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Contents

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Summary

The present deliverable aims to report the status of the review paper to be written within GOTSolar project and that should contain the major project achievements beyond state-of-the-art. This work might contribute for updating the scientific community in what concerns relevant breakthroughs both on perovskite solar cells fundamental and technological knowledge. The review will be entitled “New advancements on perovskite solar cells” and it will have the contribution of all partners. The methodology to prepare this work by the consortium is described and the sections that are already written are presented. Finally, a list of possible journals for submission are listed. The deadline to submit this work is by the end of February.

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Table of Contents and Partners Assignment

1. Abstract - UPorto
2. Introduction – UPorto
3. Fundamentals – CNRS and EPFL
4. Materials and performances – EPFL, IChF PAN and UUlm
5. Engineering solutions – UPorto and GCS
6. Devices and quality control – GCS and Efacec
7. Economical assessment - GCS

Methodology

The title, table of contents and each partner assignment was defined in the 5th face-to-face meeting with the inputs of all partners. Then, UPorto started the Introduction section to deliver to all partners as the first draft and requested the contribution of the other GOTSolar partners as distributed above. In order to include all project main results as well as maintain the partners focus on the project main objectives, the deadline for sending the contributions to UPorto was defined to be beginning of February. This way, it is presented next only the section *Introduction* that was defined to be as the kick-off the review.

Introduction

Hybrid organic-inorganic perovskite materials have become one of the most studied class of light-harvesting materials due to their exceptional properties such as high light absorption, long carrier diffusion lengths, flexible bandgap tuning and defect tolerance. Besides, these materials can be processed from solution leading to photovoltaic devices that can be fabricated at room temperature with performance levels very similar to industry giant polycrystalline silicon. This makes perovskite solar cells (PSCs) appear particularly promising for next-generation solar devices owing to their high-performance and low-cost. In the NREL efficiencies plot the most efficient certified cell presents 23.3 %, however the configuration and the detailed materials used remain unknown [1].

Perovskites are ABX_3 structures, where for photovoltaic applications A is normally rubidium (Rb), cesium (Cs), methylammonium (MA), and/or formamidinium (FA); B is tin (Sn) and/or lead (Pb); and X is chlorine (Cl), bromine (Br), and/or iodine (I). Organic-inorganic perovskites were first studied by Weber in 1978 [2, 3] but only in 2006 this material was used as photovoltaic absorber by Miyasaka and co-workers [4]. These authors were apparently attracted by the self-organization potential of perovskite in the nanoporous TiO_2 layer of dye-sensitized cells, reporting power conversion efficiencies (PCE) of 2.2 % for devices prepared with methylammonium lead bromide ($MAPbBr_3$) and lithium halide/halogen as redox couple and a Pt-coated FTO glass as counter-electrode. Three years later, replacing bromine by iodine in the perovskite

structure, they were able to increase the efficiency up to 3.8 % [5]. This power conversion efficiency was further enhanced to 6.5 % by Park et al. [6] in 2011 using thinner TiO_2 films and optimizing the perovskite coating solution concentration, post-annealing conditions and TiO_2 surface modification with $Pb(NO_3)_2$. The liquid electrolyte was identified as the limiting component since the perovskite material was dissolved in the electrolyte, resulting in a rapid performance degradation. One year later, the same authors demonstrated that the use of a solid hole conductor such as spiro-MeOTAD (2,2',7,7'-tetrakis(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene) makes the device stability to dramatically improve compared to $MAPbI_3$ with an electrolyte junction. However, due to transport limitations, the typical photoelectrode thicknesses used in dye-sensitized solar cells, 10 – 20 µm, was greatly decreased to thicknesses below 1 µm. Both developments not only improved the stability but they also boosted the reported energy conversion efficiency to 9.7 % [7].

During mid-2012 and 2013, it was a very profitable period for perovskite research field since important advances were made in the comprehension of perovskite-based devices and on reported energy conversion efficiency. First, it was reported the use of mixed-halide perovskites as $MAPbI_{3-x}Cl_x$, which exhibited higher stability and enhanced carrier transport than its pure iodine analogue [8]. Then, the nanoporous TiO_2 conductive scaffold was substituted by a non-conductive Al_2O_3 similar structure, coupled with the mentioned mixed iodine/bromine perovskite; these devices delivered more than 10.9 % power conversion efficiency under full solar illumination and an impressive open-circuit voltage (V_{oc}) of more than 1.1 V, despite the relatively narrow absorber band gap of 1.55 eV [8]. This work demonstrated the wide potential of perovskite material to transport efficiently both holes and electrons. Third, a new device configuration was proposed using a nanoporous TiO_2 thin layer impregnated with the perovskite absorber (ETA – extremely thin absorber); these devices exhibited power conversion efficiencies near to 8 % [8]. In mid-2013, Grätzel's group used TiO_2 scaffolds and two-step iodine deposition for improving the perovskite morphology and reported a certified efficiency of 14.1 % [9]. Finally, as a consequence of the perovskite ambipolar transport ability, it was demonstrated that the TiO_2 scaffold was not essential to reach high energy conversion efficiencies: by that time, a simple planar device could display solar-to-electrical efficiencies of 15.4 % [10], what was proved later by the present record efficiency of 20.7 % already reached for planar devices [11]. In 2014, it was introduced the anti-solvent crystallization method where a solvent is poured onto the perovskite solution precursor during spin coating, causing the perovskite crystal to form and precipitate into a smooth and compact film. Seok et al. [12] showed that when the perovskite was deposited using a mixture of γ-butyrolactone (GBL) and dimethylsulphoxide (DMSO), followed by a toluene drip while spinning, extremely uniform and dense layers were formed. This solvent-engineering technology enabled a fully solution-processed perovskite solar cell with a certified 16.2 % power-conversion efficiency under standard reporting conditions. Another jump in the reported efficiency occurred by the hand of the same group, who further improved the power conversion efficiency reporting a certified value of 17.9 % [13]. TiO_2 yttrium doping electron extraction layer was reported to improve the electron transport through the scaffold and modified ITO electrode work-function, resulting in overall power conversion efficiency of 19.3 % [14]. In 2015 Seok et al. [15] reported 20.1 % of power conversion efficiency with high-quality formamidinium lead iodide ($FAPbI_3$) perovskite films without residual PbI_2. A contact-passivation strategy, using chlorine-capped TiO_2 colloidal nanocrystal film, mitigats interfacial recombination and improve interface binding in low-temperature planar solar cells. This approach allowed

cells that retained 90 % of their initial performance after 500 hours of continuous room-temperature operation at maximum power point under 1-sun illumination [16]. Still using formamidinium with multiple cations and mixed halide anions, Seok et al [17] showed that the introduction of additional iodine ions into the organic cation solution decreases the concentration of deep-level defects and is essential for realizing thin-film PSCs exceeding 21 %; indeed, a certified power conversion efficiency of 22.1% was reached. So, using perovskites with mixed cations and halides has become a trend since pure perovskite compounds as the primarily used $MAPbX_3$, $FAPbX_3$ and $CsPbX_3$ (X = Br or I) present several limitations, like structural phase transition at low-temperatures, degradation upon contact with moisture, thermal stability, light-induced trap-state formation or halide segregation in the case of "mixed halide" perovskites [18-20]. Following this strategy, Saliba et al. [21] reported high efficiency perovskite solar cells with a stable PCE of 21.1% using a mixture of three cations, Cs/MA/FA. This cation combination suppresses yellow phase impurities (namely PbI_2) and generates highly uniform perovskite grains besides allowing more stable and reproducible devices. The same research group also showed that rubidium cation can be embedded into a "cation cascade Rb/Cs/MA/FA" to create perovskite materials with stabilized efficiencies of up to 21.6 % and an open-circuit voltage of 1.24 V for a band gap of 1.63 eV [22]. Furthermore, these cells retain 95 % of their initial performance at 85 °C for 500 h under full solar illumination and at the maximum power point. In 2017, Graetzel's group reported a device displaying a remarkable PCE value, operational and thermal stabilities, based on all-inorganic charge extraction layers (mesoporous TiO_2 as electron extraction layer and CuSCN as hole transport layer) [23]; devices with 20.2 %, retaining more than 95 % of their initial efficiency after aging at a maximum power point for 1000 hours at 60 ºC, were reported.

A loss of efficiency is inevitable when size scale changes, namely from small-area cells (~0.1 cm^2) to large-area cells (~1 cm^2) and modules (> 10 cm^2). In individual cells this loss results mainly from higher series resistances and lower shunt resistances (due to non-uniform coatings), but when speaking about modules it has to be considered also the unavoidable dead areas of bus bars and interconnections. Several works have been done to develop scalable deposition techniques, enabling uniform perovskite films free of pinholes [24-27]. Nevertheless, the majority of reported works is still focused in the validation of these techniques at small scale, for allowing comparison with perovskite films prepared by spin-coating, and no information is given for areas bigger than 1 cm^2.

In 2014 Di Carlo et al. [28] produced the first solid state modules based on organometal halide perovskite $MAPbI_{3-x}Cl_x$ using Spiro-OMeTAD and poly-(3-hexylthiophene) (P3HT) as hole transport materials, reporting a PCE of 5.1 % for an active area of 16.8 cm^2. Each module was composed by 5 series-connected solar cells, each individual cell with area of 3.36 cm^2 and 6.3 % efficiency. Seo et al. reported a p–i–n perovskite planar device based on a flat and thick $MAPbI_3$ film, a thin PCBM film and a thin LiF layer (~0.5 nm) as a buffer layer, prior to deposition of the Al electrode.

Through solution process at low temperature, 10 cells were serially connected into a module with an overall active area of 60 cm^2 (actual size 10 × 10 cm^2) showing 8.7 % PCE [29]. Then, it was described an air-assisted blade coating process of PbI_2 layers for facilitating the fabrication scale up of perovskite modules. A 100 cm^2 module composed by 9 cells series-connected and made of $MAPbI_3$ perovskite and P3HT hole transport material delivered a V_{OC} of 9.6 V and an efficiency of 4.3 % with an aperture ratio of

73 % [30]. In 2015 Heo et al. proposed inverted ITO/PEDOT:PSS/$MAPbI_3$/PCBM/Au planar hybrid solar cells for supressing hysteresis by preparing dense pinhole-free $MAPbI_3$ perovskite on a PEDOT:PSS/ITO substrate via a single-step spin-coating of $MAPbI_3$ solution with controlled solubility. They succeeded in assembling a 10 × 10 cm^2 perovskite module by serially connecting 10 cells; this module with an active area of 40 cm^2 exhibited a power conversion efficiency of 12.9 % and delivered 10.1 V under full sun conditions [31]. Later in 2015, it was fabricated modules containing $MAPbI_{3-x}Cl_x$ perovskite active layer impregnated into a scaffold of TiO_2 nanorods (NR) with active area of 10.8 cm^2, presenting an efficiency of 10.5 %; besides, this configuration had better stability than TiO_2 nanoparticles analogue due to the macroporous nature of the nanorods [32]. In the same month, Nano-electronics Research Center Imec, part of the Solliance thin-film PV research organization, reached 11.3 % with a 16 cm^2 aperture area perovskite module and a geometrical fill factor of more than 95 %[33]. Again Imec and Solliance announced on may 2016 the first semi-transparent perovskite PV-module of 16 cm^2 with a power conversion efficiency up to 12 %. This technology is particularly interesting for semi-transparent PV-windows to be integrated into Zero-Energy Buildings. Moreover, these semi-transparent perovskite modules were coupled to 4 interdigitated back contacted (IBC) silicon solar cells in a tandem arrangement and an unprecedented 17.2 % power conversion efficiency was achieved for larger areas of up to 16 cm^2 [34]. Company Solaronix from Switzerland presented a PSC prototype of 500 cm^2 based on printing techniques, thus avoiding the expensive methods such as chemical vapor deposition or metal evaporation. The assembled solar modules reached 12 % solar-to-power efficiency and 5000 h continuous illumination stability [35]. In the same year, a research group in Shanghai Jiao Tong University (SJTU) obtained an energy conversion efficiency of 12.07 % on a perovskite module with an active area of 36 cm^2, certified by the National Institute of Advanced Industrial Science and Technology (AIST) [36]. Heo et al. reported another perovskite module with an active area of 40 cm^2 delivering 10.5 V open circuit voltage, 84.15 mA short circuit current and 15.5 % power conversion efficiency under 1 sun conditions. This power conversion efficiency was a remarkable value obtained by controlling the re-dissolution and crystal grain growth of the $MAPbI_{3-x}Cl_x$ mixed halide perovskite film via spray coating [37]. Still in 2016, monolithic perovskite modules with active areas of 31 cm^2 and 70 cm^2 were fabricated using scalable printing processes, which allowed power conversion efficiencies of 10.46 % and 10.74 %, respectively. These power conversion efficiencies values were attributed to the very good electrical quality of the mesoscopic hole transporter and its efficient infiltration [38]. Moreover, very good stability at ambient environment was obtained with only 5 % reduction in efficiency after 2000 h. In 2017, a fully printed 10 × 10 cm^2 $(5\text{-}AVA)_x(MA)_{1-x}PbI_3$-based mesoscopic solar cell with a double layer of mesoporous TiO_2 and ZrO_2 covered by a porous carbon film, exhibited an average PCE value of 10.3 % [39]. Yang et al. demonstrated a 12.6 cm^2 four-cell module (88 % geometric fill factor) with 13.3 % stabilized active-area efficiency output using a $MAPbI_{3-x}Cl_x$ precursor formulation along with solvent tuning to enable a wide precursor-processing window and a rapid grain growth rate. This ink was deposited by blade-coating [25]. Another deposition route of $MAPbI_3$ perovskite films was proposed later on by Grätzel et al. [40], relying on the rapid conversion of amine complex precursors to perovskite films, followed by a pressure step. This deposition approach presents several advantages as it can be performed in air at low temperatures, facilitating fabrication of large-area perovskite devices. A certified power conversion efficiency of 12.1 % with an aperture area of 36.1 cm^2 was reached for a mesoporous TiO_2-based perovskite solar module architecture.

A record-breaking module was prepared in May 2017, composed by 6 series-connected cells with an aperture area of 16.29 cm^2 and a certified PCE of 16.0 % belonging to Microquanta; this value was certified by the National Renewable Energy laboratory in United States [41]. The major breakthroughs in 2017 continued with Imec and Solliance team by improving their previous glass-based modules of 16 cm^2 to 12.4 % PCE and, last April, further to 13.8 % for even larger modules of 144 cm^2 aperture area [42, 43]. Some prototypes in flexible substrates were also presented by Toshiba and Solliance based on roll-to-roll (R2R) production processes. Toshiba prepared a 25 cm^2 module with 10.5% [44] and Solliance and Greatcell Solar a 160 cm^2 with 10.2 % efficiency [45]. Last July, Microquanta Semiconductor has announced a new world record of for perovskite solar modules of 7-series connected cells and exhibiting 17.3 % conversion efficiency. This result was certified by the international test center Newport Corporation [42]. Table 1 summarizes the most relevant perovskite prototypes fabricated so far with relevant details concerning modules configuration.

From the recent history of perovskites, it is evident that this technology is marching ahead towards commercialization. In this review work, it will be provided the most recent breakthroughs in terms of materials and configurations, as well as processing techniques for upscaling and appropriate device characterization.

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Device architecture	Area cm^2	PCE %	Module details	Year	Ref.
FTO/c-TiO_2/m-TiO_2/$MAPbI_{3-x}Cl_x$/Spiro-OMeTAD/Au	16.8 (a)	5.08	Glass substrate; 5 series-connected cells of 3.36 cm^2; 25 cm^2 aperture area.	2014	[28]
FTO/c-TiO_2/m-TiO_2/$MAPbI_{3-x}Cl_x$/P3HT/Au		5.10			
ITO/PEDOT:PSS/$MAPbI_3$/PCBM/LiF/Al	60 (a)	8.7	Glass substrate; 10 series-connected cells; 10×10 cm^2 total area.	2014	[29]
FTO/c-TiO_2/m-TiO_2/$MAPbI_3$/Spiro-OMeTAD/Au	10.1 (a)	10.4	Glass substrate; 4 series-connected cells of 2.5 cm^2; spin-coating.	2014	[30]
FTO/c-TiO_2/m-TiO_2/$MAPbI_3$/P3HT/Au	100 (t)	4.3	Glass substrate; 9 series-connected cells; blade-coating; 73 % aperture ratio.		
ITO/PEDOT:PSS/$MAPbI_3$/PCBM/Au	40 (a)	12.9	Glass substrate; 10 series-connected cells; 10×10 cm^2 total area.	2015	[31]
FTO/c-TiO_2/NR-TiO_2/$MAPbI_{3-x}Cl_x$/Spiro-OMeTAD/Au	10.8 (a)	10.5	Glass substrate; 4 series-connected cells of 2.6 cm2 each	2015	[32]
-	16 (ap)	11.3	Glass substrate; 95 % geometrical FF 95; spin coating[1].	2015	[33]
-	16[2]	12	Glass substrate; semi-transparent.	2016	[34]
-		17.2	Glass substrate; Tandem perovskite/silicon solar cells.		
-	500 (t)	12	Glass substrate; fully-printed.	2016	[35]
-	36 (a)	12.07	Glass substrate.	2016	[36]

[1] Using blade coating PCE is 9 %

[2] Not specified if it is active, aperture or total area

FTO/TiO_2/$MAPbI_{3-x}Cl_x$/PTAA/Au	40 (a)	15.5	Glass substrate; 10 series-connected cells; 10×10 cm^2 total area; 40 % geometrical FF; spray coating.	2016	[37]
FTO/c-TiO_2/m-TiO_2/Perovskite/ZrO_2/Carbon	31 (a)	10.46	Glass substrate; fully-printable; drop casted perovskite; 10×5 cm^2 total area.	2016	[38]
	70 (a)	10.74	Glass substrate; fully-printable; drop casted perovskite; 10×10 cm^2 total area.		
FTO/c-TiO_2/m-TiO_2/$(5\text{-AVA})_x(MA)_{1-x}PbI_3$/$ZrO_2$/Carbon	49 (a)	10.4		2017	[39]
FTO/c-TiO_2/$MAPbI_{3-x}Cl_x$/Spiro-OMeTAD/Ag	12.6 (t)	13.3	Glass substrate; 4-monolithically interconnected cells; 88 % geometric FF; blade coating.	2017	[25]
FTO/c-TiO_2/m-TiO_2/$MAPbI_3$/Spiro-OMeTAD/Au	36.1 (ap)	12.1	Glass substrate; 10 series-connected cells of 3.6 cm^2.	2017	[40]
-	16.29 (ap)	16	Glass substrate; 10 series-connected cells.	2017	[41]
-	14.4 (a)	12.4	Glass substrate; 4 ×4 cm^2 total area.	2017	[42]
-	25 (t)	10.5	Flexible Substrate (R2R).	2017	[44]
-	160 (ap)	10.1		2017	[45]
-	17.277 (a)	17.3	Glass substrate; 7 series-connected cells.	2018	[42]

Abbreviations: (ap), aperture area; (a), active area; (t), total area.

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List of journals to be submitted

1. Nature reviews chemistry https://www.nature.com/natrevchem/about
2. Nature reviews Materials https://www.nature.com/natrevmats/
3. Nature energy https://www.nature.com/nenergy/
4. Renewable & Sustainable Energy Reviews https://www.journals.elsevier.com/renewable-and-sustainable-energy-reviews
5. ACS Sustainable Chemistry and Engineering https://pubs.acs.org/journal/ascecg

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