

Abstract

It is crucial to increase the harnessing of solar energy and thus diminish our dependency on fossil fuels, which is one of the solutions to mitigate the global warming by slowing down the rapid increase in greenhouse gas emissions. Therefore, photovoltaics plays an important role in the efficient decarbonization of power generation by transition from fossil fuels to renewable and sustainable energies. Recently, the most promising candidate among the new photovoltaic technologies are the perovskite solar cells due to the cost effectiveness, high power conversion efficiency, and simple fabrication process. The key component in a perovskite solar cell device is an organic-inorganic hybrid photo absorber of general formula ABX_3 (where $A = Cs^+, Rb^+$, methylammonium (MA) $(CH_3NH_3^+)$, formamidinium (FA) $(CH(NH_2)_2^+)$, guanidinium (GA) $(CH_6N_3^+)$, $B = Pb^{2+}, Sn^{2+}$, $X = I^-, Br^-$). The intrinsic properties of perovskite materials are highly attractive due to high electron mobility, high charge carrier diffusion length, ambipolar charge transport behavior, high absorption coefficient, low exciton binding energy, long carrier lifetime, tunable bandgap, and excellent structural defect tolerance. The rapid development of perovskite-based photovoltaics has meanwhile recorded power conversion efficiencies exceeding 25% to become a key player of third-generation photovoltaics.

Despite their impressive photovoltaic features, inhomogeneity in the polycrystalline films and the anomalous photo current density-voltage responses are holding back the commercialization of perovskite solar cells. In this work, GA has been introduced in perovskite composition to synthesis uniform and highly crystalline perovskite films via two step sequential deposition method. Using this approach, the power conversion efficiency of GA-based perovskite solar cells in this work has been increased from 10.4% to 12.9%.

In this thesis, the possible compositional variations of the perovskite absorber by addition of secondary cations were investigated. Multi-cation mixed halide perovskite single crystals were grown using an inverse temperature crystallization process in γ -butyrolactone solvent. Moreover, thin films of same perovskite compositions $((A^1, A^2, A^3, A^4)B(X^1, X^2)_3)$ were also prepared via solution processing approaches and integrated into thin film solar cell devices. The influence of multi-cation, processing parameters and the impact of chemical modification on the A-site of the hybrid perovskite thin films with GA cation have been studied and evaluated by a variety of spectroscopic and analytical techniques. Efficiently operating organic-inorganic lead halide perovskite solar cells in *n-i-p* device stack configuration could be

obtained. It was observed that a mixture of the A-site cations in perovskite lattice leads to its stabilization and band gap narrowing in the perovskite which allows more photons to be converted into electrons. As a result, the highest power conversion efficiency above 20% with negligible hysteresis was achieved in this work.

In the second part of the thesis, alternative hole transport materials were studied. The use of hole transport materials is still essential for efficient charge carrier extraction in perovskite solar cells.

To date, Spiro-OMeTAD (2,2',7,7'-tetrakis-(N,N-di-4-methoxyphenylamino)-9,9'-spirobifluorene) and PTAA (poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine]) have been selected as hole transport materials to maintain an efficient performance, however, the high price of these materials, which cover more than 30% of the total module cost can disrupt the development of perovskite solar cell manufacturing. Furthermore, such wide band gap hole transport materials typically require doping to match the required electric conductivity, which is problematic because the humidity makes the hole transport layer hydrophilic that negatively affects the stability of the entire solar cell device. In this work, a new alternative of the different types of hole transport materials based on tetra-phenyl-porphyrin was studied. Highly ordered characteristic of tetra-phenyl-porphyrin based hole transport materials enhance charge carrier transport within a perovskite solar cell device leading to a power conversion efficiency over 18%. Moreover, hydrophobic nature of tetra-phenyl-porphyrin may help to protect the perovskite layer from the humidity and prevent the diffusion of external moieties.