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THESIS DEFENSE

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«Cellules Photovoltaïques à Pérovskite : Architectures à Base d'électrode de Carbone»

Time : «February 25, 2023» «10 a.m.»

Location : «FST Tangier (Conference room, Block F)»

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ABSTRACT

Perovskite solar cells (PSCs) are a revolutionary thin-film based photovoltaic technology, that has gained a lot of attention thanks to its ease of manufacturing processes, and rapidly growing efficiencies. However, despite the numerous attempts at improving the stability of PSCs, their potential for commercialization remains restricted as no viable operational lifetimes have so far been achieved. This doctoral thesis focuses on PSCs employing carbon as a back contact electrode. Carbon electrodes have been widely investigated to overcome the stability issues reported for PSCs, achieving promising results in terms of device durability. Nonetheless, their transformational structure has been yielding an insufficient photovoltaic performance, despite their superior stability compared to their conventional metal electrode based counter parts.

A high-temperature carbon electrode based PSC structure was used as a reference at the start of this thesis. The absence of an effective hole selective layer in such architectures has been shown to limit their performance, particularly affecting the fill factor and photovoltage. As such, derived from a fill factor obtained from certified measurements of 78.8%; a value considered especially high for such structures; a quantitative assessment of the loss mechanisms affecting the fill factor in hole selective layer free carbon electrode based PSCs was performed. Such an assessment, consisting of applying methods commonly used for established photovoltaic technologies to analyze the impact of recombination and transport losses to the fill factor, is believed to have performed on carbon electrode based perovskite devices through this thesis for the first time. For carbon electrode based PSCs without a hole selective layer, non-radiative recombination was found to contribute to only 3% of the loss with respect to the fill factor, while charge transport losses were found to be the dominating factor affecting the performance. Moreover, the perovskite/carbon interface was emphasized as a main loss channel, highlighting the importance of improving the quality of the contact between the perovskite absorber and the carbon back electrode.

For that, the low-temperature carbon device concept was introduced, as an approach offering more versatility, allowing to overcome the structural constraints faced by the high-temperature structure. The optimization of the deposition and layout of the low-temperature carbon electrode was essential to ensure an optimal contact with the perovskite layer underneath, and allow for a favorable performance. The thesis then explored the new degrees of freedom the novel structure offered by introducing a range of materials at the perovskite/carbon interface. The most effective hole selective layer employed in PSCs, spiro-OMeTAD, was found to be incompatible with the solvents in the carbon paste employed, other hole selective

materials were investigated, namely carbon nanotubes and phthalocyanines. These materials were found to be especially challenging to apply, considering their big molecules hindering their optimal dispersion, thus leading to mediocre cell performances. P3HT was also tested, allowing for promising efficiencies, though the high pin hole density in the film caused the rapid degradation of the perovskite absorber.

Applying a two dimensional perovskite layer developed by partners at EPFL between the perovskite absorber and the carbon back electrode in the lowtemperature structure was found to effectively compensate the losses perceived at the interface by acting as an electron blocking layer. The low dimensional perovskite allowed to successfully overcome the interfacial recombination losses and decrease charge transport losses, suggesting an effective suppression of electron back transfer. This allowed for an improved fill factor and photovoltage, leading to an efficiency of 18.5% with a photovoltage of above 1 V.