

Abstract

In this thesis, thin films of organic-inorganic lead halide perovskites (ABX_3) have been prepared *via* solution processing approaches and integrated into thin film solar cell devices. For this purpose, a low temperature single-step fast crystallization method – the instantaneous perovskite crystallization process (IPC) – was developed for the deposition of $CH_3NH_3PbX_3$ ($X = \text{halide}$) thin films. The influence of short-chain lead (II) carboxylate precursors, processing parameters and the impact of chemical modification on the X-site of the hybrid perovskite thin films with methylammonium cation have been studied and evaluated by a variety of spectroscopic and analytical techniques. Efficiently operating planar organic-inorganic lead halide perovskite solar cells in both device stacks (*n-i-p* and *p-i-n*) could be obtained by this approach. The highest power conversion efficiency of 15.6% was obtained *via* IPC process at a processing temperature of 75 °C for $CH_3NH_3PbI_3$ thin films in *n-i-p* device stack configuration prepared using $Pb(CH_3COO)_2$ and CH_3NH_3I precursors in *N,N*-dimethylformamide solvent. Time resolved photoluminescence spectroscopy measurements revealed mono-exponential decay characteristic with high charge carrier lifetime of 691 ns, coherent with the solar cell measurements under simulated sunlight conditions. By using the same combination of precursor in γ -butyrolacton, room temperature processed thin film solar cells in *p-i-n* device configuration with a power conversion efficiency of 10.4% were obtained. For $CH_3NH_3PbI_{3-x}Br_x$ perovskite films the charge carrier lifetime decreased with increasing bromide content. Here, phase separation of nanoaggregates was observed, as evident from the large deviation of the electronic band gap from the optical, probed by steady-state photoluminescence spectroscopy. For $Pb(CH_3CH_2COO)_2$ precursor, the measured champion solar cell efficiency in *p-i-n* device configuration was slightly higher for the IPC processed specimen (5.3%) in comparison to post-annealed $CH_3NH_3PbI_3$ thin films, due to hysteretic current-voltage behaviour. For $Pb(HCOO)_2$ precursor no operational solar cell devices could be obtained in both device stacks, due to bad film quality and the formation of byproducts of $(CH_3NH_3)_4PbI_6$.

Furthermore, protic ionic liquids with methylammonium cation and carboxylate anion have been synthesized and successfully employed as alternative solvents for hybrid lead halide perovskite processing in their pristine state and in binary blends with co-solvents such as water, alcohols and acetonitrile. By using a ternary solvent system, based on the new protic ionic liquid methylammonium propionate, acetonitrile and dimethyl sulfoxide, $(MA_{0.15}FA_{0.85})Pb(I_{0.85}Br_{0.15})_3$ perovskite thin films could be prepared by a sequential deposition method and successfully integrated into solar cell devices. In addition, the effects of surface

modification of mesoporous TiO₂ thin films by alkali bis-(trifluoromethanesulfonyl) imide (TFSI) treatment were studied by X-ray photoelectron spectroscopy (XPS). Here, a trade-off between type of alkali-metal and precursor concentration was observed. Through potassium-TFSI surface treatment with optimized precursor concentration of 0.2 mol/l, air processed perovskite solar cells with stabilized power conversion efficiency of 20.6% could be obtained.