Upscaling of Carbon-Based Perovskite Solar Module

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Perovskite solar cells (PSCs) and modules are driving the energy revolution in the coming photovoltaic field. PSCs reached efficiency close to the silicon photovoltaic technology by adopting low-cost solution processes. Despite this, the noble metal (such as gold and silver) used in PSCs as a counter electrode made these devices costly in terms of energy, CO_2 footprint, and materials. Carbon-based perovskite solar cells (C-PSCs) and modules use graphite/carbon-black-based material as the counter electrode. The formulation of low-cost carbon-based inks and pastes makes them suitable for large area coating techniques and hence a solid technology for imminent industrialization.

perovskite solar cells upscaling carbon counter electrode

1. Introduction

In the last decade, perovskite has earned a lot of attention in solar technology as a photoactive material due to its high carrier mobility, ambipolar transport properties, and bandgap tunability [1][2][3]. Since its discovery in 2009 [3], the perovskite solar cell (PSC) technology has reached a power conversion efficiency of 25.7% for the single junction and 31.25% in tandem with silicon [4][5]. The excellent optoelectronic and light-absorbing properties along with easy and low-cost fabrication are leading this technology to be industrialized and able to supplant the well-established silicon photovoltaics [1].

PSCs are made on top of a glass/plastic substrate coated with a Transparent Conductive Oxide (TCO) that works as a front electrode. The absorber is sandwiched between p-type (HTM, Hole Transporting Material) and n-type (or ETM, Electron Transporting Material) semiconductors to improve the charge extraction from perovskite. A top metal electrode (Au, Ag, Cu) completes the device. Perovskite devices could adopt direct (n-i-p) or inverted (p-i-n) configuration. n-i-p or p-i-n junctions are equally used, and which is the most efficient and stable configuration is still under debate in the scientific community [6][7][8].

2. Materials and Methods

2.1. Carbon Electrodes

In PSCs, carbon-based electrodes demonstrated the ability to extract photogenerated holes from perovskite by themselves, opening the opportunity for researchers to explore HTL-free monolithic perovskite solar cells [9][10][11].

The control of graphite and carbon amounts inside precursor paste provides the optimal electrical properties according to the final device. Depending on which binder is used to realize the paste, a carbon electrode could be obtained by high-temperature (500 °C) or low-temperature curing (generally ≤ 120 °C) [12][13].

2.1.1. High-Temperature Carbon Electrodes (HTCEs)

High-temperature carbon electrodes (HTCEs) are the first ones used in perovskite photovoltaics [14]. The core of the cell architecture is based on a fully mesoscopic structure made of three highly porous layers, called the triple-mesoscopic configuration. After the deposition of the anatase mp-TiO_2 layer, a porous ZrO_2 or Al_2O_3 spacer insulator layer avoids recombination and shunts with respect to the porous carbon counter electrode. All porous layers need high-temperature curing, about 400–500 °C. Perovskite is added by percolation through the carbon electrode by different coating techniques [14][15][16][17]. The easy fabrication of fully-printable triple-mesoscopic carbon-based (TiO_2/ZrO_2 or Al_2O_3/carbon) perovskite solar cells has gained widespread attention due to the exceptional stability and strong upscaling potential of such devices [14][15][18]. The first application of the triple-mesoscopic device was reported in 2013 with a power conversion efficiency (PCE) of 6.6% [19]. Since then, substantial improvements in performance have been made to generate a PCE above 17% [20][21]. Despite their good qualities, triple-mesoscopic carbon perovskite solar cells show different issues, such as the complete removal of solvent from perovskite percolated into the mesoscopic structure, the thickness of spacing layers, the poor filling of perovskite through a very thick carbon film (20–40 µm), and the photo-absorber morphology [22][23][24]. Moreover, the absence of any selective contact for photogenerated holes causes recombination pathways at the interface with the perovskite. In this context, Raptis et al. implemented metallic grids inside the carbon layer for better conductivity [25]. The stack utilized is TCO/TiO_2/ZrO_2/carbon where 25 µm of copper grids were applied between two layers of carbon. On the same stack configuration, Jiang et al. worked on a precursor solution concentration, demonstrating how a systematic study on perovskite precursor solutions led to an efficiency above 16% [20]. Liu et al. found critical parameters in the layer's thickness that can improve the efficiencies and avoid poor filling issues of the full printable stack [23]. Liu et al. added a selective mesoporous layer of NiO_x as an HTL in a FTO/TiO_2/Al_2O_3/NiO_x/carbon device configuration. NiO_x accelerates the extraction of photogenerated holes and improves photovoltaic performance up to 17.02% [21]. In the literature, there are many approaches to solve the efficiency gap with respect to gold-based devices. The main strategies are focused on the charge extraction through hole Fermi level shift, the increase in carbon electrode conductivity, optimization of precursor solutions, and thickness of all the involved layers [20][21][23][25][26][27]. Despite these improvements, the high environmental and cost impact related to the high-temperature processes and non-radiative losses induced by the stack morphology reduce the feasibility on an industrial scale [28][29]. In addition, there are limited materials (e.g., inorganic HTM) compatible with high-temperature curing [30].

2.1.2. Low-Temperature Carbon Electrodes (LTCEs)

In the recent years, low-temperature carbon electrodes (LTCEs) in PSCs have earned a lot of attention for their low-cost manufacturing and long-term stability [31][32]. Moreover, improving the binder formulation to realize the carbon paste or ink makes it possible to obtain a good film with low-temperature annealing processes. LTCEs are

used both in n-i-p (mesoscopic or planar) and p-i-n configuration [19][33][34][35][36][37]. LTCEs grant selective holes transport, a good conductivity for external contact, thermal and chemical stability, and high hydrophobicity [34]. This type of counter electrode and its relatively low curing temperature permit a deposition directly on top of the perovskite, without the poor filling and morphology control issues of HTCEs. Furthermore, organic/inorganic HTM can be used to achieve better charge extraction and efficiencies. The first attempt to make a device with LTCEs was made by Wei et al. by directly printing the carbon ink with perovskite, without any HTM as an interlayer, with an efficiency above 11% [38]. With the growth in interest in these materials, many techniques and optimizations were implemented in C-PSCs, including HTMs between PVSK and carbon electrodes and phase engineering of active materials. Recently, the incorporation of graphene-doped P3HT as an HTM in C-PSCs shows excellent PCEs above 18%, with a shelf life and light-soaking stability of 1600 and 600 h, respectively [31]. Formamidinium-based perovskite coupled with Spiro-OMeTAD and pressed LTCE foils demonstrate the biggest efficiency for these devices, above 20% with 1000 h of shelf life stability [39]. Metal phthalocyanine [40][41], CuSCN [42], P3HT/NiO_x-CNT [31][43], NiO nanoparticles [44], Cu_2ZnSnS_4 [45], and TPDI [46] are new relevant examples of HTMs employed with a low-temperature carbon electrode. Lately, low-dimension perovskite layers employing large cations like phenethyl ammonium iodide (PEAI) and octyl ammonium iodide (OAI) were used on top of 3D perovskite with LTCEs for interfacial engineering and 2D perovskite growth, with promising efficiencies of 15.6 and 18.5%, respectively [47][48]. Calabrò et al. show the thermal (85 °C) stability improvement of a KI-doped perovskite cell by substituting the gold counter electrode with an LTCE [49]. He et al. doped the carbon paste with a small amount of CuPC as a p-dopant to modify the work function of the electrode and improve the band gap alignment.

2.2. Methodologies

The upscaling of perovskite photovoltaic technology from small-area cells to modules and the related industrial and economical transition are achievable by scalable manufacturing processes, module design, and interconnection patterning [50][51][52]. The first point is related to the deposition of the full stack by scalable deposition techniques. In the literature, many efforts are related to the deposition of the perovskite layer to get homogeneous and highly efficient modules [53]. The carbon-based devices give the chance to deposit the full stack by printing techniques. The blade-coating/doctor-blading technique deposits material using a blade on a rigid or flexible substrate. Highly homogeneous films are obtained by modifying the material amount, the meniscus gap, and the concentration and composition of precursor solutions. After deposition, an annealing process is required, sometimes in a vacuum chamber to make more efficient the solvents'/binders' evaporation [54]. The screen-printing technique adopts a patterned screen to deposit the material with a controlled thickness. These scalable deposition methods could be implemented in manufacturing processes such as roll-to-roll [23][40][46][55]. Press transfer and hot press are alternative methods to obtain single-carbon film on the substrate [56][57][58]. These types of techniques avoid the annealing process and preserve organic HTM or the passivating agent on top of the perovskite.

3. Upscaling Carbon-Based Perovskite Technology

3.1. Upscaling of High-Temperature Carbon Electrodes and Perovskite Devices

In 2013, Ku et al. showed the first HTCE heterojunction perovskite solar cell by a printable deposition method with an efficiency of 6.64% [14]. Over the last 9 years, many large-area perovskite modules working with carbon counter electrodes were reported in the literature. In 2016, Priyadarshi et al. reported a monolithic perovskite module with an active area of 70 cm^2, PCE of 10.74%, and ambient stability of more than 2000 h. The meso-TiO_2/ZrO_2/carbon stack was deposited by the screen-printing technique, and the perovskite absorber was obtained by the drop-casting method [59]. Grancini et al. demonstrated the stability of a 5-AVAI/MAPI perovskite solar module with 2D/3D perovskite interface engineering [60]. The fabrication of 10 × 10 cm^2 solar modules by a fully printable industrial-scale process reached 11.2% efficiency with zero loss at 1 sun AM 1.5 G, 55 °C for 10,000 h [60]. 5-AVAI is also used by Hu et al. combined with γ-butirrolactone (GBL) to control perovskite solvent evaporation after the infiltration step. The authors presented a 7 m^2 solar panel fully fabricated with printable techniques [15]. De Rossi et al. use 5-AVAI/MAPI perovskite in a triple-mesoscopic module with a 198 cm^2 active area and an efficiency of 6.6% after storing the module in the dark at 50–70% RH [61]. Thus, they show how the patterning optimization of the blocking layer could improve device performances. More recently, a modified mesoporous scaffold with CsX salts showed a boosted open circuit voltage of 960 mV with respect to the reference 920 mV on cells (0.7 cm^2 active area) and modules (70 cm^2) with a PCE of 12.59% and 11.55%, respectively, and stability over 2000 h in ambient conditions [62]. Xu et al. present 60.08 cm^2 active area module with a controlled infiltration method by slot-die coating. The control of the precursor solution and deposition of perovskite above the triple-mescocopic scaffold grant a final PCE of 12.87%, which is the highest value reported for such large devices (60 cm^2) [63]. Recently, Kobayashi et al. reported an evaluation about the stability of HTC-based devices. They show 4.32 cm^2 active area modules with a triple-mesoscopic high-temperature stack (meso-TiO_2/ZrO_2/carbon) [64]. The module had a PCE equal to 8.7% and was stable in damp-heat aging conditions (85 °C/85% RH); it showed stability for more than 3000 h. This stability is attributed to the light-induced performance-increasing phenomenon. The mechanism is associated to the organic molecules 5-ammoniumvaleric acid and methylammonium forming a quasi-2-dimensional perovskite/metal oxide interface with a positive effect on charge transport and suppression of ion migration [64].

3.2. Upscaling of Low-Temperature Carbon Electrodes and Perovskite Devices

In the literature, few works are present investigating LTCE-based module fabrication. In 2017, Cai et al. coupled gas-pumping perovskite with a slot-die-coating method and a low-temperature carbon electrode [65]. This approach led to reproducible solar modules (FTO/ZnO/PVK/carbon) with 17.6 cm^2 active area, a PCE of 10.6%, and no significant degradation after 140 days of outdoor testing [66]. In 2019, He et al. reported a full low-temperature-processed n-i-p mesoscopic module by doping the carbon counter electrode with copper (II) phtalocyanine. The aperture area was 22.4 cm^2, the geometrical fill factor was 89.6%, and the efficiency was 7.2% [67]. In 2021, Yang et al. added 10% guanidinium chloride ($CH_6N_3^+Cl^-$) to the perovskite precursor solution and used P3HT as an HTM [39]. The morphology and crystallinity of perovskite film were greatly enhanced, resulting in enlarged grain sizes and lowered defect densities. Moreover, they studied the passivation of the active layer with PDCBT and the interface engineering between HTM and carbon with Ta-WO_x. All these optimizations led to amazing efficiency of 15.3% on a 4 cm2 module with a fully printable fabrication process.

4. Conclusion and Perspectives

In just a few years, state-of-the-art perovskite solar cells and modules have reached impressive efficiency. Metal electrodes are intensively used despite the high costs of the materials and processes. The future of photovoltaic energy cannot base its growth on high-cost materials with a high CO_2 footprint and energy consumption. Carbon-based materials are processable with scalable techniques at high and low temperature as PSC counter electrodes. In the literature, it is found different approaches for improving and scaling up HTCE on PSM technology both on modules and panels. Fully printable triple-mesoscopic devices are a suitable choice for their easy adaptability in a possible pilot line production, but despite the excellent stability, huge differences in efficiency are still present for such devices with respect to the metal-based perovskite cells. LTCE grants low-temperature processing with relatively less energy costs. Moreover, it is possible to use various HTMs as a selective contact between perovskite and carbon material, retaining perovskite morphology as well. All these features will drive the fabrication of low-temperature carbon-based solar modules with high efficiency and stability in the coming years. Nowadays, there is a lack of literature about LTCE-based modules. Many efforts should be oriented towards scalable processes for such devices starting from the laser interconnection patterning, which has not been deeply detailed or explored. Low-temperature carbon-based PSCs have the chance to be the perovskite solar technology ready and suitable for the market. The efficiency is above 20%, and the research community is shifting its attention in this field away from the expensive and pollutant metal-electrode-based PSCs. Here, it is reported the state of the art on HT and LT carbon-based perovskite modules to help researchers understand the most feasible way towards an efficient, stable, and sustainable solar device.

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