Si wafer based PV technologies are reduced by half through use of recycled materials [5]. For CdTe PV it has been predicted that, as PCE improvements are made, and available volumes of EoL modules increase, demand for CdTe for PV could be satisfied exclusively by secondary supplies from EoL modules alone [6]. The value of take-back and recovery in this case is enormous. The magnitude of these benefits from lifecycle optimisation is determined by the effectiveness of eco-design, which is greater when conducted at the earliest possible stage in development of technologies, and in collaboration with all parties involved in product lifecycles.

3rd generation photovoltaics (3GPV) which include dye-sensitised solar cells (DSSC) [7], organic photovoltaics (OPV) [8], and lead halide based perovskite solar cells [9] are promising low cost green energy technologies in their infancy. 3GPV offers advantages over previous PV technologies including: the use of low cost abundant materials; manufacture by roll-to-roll (R2R) printing technologies; flexible light-weight devices; suitability for building and product integration; superior performance in diffuse light conditions; and a range of aesthetic possibilities such as tuneable colour and transparency.

Although viewed as a ‘green’ technology, 3GPV has environmental impacts associated with its production and potentially will contain hazardous components. Widespread deployment will require a continued supply of critical raw materials (CRMs) and full lifecycle

optimisation for CE is necessary at this early stage in development to create truly sustainable technologies. This includes: minimisation of environmental impacts associated with production; development of EoL strategies and processes; design for longevity; cradle-to-cradle design optimisation; selection of low impact materials; and substitution of primary resources and CRMs. This is achievable through the process of eco-design [10] which considers: selection of low impact materials; optimisation of production techniques and reductions in material usage; optimisation of business models and logistics systems; reduction of impacts during use; optimisation of initial life stages of products through design for longevity, upgrade and repair; and optimisation of EoL systems.

Full lifecycle optimisation also requires cross-sector collaboration between all parties involved in product lifecycles including: academics, manufacturers, waste managers and designers, to enable cascades of reuse, remanufacturing and recycling, and to ensure appropriate circular flows of products and materials for optimal economic, environmental and social benefits.

Here we present an assessment of key features of design, production, reuse and recycling of 3GPV, and the future priority research areas necessary to ensure truly sustainable photovoltaic energy generation. These include: likely applications, architectures and manufacturing processes of future commercially available 3GPV; attributes of these technologies which are conducive to circularity; appropriate materials selection; potential recycling/remanufacturing processes and strategies; barriers to circularity/sustainability for 3GPV.

2 Current position

First generation crystalline silicon (c-Si) PV devices are the dominant product on the market today, due to high PCE and stability. Devices are however fragile, expensive and have relatively high embodied energy compared to successive generations of PV [11]. The 2nd generation of thin-film technologies which include amorphous silicon (a-Si), cadmium telluride (CdTe), copper-indium-selenide (CIS) and copper-indium-gallium-diselenide (CIGS), have begun gaining market share, accounting for ~7% of global PV production in 2015 with some projections showing an increase to 50% by 2030 [12]. Although offering lower PCEs than c-Si PV, 2nd generation PV require less materials and energy for manufacturing and offer lower cost electricity generation, short energy payback time, and reduced emissions associated with electricity generation [13]. In addition, flexible devices can be created. Despite these advantages, manufacturing involves costly vacuum processes; and devices contain toxic

materials (such as Cd) and CRMs (e.g. In, Ga, Te), the use of which may limit widespread deployment of these technologies [6].

In light of these issues, we are now witnessing the emergence of 3GPV, or printable PV (PPV) which are thin-film devices based on molecular photoactive layers, potentially manufactured from earth abundant materials using cheap roll-to-roll (R2R) production. Early versions of DSSC and OPV devices for niche applications are now commercially available, and research into new materials, improved device performance and superior manufacturing processes is ongoing. Perovskite solar cells [14], the newest of the PPV technologies, are yet to emerge on the market as issues with device stability are yet to be fully addressed. However, PCEs of lab based perovskite devices have already reached 22.1% [15], which is comparable to record cell efficiencies for competing thin-film technologies.

3 Architecture and operation

3GPV modules are composed of individual solar cells, which are electrically connected and encapsulated in EVA or glass to form flexible or rigid modules respectively. The various 3GPV technologies share common features in terms of cell architectures and material sets. Substrate and electrode materials for example are commonly used in all technologies as are encapsulant materials. Some of the active materials of cells are commonly used across technologies, however a wide range of active materials and architectures have been explored in research, complicating the issue of materials selection in future devices. To highlight issues relevant to CE we limit our discussion to ‘Sandwich’ cells (Figure 1), with working and counter electrodes on different substrates.

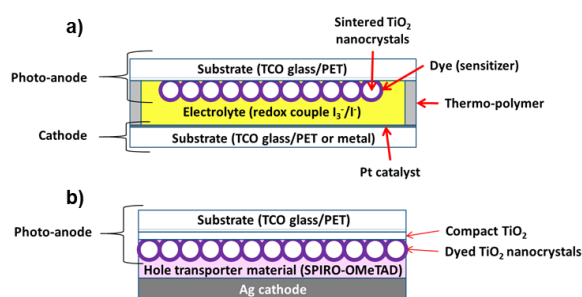


Figure 1: a) Dye-sensitised solar cell (DSSC - Grätzel Cell); b) solid-state dye-sensitised solar cell (ssDSSC). (Adapted from [14])

3GPV cells are composed of two electrodes, one of which must be transparent i.e. TCO coated glass or a plastic, such as PET, for flexible devices. The second electrode can be made of either a similar substrate or a metal foil.

DSSCs have been created with various architectures and materials sets [16]. Generally, these are composed of two electrodes, the photo-anode, and a cathodic counter electrode (Figure 1a). The photo-anode is composed of a substrate coated with a mesoscopic layer of a semiconductor, commonly TiO₂, which is dyed with a sensitizer species capable of absorbing visible light. The counter electrode is composed of either a second TCO substrate coated with a catalytic layer of Pt, or a metallic substrate such as Ti. Electrodes are sealed together with a thermopolymer, and the cavity between is filled with an electrolyte iodine/triiodide or cobalt based redox couple [17]. When illuminated, absorption of a photon by the dye species creates an exciton (electron-hole pair). Rapid injection of the electron from the dye into the conduction band of titania follows, enabling the electron to diffuse through the TCO of the anode, around a circuit, and back to the counter electrode (CE). The oxidised dye is reduced by the electrolyte, which is, in turn, reduced at the Pt of the counter electrode.

Solid-state DSSCs (ssDSSCs) (Figure 1b) have also been developed although they are yet to be commercialised. Their structure and function is analogous to liquid DSSCs with the electrolyte replaced by a solid hole transport material (HTM). These can be inorganic or conjugated organic species, capable of reducing oxidised dye species back to the ground state and thus transport the hole resulting from the generation of an exciton to the counter electrode. A compact titania layer is also used between mesoporous titania and the TCO on the anode which functions as an electron transport layer (ETL) and blocking layer preventing short circuiting through contact of the HTM with the TCO of the anode. Devices use FTO glass or ITO PET as transparent electrode substrates, or a metal laminate such as Ti. The catalytic Pt layer is not necessary in ssDSSC.

Perovskite solar cells use methylammonium lead halides (CH₃NH₃PbX₃; X=Cl, I or Br). These crystallise in a perovskite structure, which gives the cells their name, (Figure 2). The first perovskite cells used DSSC type architectures with perovskite infiltrated in a titania scaffold as a sensitizer, these are referred to as mesosuperstructured solar cells (MSSCs) (Figure 2a) [18]. Later it was found that perovskites themselves function as excellent electron transport materials and so porous perovskite heterojunction devices (Figure 2b) and perovskite p-i-n heterojunction cells (Figure 2c) without titania/alumina scaffolds have been created. The most efficient devices use Au as a

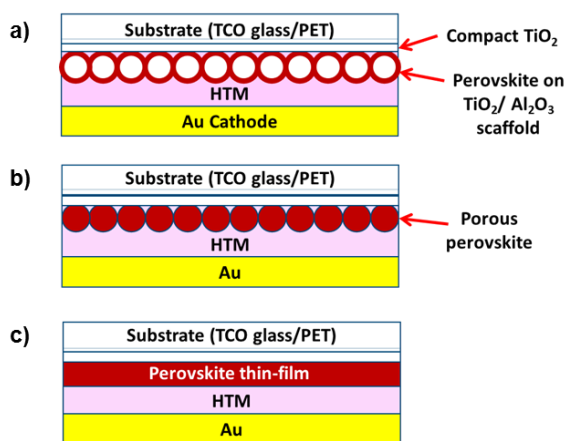


Figure 2: Example architectures of perovskite solar cell a) perovskite based mesosuperstructured solar cell (MSSC); b) porous perovskite heterojunction c) perovskite p-i-n heterojunction. (adapted from [14]).

contact. Both organic and inorganic HTMs have been employed in devices.

Between the electrodes of OPV devices are two organic materials with extended conjugated π -orbital systems. The first is generally a polymer material such as 3-hexylpolythiophene (P3HT), which acts as a light absorber. The second functions as an ETL, typically a fullerene compound such as phenyl-C61-butyric methyl ester (PCBM). These organic materials can be deposited as individual thin films (Figure 3), or can be combined and deposited as a single film polymer blend.

4 Applications of 3GPV

Currently, stability and PCE problems prevent the production of commercially viable large area 3GPV devices, and early forms of 3GPV emerging on the market are for niche applications. Building-integrated photovoltaics (BIPV) to date have been mounted on south facing roofs to achieve good efficiency. 3GPV can be applied vertically to walls and windows. Retrofitting existing buildings which were not designed to bear the additional weight of other forms of PV is also possible. Interesting new product integration possibilities for 3GPV also exist, particularly at the dawn of the ‘internet of things’ where electronics will communicate wirelessly and require off-grid energy supply. Additional possibilities arise from the transparent nature of 3GPV, such as combination with existing PV technologies in tandem devices for higher PCE.

In the immediate future DSSCs will likely find most use in product integrated applications. One example is the GCell [19], produced in Wales. This flexible prod-

uct has found applications in solar backpacks for charging consumer electronics and keyboard folios for iPads. Sony has produced prototypes of their Hana-Akari (flower lamps) for indoor use, which use glass based DSSCs to charge batteries to power LEDs. An example of BIPV DSSCs include the façades of the SwissTech Conference Centre.

The stability issue for perovskite cells is such that encapsulation in flexible devices does not at this time produce devices with sufficient stability for commercial viability [20]. Ingress of moisture and air results in rapid degradation of the perovskite, and applications for perovskites at this time are therefore limited to rigid devices which encapsulate the materials within glass. The earliest perovskite products will probably therefore be ‘tandem cells’, in which a perovskite device is combined in tandem with existing PV technologies. This is the goal of Oxford PV who are developing and commercialising thin film perovskite solar cells for printing directly onto Si or CIGS modules.

Flexible OPV products have emerged on the market including Heliatek’s Heliafilms® for use in BIPV applications, and in the automotive sector for integration with car roofs [21]. Solar phone chargers, solar adhesive tapes and flexible solar foils are commercially available [22]. Such products are suited to retrofitting of buildings, windows and consumer electronics.

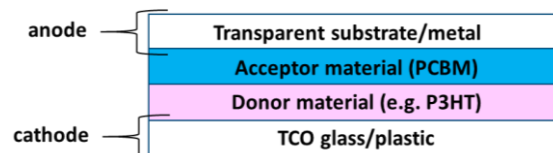


Figure 3: Organic photovoltaic (OPV) solar cell device architecture

5 Manufacturing processes

3GPV is cheap because it can be made using R2R production on flexible substrates [23] using solution deposition of materials. In such processes, rolls of substrate are run through a series of sequential deposition techniques in which each of the layers of solar cells are deposited as thin films (10 nm–10 μ m), with the final coated product recoiled at the end of the line (Figure 4). The result is rapid production at relatively low cost. Substrates include metals such as steel for functionalised building envelopes, or ITO PET for transparent devices. A variety of solution based coating techniques are possible including: bar coating, screen printing, spray deposition, dip coating, slot-die

coating and inkjet printing. Coating is followed by thermal treatment to drive off solvents and cure films. Convection ovens, and hot plates, have been employed but higher throughputs, shorter processing times, and greater energy efficiency is achievable with NIR curing [24]. Photonic flash annealing is another promising option currently under research [25]. For glass-based devices, R2R is not a possibility and glass must be processed in sheets.

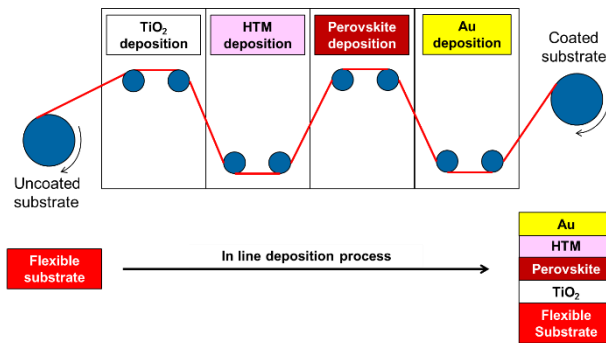


Figure 4: Principle of roll-to-roll production of planar p-i-n perovskite cell as shown in Figure 3c

Despite the suitability of R2R production for resource efficient manufacturing, there are still issues which need to be addressed: some components, such as gold cathodes require vacuum deposition techniques which are relatively high in cost, energy demand, and material wastage during processing; masks used in deposition processes retain material; there is the waste issue of production scrap; and solvent loss in thermal treatment. In addition, many of the solvents used for deposition of thin films such as DMF and chlorobenzene are hazardous and much research is underway to replace these. Many new environmentally-friendly solvents are becoming commercially available, such as Cyrene® derived from cellulose, which exhibits similar solvent properties to DMF and may be suitable for the deposition of perovskite thin films [26].

An additional advantage of solution deposition is that devices manufactured in this way are able to be recycled with ‘reverse manufacturing’ type processes using selective dissolution of layers with the same solvents used for deposition. This has been demonstrated for perovskite cells in the lab [27]. Solutions of recovered materials may then be used to manufacture new devices.

6 Materials selection

In the interest of circular economy and sustainable economic development, materials selection can no

longer be based solely upon the factors considered for commercially viable PV to date. Production cost, lifetime and efficiency of devices must be balanced with the benefits afforded over multiple product generations through refurbishment, upgrade, and the reuse of recovered components and materials. Materials selection must therefore take account of compatibility with EoL processes.

Numerous lifecycle analysis (LCA) studies have shown that substrates represent a large proportion of the embodied environmental and economic costs of PV cells [28], and that these costs are lower for PET substrates than glass [7]. Recovering substrates for reuse is therefore important and delamination of modules is necessary. Mechanical delamination usually destroys substrates and thermal delamination degrades PET. LCA has also shown that laminate materials represent a significant proportion of the embodied costs of thin-film modules [29] and so its recovery is also desirable. So there is a need to develop new delamination methods, new laminate materials, and/or alternative flexible transparent substrates which are compatible with thermal delamination. There is also a case for the use of glass (flexible glass is available through expensive) in favour of PET despite its higher embodied cost based upon net resource efficiency benefits over time. Organic active materials may also be degraded in such processes, and so consideration of the benefit of their recovery following mechanical delamination in favour of substrate recovery with thermal delamination is also necessary.

The use of metals as electrodes also requires careful consideration. These are readily recyclable, however their initial deposition in devices is usually by relatively high cost, high energy, high wastage vapour deposition techniques unless devices are printed directly onto metallic substrates.

3GPV technologies utilize numerous critical raw materials. The traditional TCO used in devices has been ITO. However, due to the rising price and global criticality of indium, resulting from supply bottlenecks and demand for ITO for flat screens, this has been replaced by FTO on glass in solar cell applications. Replacement with FTO on PET has been problematic due to the high temperature PVD process used for FTO deposition and degradation of PET in the process. Mitigation strategies enabling ITO substitution may result from research into new low temperature methods for deposition of TCOs such as RF magnetron sputtering [30-32]. Substitution with graphene coated PET may be a suitable solution, however its cost is currently prohibitive for commercial application. Carbon nanotubes (CNTs) which can be printed on substrates have also been explored as TCO alternatives [33]. Another potential CRM issue arises from

the use of Ru in the dyes for DSSCs, and there is much work on fully organic dyes as replacements [34]. Their use mitigates the criticality issue associated with Ru and they are compatible with current dyeing processes. Further investigation is necessary however into their degradation mechanisms and whether they can be converted easily back into their functional forms for reuse.

An alternative strategy to substitution for mitigating resource criticality issues is to decouple supply from primary production by developing secondary supplies from within the circular economy, including supplies from EoL devices and cascaded materials derived from wastes available within the circular economy (industrial symbiosis). Examples of lab scale processes for production of 3GPV materials from waste include the production of perovskites from lead-acid car batteries [35], production of carbon based counter electrodes from Li-ion batteries and generation of platinised counter electrodes for DSSCs from waste thermocouples [36].

Plastic substrates are derived from crude oil so biologically derived alternatives are an attractive prospect. Transparent flexible substrates composed of cellulose nanocrystals (CNCs) have been used in OPV cells and shown to have high transparency and appropriate surface roughness for this application. In addition, OPV devices on CNC substrates have been shown to be readily recyclable due to the solubility of substrates in water [37]. Where plant derived materials are used, the carbon sequestration benefit will also contribute to reducing emissions associated with electricity generation.

7 Product integration

It is likely that the lifetimes of buildings will be considerably longer than the target lifetime of 25-30 years for 3GPV. Full integration into buildings therefore presents issues once modules degrade and reach EoL, and a 'roll-on/roll-off' approach may be useful. OPV lifetimes are more aligned with that of consumer electronics products and so these devices may be most suitable for integration with product. DSSCs on the other hand present an interesting opportunity in terms of *in-situ* refurbishment. DSSCs fail due to degradation of electrolytes and dyes. But cells can be flushed of electrolyte and re-dyed in a period of 5 mins [38]. Modification of DSSC design to accommodate a re-dyeing process presents possibilities for *in-situ* refurbishment and upgrade.

Integration of PV into consumer electronics will also result in complex EoL issues. It is likely that much 3GPV will end up incorporated into low value domes-

tic appliances that currently do not justify manual disassembly. Common practice is to shred such devices and separate materials with automated processes. If this occurs, then the material resources in the PV will be dissipated amongst the bulk material fractions and lost from the CE. To address this issue cooperation between PV manufacturers and their clients who purchase PV for integration with their products will be necessary in order to optimise the design of electronics so that PV can easily be isolated. The cost benefit of returning PV from where it is globally distributed to electronics manufacturers for reuse in new/refurbished products, or to PV producers for recycling, is likely to be poor due to the low inherent material value of 3GPV devices small enough for product integration. The 'refurbishability' of DSSCs also presents interesting opportunities for product integration. As the devices could potentially be 're-charged' they could retain value after degradation which may justify isolation from products and return to manufacturers for refurbishment using reverse logistics system resembling those currently used for printer cartridges for example.

8 Conclusion

3GPV technologies hold great potential as a sustainable renewable energy source for the future. With full lifecycle optimisation which takes account of EoL processes during design, enabling reuse of substrates and active materials in successive product generations, these technologies could provide the lowest levelised cost of electricity for PV to date.

DSSCs show great potential as the first 'refurbishable/upgradable' PV device due to the ability to replace dyes and electrolytes repeatedly with no observable loss in functionality over many product generations. Coupled with the numerous aesthetic possibilities for all 3GPV technologies, interesting possibilities in terms of building and product integration exist.

Priority research areas to enable full lifecycle optimisation include: methods of module lamination/delamination which do not degrade material components of cells and modules; substitution of CRMs; processes for generation of secondary resources from 'wastes' available within the CE and EoL devices; development of biologically derived cell components such as CNC based substrates; and methods which enhance resource and energy efficiency of R2R manufacturing such as solvent capture and recovery of production scrap.

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