PREFACE

The worldwide increase in energy consumption has motivated scientists to explore the potential of solar energy. At present, solar energy harvesting is carried out mostly (more than 90%) by the inorganic crystalline silicon (Si) solar cells. Moreover, Si-based solar cells are now approaching their maximum theoretical limiting efficiency of ~29.43%. The latest advancements in technology and materials have led to the development of novel photosensitive materials that enabled the fabrication of low cost and highly efficient photovoltaic devices. The state of the art growth in various perovskite materials like inorganic ($CsPbX_3$: X=I, Cl, Br) and hybrid (ABX_3; A= organic compound, B= inorganic compound, and X= halides) have attracted huge interest among the researchers. Recently, methylammonium lead halides ($CH_3NH_3PbX_3$) have shown highly encouraging photosensitive for photovoltaic application, which gained immense research attention since its discovery and has boosted hopes for new photovoltaic technologies.

Typically, the optical and electrical characteristics of photovoltaic devices depend upon the device design, fabrication procedures, semiconductor/active material, etc. The different synthesis and deposition techniques may be used for the tuning of the electronic and optical properties of as-grown thin films. Additionally, the low-dimension nanostructures such as nanorods or nanowires that offer large surface-to-volume ratios are mostly used for the charge transport layer in perovskite solar cells (PSC). From this perspective, the present thesis deals with the TCAD simulation, fabrication, and characterization of TiO_2/ZnO nanorods electron transport layer (ETL) based hybrid perovskite ($CH_3NH_3PbI_3$) solar cells. In PSCs' design, the PTAA/Spiro-OMeTAD polymer is employed as hole transport material where small molecule materials (Li-TFSI and TBP) are used as doping elements to enhance the conductivity of the hole transport layer (HTL).

The primary focus of this thesis is to explore the performance parameter of PSC by means of the synergic effects of the modified synthesis process for ETL through optimized doping in HTL. The uniformly distributed and vertically aligned TiO_2/ZnO Nanorods have been grown on FTO substrates by the hydrothermal method, whereas both HTL and absorber layers are deposited via spin-coating techniques. Later, the optical and electrical properties of TiO_2/ZnO Nanorods have been explored in detail. The thesis consists of six chapters, which are briefly outlined as follows:

Chapter-1 introduces the hybrid perovskite material's optoelectronic properties, thin-film synthesis process, and the working principle of perovskite solar cells. A brief introduction about device models and thin film characterization techniques have been discussed. Finally, a detailed literature survey, motivation, and scope of the thesis have been presented.

Chapter-2 presents TCAD simulation and fabrication of hybrid perovskite solar cells. The electrical and optical characterization has been investigated for the device structure Pd/PTAA/hybrid perovskite ($CH_3NH_3PbI_3$)/TiO_2 Nanorods (TNRs) grown on an FTO coated glass substrate. The TNRs layer is synthesized by a low-cost hydrothermal process and acts as the ETL, whereas the PTAA acts as the HTL. The solar cells are optimized, fabricated, and characterized for different TNRs thickness and then several device performance parameters such as short circuit current density (J_{SC}), open-circuit voltage (V_{OC}), fill factor (*FF*), and power conversion efficiency (*PCE*) and etc. are studied. The *Solar Cell Capacitance Simulator-One Dimensional* (SCAPS-1D) is used to simulate the proposed solar cell structure and validated via our experimental results. The effects of thickness variation of ETL on the solar cell parameters have been investigated by solving the drift-diffusion model. The measurements show that the efficiency of the solar cell is decreased with the increase in ETL thickness, which is attributed to higher trap sites in the active layer and ETL. The maximum optimized efficiency of 15.04% is obtained for ETL thickness of 500 nm. On the other hand, the simulated results are in close resemblance to the experimental results, having an efficiency of 15.69%.

Chapter-3 reports the efficiency improvement of perovskite solar cells (PSCs) by solvothermal etching and $TiCl_4$ treatment of TNRs based ETL. The TiO_2 NRAs have been explored for the ETL due to their better direct carrier transportation over other TiO_2 nanostructures. The solvothermal etching of TiO_2 NRAs enhances the surface-to-volume ratio of the ETL, which, in turn, enhances the power conversion efficiency (PCE) of the PSCs. All the measurements have been performed at room temperature and high humid (with ~65% humidity) conditions to demonstrate the performance of the PSCs under normal environmental conditions. A noteworthy efficiency of 15.16% with an improved fill factor (FF) and short circuit current density (J_{SC}) has been reported in the proposed PSCs. The PSC performance is further improved by the $TiCl_4$ treatment of the solvothermally etched TiO_2 NRs as the ETL in the device.

Chapter-4 discusses the effect of the seed layer, which directly affects the growth of the ZnO Nanorods (ZNRs) and related photovoltaic parameters of the hybrid perovskite solar cell. Four different types of ZnO seed layer samples have been synthesized using four methods, namely: ZnO drop-cast, ZnO nanoparticles (NPs), ZnO quantum dots (QDs), and ZnO solvothermal on the FTO substrate. ZnO drop-cast results in less density, tilted, and non-uniform deposition of the nanorods. But uniform coverage, high volume to the surface, and vertical direction growth of ZNRs are observed in the ZnO QD seed layer sample compared to the other three samples. Morphology and crystalline structure were analyzed with HRSEM, TEM, and XRD, whereas optical absorption, emission, and transmittance are recorded using UV-Visible, and Photoluminescence, respectively. Subsequently, the electrical characterization reveals that the optimum photovoltaic parameters are obtained on the seed layer of ZnO QD, which leads to the power conversion efficiency of 10.69% for ZnO NR based perovskite solar cell structure (FTO/ZNRS/$CH_3NH_3PbI_3$/PTAA/Au).

Chapter-5 presents the simulation, fabrication, and characterization of ZnO Nanorod (ZNRs) based PSCs under ambient air conditions. The proposed PSC structures use a $CH_3NH_3PbI_3$ hybrid perovskite-based active layer sandwiched between a ZnO Nanorods (NRs) ETL and a Spiro-OMeTAD (undoped and doped) HTL. The ZnO NRs are grown using a low-cost solvothermal process at relatively low temperature. The performance of fabricated PSCs is analyzed for both the undoped and doped (with TBP and LiTFSI) spiro-OMeTAD based HTLs. All the solar parameters, namely, short circuit current density (J_{SC}), open-circuit voltage (V_{OC}), fill factor (*FF*), power conversion efficiency (*PCE*), and external quantum efficiency (*EQE*), are calculated from experimentally measured current density versus voltage (*J-V*) and wavelength transient characteristics in ambient condition. The maximum *PCE* of 10.18% is obtained for the doped HTL, whereas 9.51% for undoped HTL. The improved performance due to HTL doping is attributed to the enhanced charge transportation of the HTL. The experimental results obtained from the fabricated PSCs are also compared with the SetFos^{TM} TCAD simulation data using the drift-diffusion model. The simulated results are observed to be well matched to the experimental data.

Chapter-6 includes the major findings of the thesis along with a brief outline for the future scope of research related to the present thesis.

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