Perovskite Solar Cells: Understanding the Degradation and Enhancement in the Optoelectronic Properties of $MAPbI_3$ Thin Films

Abstract

Organic-Inorganic halide perovskites have been a breakthrough in the field of photovoltaic research. The eminent interest of the photovoltaic community in perovskite research has been developed due to the exceptional optoelectronic properties and low-cost solution-processable fabrication techniques offered by the material. For perovskite solar cell technology to prove its technological viability, two major aspects have to be achieved: stability in the operating environment and performance. In this thesis work, we have worked towards understanding the degradation mechanism of perovskite solar cells and improving the stability as well as enhancing the photophysical properties of perovskite films. Here-in the fabrication techniques and precursor materials that are used to prepare the perovskite film are investigated, developing them in a way to provide a stable and high performing perovskite film.

The initial stage of thesis work is oriented towards understanding the degradation mechanism in $MAPbI_3$ perovskite solar cells. The effect of different fabrication routes on the degradation behavior of perovskite solar cells has been explored. It is investigated that different fabrication route shows different decay rate and follow a dissimilar route while decomposing into its by-product. A detailed analysis based on structural data has been carried out to probe into the difference in the degradation pathways. The scientific outcomes of this work unravel that the root cause for the distinct degradation mechanism lies in the crystal symmetry and lattice orientation of the crystallized perovskite films.

Based on the understanding on degradation mechanism, stability of $MAPbI_3$ perovskite solar cells has been improved by improving the surface morphology. The impact of temperature-assisted crystallization of precursor PbI_2 layer on the surface characteristics and performance improvement of perovskite solar cells has been demonstrated. We noted a significant improvement in the surface features and crystallite size of PbI_2 film annealed at 200˚C that led to improved efficiency of 13.51% with respect to 4.27% for the controlled film. Significant stability improvement noticed for perovskite prepared from PbI_2-200˚C showing less than 10% degradation in efficiency even after 384 hours of ambient exposure

whereas, the controlled film indicates a 50% efficiency loss within 72 hours of aging. The rationales behind the improvement in the overall performance have been discussed thoroughly. Furthermore, a detailed analysis of the role of lead iodide surface characteristics on the perovskite morphology and performances has been investigated.

A novel Pb-precursor composition has been reported to target the issue of incomplete conversion of lead iodide into perovskite in Two step (Sequential) deposition process. The work reports that the incorporation of $Pb(Ac)_2.3H_2O$ in the PbI_2/DMF solution improves the deposition process. A novel Pb-precursor composition containing PbI_2 (70vol%) and $Pb(Ac)_2.3H_2O$ (30vol%) results in a change in the crystallization process of Pb-precursor film and allows it to crystallize in Trigonal crystal symmetry with (104), (101), (110) and (003) lattice orientation instead of its standard highly ordered Hexagonal phase with (001) orientation. Besides $Pb(Ac)_2.3H_2O$ incorporation gives a porous PbI_2 film that provides ease in volume expansion. Moreover, we evidenced a more than 4-fold faster conversion reaction by using the novel-Pb precursor without residual PbI_2 whereas, the controlled PbI_2 film didn't show complete conversion even after prolonged reaction time. Detailed investigation on the mechanism for faster and complete conversion has been discussed using structural analysis.

An easiest and effective approach has been published to fabricate high-quality perovskite films with enhanced optoelectronic properties. It reports that Pb-precursor solution prepared from 30vol% of $Pb(NO_3)_2$ and 70vol% of PbI_2 salts in DMF solvent, significantly improves the photophysical properties of perovskite as compared to the controlled sample. Interestingly, we observed significant spatial photoluminescence heterogeneity in the perovskite sample prepared from pristine PbI_2 film. The possible reasons for fluorescence heterogeneity and PL shoulder peak have been discussed. Detailed analysis on the correlation between the surface morphology of perovskite film and emission heterogeneity across the sample has been included in the chapter. Furthermore, the synergistic effect of $Pb(NO_3)_2$ addition on the morphology of the Pb-precursor layer and the benefit in subsequent conversion process has been discussed in detail using atomic force microscopic investigations.