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## ABSTRACT:

### **Perovskite Solar Cell Technology:**

#### **Materials, Printing and Engineering Routes Towards the Module Upscaling**

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Owing to their unique physical-chemical properties, [1,2] organometal halide perovskites permitted to develop a solution process photovoltaic (PV) technology able to deliver power conversion efficiencies higher than any thin film PV and closely resembling the one of silicon. [3] Thanks to a fine tuning of material compositions, device architectures and fabrication processes the efficiency of perovskite (PVSK) solar cells (PSCs) reached 26.1% for the single junction and 33.9% when used in tandem with silicon. [3,4] The efficiency transfer from laboratory cells to module devices is of paramount importance to exploit at market level the PVSK PV technology. The losses due to the layers' inhomogeneity are related to the difficulties in transferring to module devices deposition processes and material compositions optimized for the small area cells. [5] In this context, the polycrystalline nature of solution-processed PVSK layers induces defects, such as at grain boundaries and vacancies during the fabrication process. [6] A scalable, repeatable and optimized process is the baseline to obtain high efficiency devices and to upscale the PV technology. [7–10] Recently, FAPbI<sub>3</sub> has been developed as an effective absorber, with a narrow band gap of 1.47 eV. In literature, researchers scaled up to module the FAPbI<sub>3</sub> formulation by air-assisted bar-coating, [11] spin-coating, [12] or blade coating techniques, [13] but still adopting unstable HTM (Hole Transporting Materials) and gold counter-electrode. The widely used gold cathode materials for high efficiency cells and large area modules [14–16] are corroded by halogen ions, need high energy process during the device fabrication and are not cheap [17,18]. Carbon-based perovskite solar cell technology can face these issues by replacing the gold counter electrode with the cheap and stable carbon black/graphite layer [19]. In the low-temperature architecture, the carbon layer is deposited directly on top of the adsorber layer and the PVSK can have large crystals because it is not constricted in the pore size of the carbon layer. Moreover, the hole transporting material can be easily inserted to improve the hole-

extraction and reduce the non-radiative recombination at the perovskite/carbon interface.

In this work, we stabilized the  $\alpha$ -FAPbI<sub>3</sub> phase by doping with methylammonium chloride (MACl) and achieved a short-circuit current density above 24 mA/cm<sup>2</sup> and efficiency approaching 21%. We adopted low-cost and solution processed stable HTM. We performed module designing and related interconnections optimization, exploited interfacial defects passivation to suppress nonradiative recombination losses, to improve charge carrier extraction and photovoltage on module device, and optimized the deposition process to have a homogeneous large area coating. Finally, we report fully low-temperature and fully printed carbon-based FAPI PVSK cells (0.5 cm<sup>2</sup>) and modules able to achieve a champion efficiency more than 15%. Moreover, the LCA (Life Cycle Assessment) of the PVSK highlights the importance of improving the scalability of manufacturing processes to achieve competitive environmental performances.

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