

Water-free PEDOT: PSS Formulation for Pb-Sn Mixed Perovskite Single-Junction and All-Perovskite Tandem Solar Cells

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Abstract

Single-junction Pb-Sn perovskite solar cells (PSCs) with a bandgap of 1.22eV are peaking at a power conversion efficiency (PCE) of over 23%. These efficiency values have been reached by following strategies to reduce non-radiative recombination losses by improving the quality of the absorber via additive incorporation and the interface with the electron transport layer (ETL) via surface passivation. Regardless, PEDOT:PSS is the standard hole transport layer (HTL) due to its high conductivity and selectivity. Its incorporation introduces drawbacks to the processing of perovskite devices, as it is typically processed from water-based dispersions, not being compatible with inert glovebox conditions. Additionally, it can permeate through the recombination layer and degrade the wide bandgap perovskite absorber (WBG), reducing reproducibility and increasing batch-to-batch variations in tandem cells. Due to the presence of PSS, the formed layer is acidic and hygroscopic, adding to the unfavorable stability of narrow bandgap perovskites and thus all-perovskite tandems. To tackle these problems, an anisole-based PEDOT:PSS formulation compatible with processing in the glovebox was introduced. By doing so, the WBG sub-cell never encounters ambient conditions. From contact angle measurements it is shown that the new PEDOT:PSS is more hydrophobic which is in agreement with the lower PSS content. Thickness measurements revealed an HTL thickness of 100 nm which can be tuned down by changing the processing parameters or diluting the coating solution. While thinner layers resulted in higher visual transmission, the increase in photocurrent generation was found to be negligible. Optical simulations are performed to analyze this behavior. At the same time, photoluminescence quantum yield (PLQY) measurements were conducted, showing a 60 mV increase in the implied open circuit voltage (iV_{OC}) in comparison to the standard device. The optimally thick HTL also positively impacts charge extraction, as can be seen from transient PL measurements. Finally, from current-voltage measurements an increased efficiency (from 19.4% to 23.3%) as well as improved reproducibility and shelf-life stability in all-perovskite tandem devices utilizing the new PEDOT:PSS formulation was observed. Tracking of the maximum power point revealed $\geq 21\%$ stabilized efficiency, in contrast to a decreasing PCE for the standard device. Overall, our research provides insights into

the application of a water-free PEDOT:PSS dispersion which can substitute the standard PEDOT:PSS without compromising any of its benefits, but instead paving the way for more efficient, reproducible, and stable PSCs.

Keywords

Narrow Bandgap, All-Perovskite Tandems, PEDOT:PSS, Stability