ABSTRACT

This thesis encompasses optimization studies on single-junction PSCs, recycling of the end-of-life (EOL) devices, experimental interface engineering studies at 3D perovskite/hole transport layer interface by using 2D perovskites, and simulation studies on perovskite-silicon and all-perovskite tandem solar cells. A systematic comparison between doping and post-treatment of SnO_2 ETLs with KCl was explored. Doped SnO_2-based ETLs exhibited superior performance, with higher conductivity and transmission, offering high perovskite grain size, compressive strain to the perovskite film, robust perovskite material, higher charge carrier lifetime, and reduced trap density, leading to enhanced power conversion efficiency (PCE) compared to treated ETLs. We explored the recycling of EOL PSCs by realizing the cost and carbon footprint associated with expensive FTO-coated glass substrates. Expensive substrates were recovered successfully by establishing a novel multi-step (MS) recycling process involving KOH and dilute HCl post-treatment. This approach effectively removed residual lead and other impurities from the recycled FTO/ETL substrate. For the first time in this work, we established a parallel correlation between the presence of residual impurities and reliable device fabrication on recycled substrates. The role of 2D perovskite capping layers in passivating the 3D perovskite/hole transport layer interface was investigated for single-junction PSCs because of its electron blocking, tunable bandgap, and moisture blocking abilities. An optimized 2D spacer cation with a concentration of 2.5 mg/ml solvent washed yielded a PCE of 18.8%. Further, optoelectronic simulations demonstrated the potential of 3D/2D perovskites in perovskite/silicon tandem cells, achieving open-circuit voltages above 2 V due to surface passivation effects, and PCE above 40% was also observed. Lastly, the numerical studies of all-perovskite tandem solar cells, exploring the impact of various ETLs directly interfaced with the perovskite layers to resolve the issues with the C_{60}/perovskite interface, were studied. A PCE above 35% and alternative ETL (PCBM) were observed to perform better than the C_{60} ETL. The discovery of better-quality ETLs, successful recycling of EOL devices, passivation using 2D perovskites, and optimization of tandem architectures pave the way for highly efficient, stable, and sustainable perovskite-based photovoltaic technologies.

Keywords: Perovskite Solar cells, Ambient fabrication, Recycling, ETL Modifications, Perovskite/Silicon Tandems, All Perovskite Tandems, Interfacial Defect Passivation.