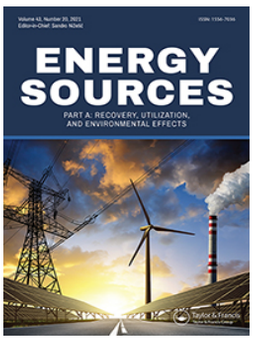

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Environmental assessment of transparent conductive oxide-free efficient flexible organo-lead halide perovskite solar cell

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ABSTRACT

Perovskite solar cells (PSCs), one of the third-generation photovoltaic (PV) technologies, have recently become a very popular topic in photovoltaic research. This technology, which is a candidate for commercialization in the future, needs to be evaluated from an environmental point of view. The amount of electricity consumption is the most important factor that directly determines the environmental impact values of photovoltaic cell manufacturing. Transparent conductive oxide (TCO) coated glass is one of the major contributors to electricity consumption in PSC architecture. It is therefore useful to investigate the environmental profile of TCO coated glass-free PSC architecture with conventional PVs. One of the solutions to this issue is manufacturing PSC on a flexible substrate. Flexible PVs are considered to be one of the most promising candidates for mass production with its advantages of low-temperature manufacturing, higher efficiency with a lower weight, portability, and compatibility with a roll to roll fabrication. In this work, we show that the environmental impacts of a representative PSCs with a flexible substrate. While the energy payback time (EPBT) of the flexible PSC is already competitive with commercial PVs, the device must reach a 25-year cell lifetime for its global warming potential (GWP) to reach a reasonable range.

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KEYWORDS
Life Cycle Assessment; perovskite PVs; sustainable Energy

Introduction

There is a growing consensus that a large shift from fossil fuels to renewable energy resources is necessary in order to combat climate change and other pressing environmental issues. One of the most important renewable energy sources is solar energy due to its abundance, wide availability, and increasing cost-effectiveness. Today's photovoltaic (PV) technology converts photon energy from the Sun directly into usable electrical energy with remarkable efficiency. Although the contribution of PV electricity generation to global energy production is only small today, the rapid increase in investments made in this sector is an indication that these technologies will be an important alternative to fossil fuels in the long term (Gbadamosi and Nwulu 2020).

PV technologies are generally defined in three generations, which are wafer-based crystalline silicon solar cells, thin-film solar cells, and third-generation PV technologies. First-generation silicon-based photovoltaic technologies currently account for more than 90% of commercialized solar cell applications, while second-generation thin-film technologies which have a lower cost but also lower efficiencies than silicon solar cells represent the remaining percentage in commercial applications (Fraunhofer Institute for Solar Energy Systems I 2020). Third-generation PV technologies have many potential advantages in terms of ease of production and cost-effectiveness, but they are not at the level where they can compete with the commercialized technologies in terms of stability.

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Perovskite solar cell (PSC) technology is an example of third-generation PVs. Organo-lead iodide perovskite ($CH_3NH_3PbI_3$) was first used in dye-sensitized solar cells instead of organic dye (Kojima et al. 2009). Research has shown that perovskite materials have better absorption ability than traditional dye (Im et al. 2011). In 2012, $CH_3NH_3PbI_3$ started being used as an active layer with a solid hole conductor material instead of a liquid electrolyte (Kim et al. 2012). This new technology offers many advantages in terms of low cost and material flexibility (Wang et al. 2015). The large-scale photon absorption ability of $CH_3NH_3PbI_3$ allows the production of highly efficient devices with very thin active layers (~300 nm) compared to silicon (300 μm) and thin film (2 μm) absorber layers (Yin et al. 2015). This opens a path to producing more efficient devices using less material (De Wolf et al. 2014). Today, PSCs have exceeded 25.2% of power conversion efficiency (NREL 2019). Although PVs are environmentally friendly technologies, some environmental impacts ensue throughout device production processes. Therefore, preparatory to the commercialization of PSCs, it is crucial to investigate its environmental performance.

Major features of a transparent conducting oxide (TCO) layer deposited on a glass substrate are a metal-like transmission capability, appropriate work function, and the ability to transmit photons into a cell with a wide range of wavelengths (Minami 2005). Two commonly used TCO materials are Indium Tin Oxide (ITO) and Fluorine Tin Oxide (FTO) (Xie et al. 2017; Zhao et al. 2017). Researches have shown that ITO has several advantages in respect of ohmic behavior and conductivity (Chander et al. 2017; Purohit et al. 2017). On the other side, FTO is generally preferred in thin film solar cell fabrication because of crystallinity and electrical conductivity performances (Chander and Dhaka 2017a, 2017b), price advantage (Michael 2012) and avoidance of the critical metal Indium, which is commonly used in thin film applications (Department of Energy 2018). On the other hand, it is shown that TCO substrates can cause transmission loss in PSC device (Hu et al. 2017). Previous life cycle assessment studies also have shown that TCO coated glass has the highest portion of electricity consumption in the perovskite solar cell manufacturing process (Espinosa et al. 2015; Sarialtin, Geyer, and Zafer 2020; Serrano-Lujan et al. 2015). Considering that the amount of electricity required for manufacturing PSCs is the largest factor in determining their environmental profile (Espinosa et al. 2015), it is important to investigate device architectures with the elimination of this layer. Flexible substrate applications such as Polyethylene terephthalate (PET) are suitable for PSCs because it allows the device to be more lightweight and bendable compared to glass or other rigid substrates (Dou et al. 2017; Popoola, Gondal, and Qahtan 2018). It was shown that the production temperatures of organic-inorganic $CH_3NH_3PbI_3$ perovskite technologies can be controlled to be in the range of 100–150°C (Heo et al. 2013) and the flexibility (Poisson's ratio) of the perovskite material is suitable for flexible production (Feng 2014).

In the presented work, life cycle assessment is performed for one representative flexible PSC device with 14% power conversion efficiency as described in the original publication (Li et al. 2016). First, the environmental impact values (amount of energy Joule per m^2 of cell produced) for each layer in flexible PSC architecture were determined. Second, electricity consumption values of manufacturing the device were specified and EPBT was calculated. Finally, GWP values (per m^2) were converted to 1kWh electricity production (second functional unit) in order to compare this value with those of commercial PVs.

Methodology

Life cycle assessment

Life cycle assessment (LCA) is a method that is used to determine the environmental impacts of products or processes across all stages of their life cycles, including the acquisition of raw materials used in the production, transportation, consumer use, and disposal (Guinée et al. 2001). Decision-making regarding emerging technologies increasingly accounts for environmental considerations such as natural resource use, climate change potential, and other environmental impact categories. LCA is

a method that is frequently used in decision-making processes and has its own ISO standards (ISO 14040:2006 2018). It consists of four stages, which are goal and scope definition, inventory analysis, impact assessment, and interpretation.

LCA of photovoltaics is usually performed using electricity generation (for example, 1 kWh) or examined cell area (m^2) as functional unit (Frischknecht and René Itten 2015). Two types of system boundaries are common in LCA: from raw material to end of life (Cradle to grave) and from raw material to factory gate (Cradle to gate). Since PSC has not yet been commercialized, analyses are usually carried out using the cradle to gate method.

Goal and scope

The goal of this work is to determine the life cycle impacts of a representative flexible PSC device that has demonstrated good productivity in a novel work (Li et al. 2016). The structure of a typical perovskite solar cell is composed of five main layers which are fluorine-doped tin oxide (FTO)/ indium-doped tin oxide (ITO) glass substrate, TiO_2 electron transfer layer (ETL), $CH_3NH_3PbI_3$ active layer, Spiro-OmeTAD hole transfer layer (HTL) and Au/Ag back electrode (Chilvery et al. 2016). The flexible PSC device examined in this study used alternative materials different from conventional architecture not only for the substrate but also for other layers except for the perovskite active layer. Table 1 shows the handicaps of conventional materials and the alternative materials studied in this work. Due to the toxicity of lead, researchers have been trying to find appropriate substitute material such as tin. However, tin is reported to have higher environmental impacts than lead in PSC devices (Serrano-Lujan et al. 2015). According to (Minami 2005), other disadvantages of tin are its cost and high resource scarcity. Therefore, lead is still a better option as active material in PSCs.

The functional units of the assessment are the manufacturing energy (Joule) per 1 m^2 of cell production and 1 kWh of electricity generated by the cell. The system boundary for this LCA is chosen as cradle to gate. Other important parameters are shown in Table 2.

Life cycle inventory

An inventory table has been created by the benefit of the literature and the ecoinvent database (Table 3). Several customizations have been implemented for the materials that are not found in databases (see supplementary material).

In the reference work, 7 µm ultraviolet resin (we assumed butyl acrylate) coated 50 µm PET substrate is printed by nanoimprinting lithography (% 3.2 Surface area with 2 µm thickness) and

Table 1. Handicaps of conventional materials in PSC architecture and alternatives in this study.

PSC Layers	Conventional Material	Handicaps	This study
TCO Substrate	FTO/ITO glass	High cost. Indium scarcity	Flexible substrate PET/Ag/PH1000
ETL	TiO_2	Sintering at high temperatures (additional complications and cost)	Phenyl-C61-butyric acid methyl ester (PCBM)
Active Layer	$CH_3NH_3PbI_3$	Lead toxicity	$CH_3NH_3PbI_3$
HTL	Spiro-OmeTAD	Cost and lack of thermal stability	PDOT: PSS (polystyrene sulfonate)
Metal Electrode	Au/Ag	High cost	Al

Table 2. Specifications of flexible PSC device.

Active Area (A) (m^2)	75%
Performance Ratio (PR)	80%
Cell to Module Efficiency Loss	20%
Annual Solar Insolation (I) (Global Solar Atlas 2018) (kWh/m^2-yr)	1700 (kWh/m^2-yr)
Device Lifetime (LT) (year)	5 years
Flexible PSC Efficiency (η)	14%

Table 3. Inventory table of manufacturing the flexible PSC. Detailed values are showed in the supplementary document. I denotes input, P for production electricity demand.

Materials and processes	Input	Unit	Assumptions	Ref.
PET/Ag/PH1000 mesh Substrate			PET: 50 µm thickness and Density is 1.38 g/cm^3 Ag: % 3.2 Surface area with 2 µm thickness PH1000: 150 nm thickness and density is 1.011 g/cm^3	(Thompson and Woods 1955)
I Polyethylene terephthalate compound (PET)	6.9E-02	kg		
I Silver, at regional storage	0.67E-03	kg		
I Market for butyl acrylate	6.17E-03	kg	Assumed as UV resin material Density is 0.89 g/ml	(CDC 2018)
I PH1000 Production	3E-03	kg	Assumed same as PDOT: PSS	(Louwet et al. 2003)
P Electricity (Nanoimprinting, Substrate Annealing, PH1000 Spin Coating, PH1000 Annealing)	183.28	MJ	Assumed as printing deposition	(García-Valverde, Cherni, and Urbina 2010, Celik et al. 2015)
Hole Transport Layer			Thickness 35 nm Density: 1.011 g/cm^3	(Louwet et al. 2003)
I PDOT: PSS Production	0.07E-03	kg		
P Electricity (Spin Coating, Annealing)	91.31	MJ		
Perovskite Active Layer			Thickness of Active Layer is 280 nm Density of Methylammonium Lead Iodide is 1.368 g/ml	(Lin et al. 2018, Sigma-Aldrich 2018)
I PbI_2 Production	0.57E-02	kg		
I CH_3NH_3I Production	1.97E-03	kg		
P Electricity (Spin Coating, Stirring, Annealing)	270.84	MJ		(García-Valverde, Cherni, and Urbina 2010, (Celik et al. 2015)
Electron Transport Layer			60 nm thickness and 1.5 g/cm^3 density	(Sun, Han, and Liu 2013, Ueda et al. 2017)
PCBM Production	1.8E-03	kg		
Energy Input				(García-Valverde, Cherni, and Urbina 2010, Celik et al. 2015)
P Electricity (Spin Coating)	0.9	MJ		
Metal Electrode			100 nm thickness	
I Aluminum	0.54E-03	kg	Deposition efficiency is assumed 50%	
P Electricity (Evaporation, Vacuum)	166.6	MJ		

filled with Ag ink. Then, PH1000 (we assumed PDOT: PSS) is spin-coated on it. The substrate is sintered after lithography at 80°C for 10 min and annealed at 120°C for 20 min. The hole transport layer (PDOT: PSS) is coated by spin coating and annealed at 120°C for 20 min. The perovskite active layer is coated by a two-step spin-coated method (First, PbI_2 is spin-coated and dried at 70°C for 10 min, then CH_3NH_3I is coated on the PbI_2 layer and annealed at 130°C for about 2 min). 60 nm PCBM electron transport layer is spin-coated, and 100 nm Al deposition is implemented by thermal evaporation.

Results and discussion

Environmental impact assessment

ILCD (International Reference Life Cycle Data System) model has been selected to calculate environmental impacts with Gabi 8.1 software. Table 4 shows the environmental impact values of each layer of the flexible PSC required to manufacture 1 m^2 of the solar cell.

In Figure 1, the share of the total environmental impacts of each PSC layers is illustrated. It is seen in the figure that the largest impact values are derived from the perovskite active layer in all environmental impact categories. The reason for this is the long stirring process used in the manufacturing of this layer (260 MJ). The share of PET/Ag/PH1000 substrate impact values to total cell values varies between 20 and 24.7%. This range is compatible with the rate (25.6%) of electricity consumption used in the production of this layer (183.71 MJ) to the entire device (712.75 MJ). Half of the manufacturing electricity consumption of the flexible substrate is derived from the annealing process of the PH1000 layer (90.41 MJ). PET substrate annealing needs one third (60.27 MJ) of the total electricity consumption of flexible front electrode manufacturing. The electricity demand for nanoimprinting of the ultraviolet resin layer is 31.7 MJ and the spin coating of PH1000 is just 0.9 MJ.

Figure 2 shows the contribution of electricity consumption to selected environmental impact categories in this study. According to this, manufacturing electricity consumption is responsible for

Table 4. Environmental Impacts of manufacturing 1 m^2 flexible PSC.

Impact Category	PET/Ag/PH1000	PCBM	Perovskite	PDOT: PSS	Al	Total
Acidification [Mole of H+ eq.]	0.23	0.002	0.48	0.16	0.29	1.16
Global Warming Potential [kg CO2 eq.]	27.84	0.4	56.87	19.16	34.96	139.23
Ecotoxicity [CTUe]	173.26	2.29	270.41	91.06	166.07	703.09
Eutrophication [kg N eq.]	0.024	0.00022	0.049	0.016	0.029	0.119
Human toxicity cancer effects [CTUh]	1.60E-06	2.12E-08	3.17E-06	1.07E-06	1.95E-06	7.82E-06
Human toxicity non-cancer effects [CTUh]	6.98E-06	7.4E-08	1.08E-05	3.64E-06	6.63E-06	2.81E-05
Photochemical ozone formation [kg NMVOC eq.]	7.24E-02	1.68E-03	1.44E-01	4.84E-02	8.84E-02	0.35
Primary energy demand [MJ]	468.11	10.029	948.54	319.54	582.9	2329.12

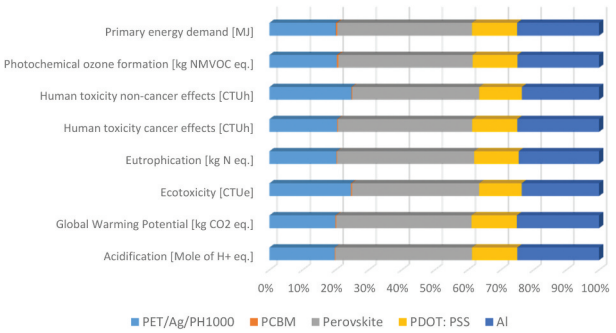

Figure 1. Share of total environmental impacts of each PSC layers.

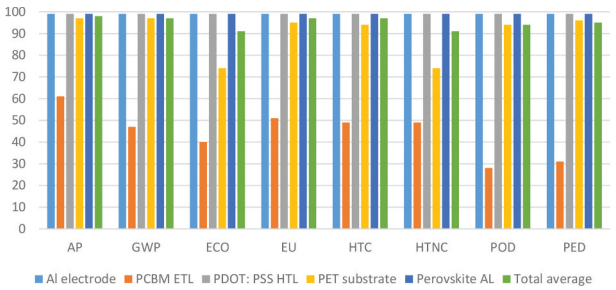

Figure 2. Contribution of electricity consumption to every impact category.

more than 90% of environmental impacts for all layers except PCBM ETL. Cumene (assumed instead of 1,2,4-Trimethylbenzene) is another major contributor to environmental impact in this layer. Considering that the PCBM layer is responsible for less than 1% of total environmental impacts, the significance of this substitution is negligible.

Energy payback time (EPBT) and global warming potential (GWP)

EPBT indicates when a system has recovered the energy it consumes during its life cycle. EPBT is calculated as the ratio of life cycle total primary energy use (kWh) to over annual power generation (kWh/year). Primary energy demand for device manufacturing has been used to calculate EPBT (see Eq. (2)) (Collier and Ellingson 2015). ε is a factor (0.35 for PVs) that converts electricity production value to primary energy demand.

$$EBPT = \frac{Primary\ energy\ demand}{(IxηxAxPR)/\varepsilon}$$

A comparison of the EPBT value for the flexible PSC studied here with these commercial PV technologies is shown in Figure 3. As can be seen in the figure, the flexible PSC device of our study has lower EPBT results than poly-silicon and mono-silicon PVs but higher than CdTe PV.

The life cycle global warming potential (GWP) of PV cells and systems indicates their potential to mitigate climate change and is by far the most widely used environmental indicator. GWP is calculated as the ratio of life cycle greenhouse gas emissions (g CO_2-eq) to lifetime power generation (kWh). Therefore, we applied the second functional unit to convert impacts from 1 m^2 cell area ($Impact_{m^2}$) to per 1kWh ($Impact_{kWh}$) by using equation 1 (Celik et al. 2015).

$$Impact_{kWh} = \frac{Impact_{m^2}}{IxηxAxPRxLT}$$

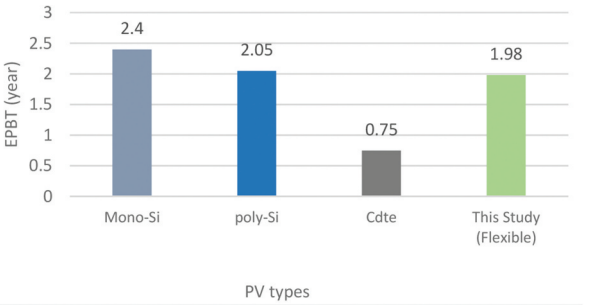

Figure 3. Energy payback time comparison of flexible PSC with commercial PVs (Peng, Lu, and Yang 2013).

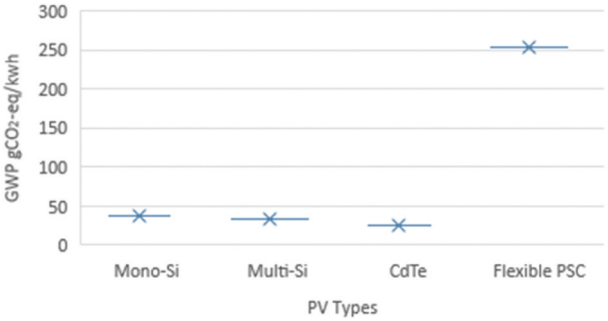

Figure 4. Comparison of the flexible PSC with commercial PVs (Peng, Lu, and Yang 2013) with regard to GWP values.

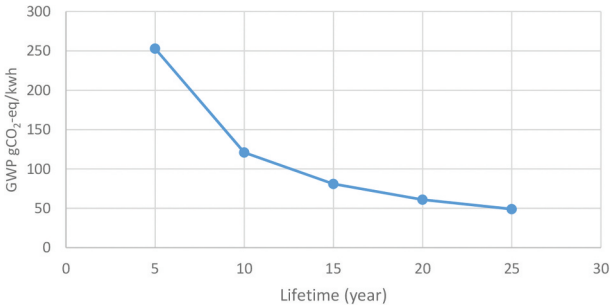

Figure 5. Sensitivity analysis of GWP of Flexible PSC.

As a result of the calculation, we found that the GWP value of flexible PSC is 243 g CO_2-eq/kWh. It is reported that the GWP of commercial PVs ranging from 29 g to 50 g CO_2-eq per kWh (Peng, Lu, and Yang 2013). Figure 4 demonstrates that flexible PSC has a much higher GWP value than commercial PVs.

A sensitivity analysis has been applied to specify what lifetime the flexible PSC requires to attain a competing GWP on commercial PVs. Considering that GWP values of commercial PVs are around 50 g CO_2-eq per kWh, the flexible PSC needs 25 years to get the same GWP value (Figure 5). This is in line with the lifetime values assumed for commercial PV cells and systems.

Conclusion

Our life cycle assessment of the flexible PSC shows that the greater part of impacts derived from perovskite active layer deposition in all environmental impact categories. The stirring process used in the manufacture of the perovskite active layer is the driver behind the high impact values. The solution-based manufacturing process was used in this study instead of the vacuum evaporation process, which would lead to even higher electricity consumption used in the manufacturing of the active layer. Nevertheless, the stirring process prevented the achievement of lower electricity consumption.

Our study showed that over 90% of the environmental impact values are caused by electricity consumption. The electricity consumption for the production of the flexible front electrode device is calculated as 183.71 MJ, which is much lower than conventional FTO glass (1735.2 MJ) and ITO glass (511.2 MJ) layer productions (Espinosa et al. 2015). This result reveals that the flexible PET/Ag/PH1000 structure used as the front electrode not only provides cheap and easy production but also reduces electricity consumption and therefore environmental impact values.

Our assessment demonstrates that the studied flexible PSC device has a competitive EPBT value in comparison with commercial PVs. However, the operational lifetime of the flexible PSC device still

needs significant improvement to achieve global warming potentials per kWh of electricity generation that are competitive with existing commercial solar cells.

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Notes on contributors

Huseyin Sarialtin completed his M.S. degree at UC Riverside. Following his degree, he pursued in depth research experience, working as a researcher in Izmir Institute of Technology and assessing photovoltaic technologies on a European Union project in Augsburg, Germany. He worked at the Bren School at UC Santa Barbara as a visiting researcher applying sustainable energy analysis on next-generation photovoltaics. He earned his PhD degree titledLife Cycle Assessment of Perovskite Solar Cells in 2019.

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