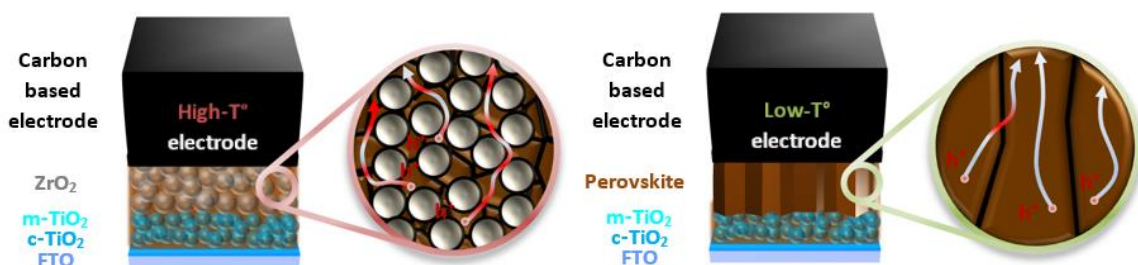


Quantifying Losses of Perovskite Solar Cells with Carbon-based Back-contacts and Outlining a Roadmap on Boosting Their Power Conversion Efficiencies

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Numerous studies have shown that perovskite solar cells with carbon-based contacts (C-PSCs) provide strong potential for delivering stable and up-scalable perovskite photovoltaic devices. However, their power conversion efficiencies (PCEs) are still lagging behind the conventional solar cells with metallic back-contacts. This necessitates a deeper understanding of the power losses present in C-PSCs in order to find effective strategies to reduce them. In principle, one can distinguish between two types of C-PSCs: (i) where the back-contact is cured at high temperatures (typically $> 400^{\circ}\text{C}$), thereby allowing perovskite to be integrated into cell stack only after its deposition and (ii) where the back-electrode is deposited at low temperatures (typically $< 120^{\circ}\text{C}$), which enables layer-by-layer deposition. Both cell structures have significant differences not only in the processing conditions, but also in the dominant losses present in the corresponding PV devices. For the first time, we conducted an objective experimental study to identify the main losses in both types of C-PSCs. We found that the major limitation of the cells with cathodes treated at high-temperatures is the non-radiative recombination happening at the numerous grain-boundaries, which are present in the mesoscopic cell stack of such cell. In contrast, the cells with low-temperature treated contacts can have large perovskite crystals due to more favorable crystallization techniques, allowed via layer-by-layer deposition. By combining experimental results with our numerical simulation we quantitatively demonstrate that the low-number of grain-boundaries reduces non-radiative recombination, thus increasing the quasi-Fermi level splitting of perovskite, prolonging charge carrier lifetime, which results in an impressive $V_{oc} > 1.1\text{V}$ in the HTL-free C-PSCs with low-temperature treated contacts. However, we note that in such cells the transport is hindered by the perovskite/carbon contact which significantly reduces the fill factor of such PV devices. Finally, we outline the promising methods of reducing non-radiative recombination and improving charge carrier transport in both types of C-PSCs to fulfill their potential. We further highlight the advantages of the C-PSCs with low-temperature treated electrodes due to higher flexibility of processing conditions, which allows to integrate wide range of charge-transport layer with favorable properties, enhanced crystallization, compatibility with roll-to-roll manufacturing and faster fabrication.



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