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Relative impacts of methylammonium lead triiodide perovskite solar cells based on life cycle assessment

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ABSTRACT

The environmental performance of four different device assembly procedures based on hybrid halide perovskite solar cell (PSC) were assessed from cradle to grave using life cycle assessment (LCA) methodology. In addition, a new environmental indicator was defined to measure the time evolution of an impact category, specifically in this case, human toxicity cancer payback time. PSCs procedures accounted for the probably three more used basic recipes for laboratory perovskite deposition: 1) spin coating of stoichiometric precursor solution, 2) spin coating of precursor solution using lead chloride precursor and 3) the two step deposition method. Also, the two most widely used substrate configurations (planar and mesoporous substrate)were considered. LCA included three realistic scenarios for the end of life: 1) residual landfill, 2) reuse and residual landfill and 3) reuse and recycling. The remaining variable parameters to assemble the device were fixed in common for all four devices, which were the major responsible of the whole PSC impact. Lead of PSCs had no significant contribution in environmental impacts. Beyond shared procedure steps, impacts generated by the two-step method and the use of mesostructured type substrate were higher. End of life scenario with reuse and recycling improved the toxicity impact categories.

1. Introduction

Perovskite solar cells (PSCs) have emerged as a very efficient type of solar cells for the last few years. As yet, they have shown efficiencies (η) over 20% on thin film cells [1,2]. Besides, it is expected they reach as high efficiencies as first-generation (25.3% for a single crystal Si non-concentrator cell) and second-generation (22.6% for a CIGS cell) solar cells do, even overstepping their efficiencies in a nearby future [3]. Owing to its versatility and the possibility of tailoring its energy band gap, perovskite also has a high potential to be combined with other materials to form a tandem device, thus reaching higher efficiencies [4]. For instance, an efficiency of 27% of a tandem of perovskite combined with Si was reported [5]. However, for a final implantation of this technology, demonstration of long term stability will be needed. Moreover, a technical and economical assessment of PSCs states that there are some limitations when manufacturing them at large scale [6]. Although stability should still be proven over a broad range of conditions [3], promising results have already been delivered [7].

One of the main concerns of PSCs is that the hybrid organic-inorganic perovskite most commonly used (with general formula $MAPbX_3$, where MA = methylammonium and X = I, Br) contains significant quantities of lead. Pb is a toxic substance whose intake in the human body causes damage through mimicry of essential ions such as Ca, Zn and Fe [8,9]. Furthermore, its use is restricted by the European Restriction of Hazardous Substances Directive [10]. Encapsulation of PSCs, to impede contact with moisture and oxygen to improve stability [11], could be a solution for the toxicity issue in order to contain possible Pb leachates. Besides the toxicity of this element, the Pb extraction consists of a high-temperature process over 1400 °C, which generates greenhouse gases and dangerous fumes as by-products [12]. Consequently, further solutions should be found in order to decrease the environmental impacts of the preparation of PSCs [13,14].

Abbreviations: EOL, end of life; EPBT, energy payback time; HTC, human toxicity, cancer; HTN-C, human toxicity, non-cancer; ET, Ecotoxicity; WDP, water depletion potential; CED, cumulative energy demand; GWP, global warming potential; S1, scenario 1; S2, scenario 2; S3, scenario 3; HTCPBT, human toxicity cancer payback time; HTCPBT20, human toxicity payback time supposing a 20-year lifetime

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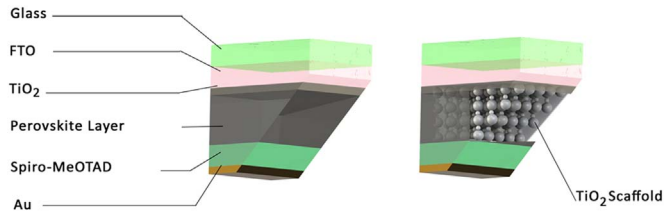

Fig. 1. Perovskite solar cells layers: 1) thin film planar perovskite layer and 2)embedded perovskite in the mesoporous layer.

Solar cells employing hybrid halide perovskite as light harvester material are mainly composed either a) by a thin film perovskite layer, known as planar configuration or b) by the perovskite deposited onto a mesoporous scaffold. The light harvesting layer is sandwiched between a hole transporting material (HTM)and an electron transporting material layer (ETM), see Fig. 1. Once the charges are photo-generated in the perovskite, the ETM separates selectively the electrons to the front contact, and the HTM layer transports the holes to the back contact. Both layers are important to ensure a high performance of the cell, although different architectures are possible [15]. The most currently used material for the HTM is the Spiro-MeOTAD, initially synthesized for incorporating in multilayer light-emitting diodes (LED) [16,17], later it was used in solid state dye sensitized solar cell as HTM [18]. A compact layer of TiO_2 is widely used as ETM which is formed in most of the cases through the hydrolysis of the titanium isopropoxide [19]. In PSCs, the front contact or light side is generally the transparent conductive oxide SnO_2:F (FTO), because of its high transparency in the visible region and its low resistivity at room temperature of the order of 1 Ω cm [20]. Meanwhile, the back contact extracting contact may be made of silver [21], gold [22] or aluminum [23], among others.

In order to lead the manufacture of this promising technology of PSCs to a more sustainable state, as it is still under development at lab-scale, life cycle assessment (LCA) methodology should be applied. In this regard, few works have been done thus far [6,8,14,24–29]. Amongst them, special emphasis should be given to the work of Espinosa et al. [24], in which two important deposition methods (spin-coating and vapor-deposition) were compared. With each deposition method, a different architecture (normal and inverted) was produced and assessed from cradle to gate. Gong et al. [30] compared two PSCs with different ETM (TiO_2 scaffold vs ZnO thin film), back contact (gold vs silver) and front contact (fluorine doped tin oxide vs indium tin oxide). This study was the first to include the disposal stage into its system boundary. Zhang et al. [31] evaluated a PSC based on titanium dioxide nanotubes through LCA methodology with data obtained from laboratory-scale. Alternatively, Celik et al. [26] evaluated a comparison of co-evaporation and spray perovskite deposition methods, which are more amenable to manufacturing, rather than laboratory specific deposition methods dipping and spinning. Recently, a perovskite/Si was assessed through LCA from cradle to grave [28] contrasting several combinations of materials for the back electrode (Au, Ag and Al) and HTM (Spiro-MeOTAD vs PEDOT:PSS). Finally, five different perovskites were compared using Cs, formamidinium (FA), and MA for the monovalent cationic position; Pb and Sn for the cationic position; and combinations of I^-, Br^- and Cl^- for the anionic position [29].

The aim of our work was to conduct a comprehensive LCA of four different devices of PSCs from cradle to grave [32,33], selecting likely the most broadly considered. Although the best efficiencies have been reached using perovskites with mixtures of organic cations (MA^+ and FA^+) and halides (I^- and Br^-) [34,35], for the sake of clarity and simplicity, we just considered the most extended $CH_3NH_3PbI_3$ halide perovskite as light absorbing material and three basic recipes of perovskite deposition: 1) spin coating of stoichiometric precursor solution of $PbCl_2$ and methylammonium iodide (MAI) in 1:3 M ratio, that we call Device 1 hereafter [36]; 2) spin coating of precursor solution of PbI_2 and MAI, Device 2 hereafter [37]; and 3) the two step method deposition, which implies the dipping of a spin coated PbI_2 film into a MAI solution (Device 3 hereafter) [38]. Also, for the preparation method of spin coating of precursor solution of PbI_2 and MAI, the two most widely used substrate types (planar and with mesoporous TiO_2 scaffold) were considered, the device with mesoporous substrate is called Device 4 [39]. The remaining variables parameters, as substrate, contacts, ETM and HTM, to assemble the device were fixed in common for all four types of devices, and correspond to the most commonly used in PSCs field, see Table 1. Due to the unreliable nature of the amount of electricity consumed in the laboratory environment for a real industrial scenario, an uncertainty analysis was performed for the most energy consuming processes.

Table 1
Characteristics of the different PSCs analyzed.

Specific layers		
Device	Perovskite deposition method	Configuration
Device 1	Spin-coating 3:1	Planar
Device 2	Spin-coating 1:1	Planar
Device 3	Spin-coating + dipping	Planar
Device 4	Spin-coating 1:1	Mesoporous
Common layers		
Layer name	Material	Deposition method
Substrate + Front contact	Glass/FTO	Sputtering
ETM	TiO_2	Spin-coating
HTM	Spiro-MeOTAD	Spin-coating
Back contact	Gold	Thermal evaporation
End of life scenarios		
Scenario	Number of uses	EOL treatment
Scenario 1	1	Landfill
Scenario 2	10	Landfill
Scenario 3	10	Recycling

For the first time, we dipped into three possible scenarios of recycling of the PSCs, which could significantly improve their lifetime, see Table 1. In scenario 1, PSCs was inertized and deposited in a residual landfill. The other two scenarios were aimed by recent researches about the potential regeneration of PSCs [40,41] and they two differ in their potential treatment at the end of the last regenerative cycle: land filling in scenario 2 or recycling in scenario 3.

By means of the power conversion efficiencies (PCE) provided in the bibliography relative to each perovskite solar cell preparation studied [36–39], the lifetime at which each PSC produces just as much energy as necessary to manufacture it (in laboratory environment) was determined. Further analyses were performed considering that all four devices were prepared with an efficiency of 20% with different scenarios of end of life. Finally, a similar assessment was made to compare the payback time of the human toxicity impact category on the four PSC devices analyzed during their potential lifetime. This latter human toxicity cancer payback time analysis was contrasted with a similar analysis for established photovoltaic technologies.

2. Materials and methods

2.1. Goal and scope definition

This study was intended to lay on the table the environmental aspects of the most promising ways through which lead halide perovskite solar cells can be conducted. Four different perovskite based devices were considered, see Table 1. The four different analyzed devices presented different light harvesting layer but the other parts of the cell were common. We considered a glass with FTO deposited via sputtering. On FTO, a compact layer of TiO_2, deposited via spin-coating worked as ETM. The next layer was the perovskite, prepared following three different procedures, and considering both planar and mesoporous TiO_2 scaffold configurations, see Table 1. Spin-coated Spiro-