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**Title of the thesis:** Study of Perovskite Semiconductor Materials for Photo-Energy Conversion.

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## **Abstract**

Hybrid organic-inorganic halide perovskites have been the subject of intense and productive research over the last few years as a third-generation photovoltaic technology. However, many aspects of this class of materials are still a mystery. For example, the origin of hysteresis in some perovskite solar cells, the underlying cause for such a high open-circuit voltage, and low recombination in films which in some materials would be considered of very low quality. Also, because of their promise for large-scale applications such as photovoltaics, perovskites' stability and mechanisms of degradation must be well characterized. In these and other remaining puzzles, there is ample room for contributions from chemists, materials scientists, and physicists working in both experiment and theory. Finding the correlation between the chemical structure and the subsequent photo-physical properties is a valuable tool for overcoming some fundamental concerns that hinder further growth.

The most important application of perovskite is light-harvesting and converting photon energy into electric current as an application in solar cells and photodectors. A lot of research is focused on the relationship between the bandgap and the chemical components. The bandgap engineering in halide-based hybrid perovskites could be accomplished by mixing anions and/or cations in the  $\text{CH}_3\text{NH}_3\text{PbX}_3$  perovskite system with varied and abundant choices. For example,  $\text{CH}_3\text{NH}_2^+$ ,  $\text{Cs}^+$ , etc. can be exchanged at A site with methylammonium cation,  $\text{Sn}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Bi}^{3+}$ , etc. at B site with the  $\text{Pb}^{2+}$  and a mixture of more than one halogens Cl, Br, or I ions can be accommodated within the perovskite matrix having different photophysical properties.

The main idea of this thesis is to (i) Synthesize different hybrid perovskite photo absorbers using different methods (ii) Investigate the photophysical properties of pure and mixed-cation perovskite systems like  $\text{CH}_3\text{NH}_3\text{PbI}_3$ ,  $\text{CsPbBr}_3$ ,  $\text{CH}_3\text{NH}_3\text{Pb}_{1-x}\text{Cd}_x\text{I}_3$ ,  $\text{Cs}_x(\text{CH}_3\text{NH}_3)_{1-x}\text{PbI}_3$ , etc. using different measurement techniques, (iii) Study the effect of the several

properties, processing conditions of charge extracting layers on the performance of perovskite photovoltaic devices. (iv) Hybrid perovskite absorbers were used in different device architectures to study the effects of various material parameters and operating conditions like interfaces between different functional layers, absorber thickness, defect states, back-contact work functions, and working temperature on the performance of parameters of perovskite solar cells (PSCs).

In chapter 3, methylammonium lead iodide,  $\text{CH}_3\text{NH}_3\text{I}$  precursor was synthesized at  $0^\circ\text{C}$  and purified before it was used along with  $\text{PbI}_2$  and  $\text{CdI}_2$  precursors in N, N dimethylformamide (DMF) to prepare  $\text{CH}_3\text{NH}_3\text{Pb}_{1-x}\text{Cd}_x\text{I}_3$  ( $x = 0, 0.03, 0.05 \& 0.08$ ) at  $80^\circ\text{C}$  in solid thin-films and solution form. The characterizations were performed to examine the perovskite crystal structure, energy bandgap ( $E_g$ ), morphology, elemental composition, and crystalline size using X-ray diffraction, UV-Visible spectrophotometry, photoluminescence, X-ray photoelectron spectroscopy, SEM, and TEM measurements. We observed the tetragonal crystal structure of the  $\text{CH}_3\text{NH}_3\text{Pb}_{1-x}\text{Cd}_x\text{I}_3$  samples was preserved throughout the doping range and the bandgap reduced slightly from 1.55 eV to 1.5 eV, while the absorption coefficient remained high ( $\sim 10^3$ ) above 750 nm in the spectrally important infrared range.

Chapter 4 is based on studying the device performance of two perovskite solar cell devices, FTO/CdS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /CuO/carbon and FTO/CdS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /Cu<sub>2</sub>O/carbon using solar cell capacitance simulator (SCAPS-1D) program. Here, we used inorganic semiconductors CuO and Cu<sub>2</sub>O as hole transport layers (HTLs) and CdS acted as a common electron transport layer (ETL). The effect of parameters such as absorber thickness, absorber defects, interface defects, acceptor density, hole mobility, work function, temperature, and ETL thickness on the photovoltaic performance was studied. The best J-V characteristics and quantum efficiency were obtained for an absorber thickness range of 500-600 nm, defect density below  $10^{14} \text{ cm}^{-3}$ , and ETL thickness in the range of 40-50 nm. The average conversion efficiency of over 21 % was achieved with the given cell configurations. We also studied the synthesis and probing of the structure, dielectric and optical properties of Cs-doped methylammonium lead tri-iodide perovskite,  $\text{MA}_{1-x}\text{Cs}_x\text{PbI}_3$  (MA =  $\text{CH}_3\text{NH}_3$ ) system. A large value of the dielectric constant for perovskite samples was observed at low frequencies which reduces the Coulomb interaction between photoexcited electrons in the conduction band and holes in the valance band by effective charge screening. The decrease in the exciton binding energy, in turn, induces ambipolar charge transport which has profound implications on the electronic and

photovoltaic operation of a device. The elemental composition, chemical state, and electronic state of the elements were investigated by X-ray photoelectron spectroscopy.

In chapter 6, the typical representative of inorganic halide perovskites, CsPbBr<sub>3</sub> nanocrystalline material was synthesized by the solvothermal method and quenched to the high-temperature cubic phase. Intermediate-bandgap (2.28 eV) polycrystalline semiconductor CsPbBr<sub>3</sub> was investigated using different characterization techniques. Morphology of the crystalline material (average size ~ 25 nm) was studied using SEM and TEM microscopy. Reflectance and emission spectra were also recorded to study optical properties. Electronic properties, the density of states (DOS), and band-structure calculations were performed from the first-principle study using the Vienna *ab initio* simulation package (VASP). It was demonstrated that the states around the bandgap arise from Pb and Br-atoms while there is no visible contribution from the Cs atom. This study offers a simplistic and adaptable pathway to rationally control the properties of inorganic lead halide perovskites leading to opportunities for applications, such as in lasing, light-emitting diodes, solar cells, photodetectors, etc.