

The fast rise in carbon emissions from 9.4 million tons to 37 billion tons since the beginning of the industrial revolution highlights urgent requirement of sustainable energy sources as the world confronts climate change and endeavors to achieve sustainability. During daylight, the solar cells can offer constant power supply for low power residential to high power industrial usages, however the intermittent nature of solar radiation requires energy storage systems to store the solar energy into electrochemical energy for usage in nighttime or at later stage. This conventional combination of solar cell and battery reduces energy poverty, but is very bulky and expensive due to the use of complex and multiple electronic components, duplication of electrodes as well as require use of various materials for these two very different type of devices. In contrast, photo-batteries, also known as photo-rechargeable batteries (PRBs), offer a compact energy solution by integrating solar harvesting and energy storage functionalities in a single device. These PRBs are composed of either single or dual active materials which can perform solar energy harvesting like a PV device and concurrently stores it in the form of electrochemical energy like a battery. The overall performance of these PRBs highly depends upon the optoelectronic and electrochemical properties of the active materials. This thesis involves the investigation of advanced PRB systems by exploring the single active material systems based on metal halide perovskite (MHPs). Bulk (3D) MHPs of type AMX_3 (formula defined below) are solution processed semiconductors and offer an ideal material system for low-cost PVs, however these materials suffer from stability issues due to degradation of their structural and optical properties under atmospheric and illumination conditions. Therefore, due to improved structural and optical properties of two-dimensional (2D) MHPs, compared to 3D MHPs, this thesis work highlights the investigation of optoelectronic, electrochemical and photo-electrochemical properties of 2D MHPs for PRB applications. Briefly, the Ruddlesden Popper (RP) phase 2D MHPs $(R')_2(A)_{n-1}M_nX_{3n+1}$ ($n=1-4$) are studied by exploring their structural, optical, optoelectronic, electrochemical as well as photo-electrochemical properties. Here, R' is the monoammonium alkyl organic spacer cation, A is the smaller organic cation ($MA=CH_3NH_3$, $FA=HC(NH_2)_2$, Cs etc.), M is the divalent metal (Sn^{2+} , Pb^{2+} , Ge^{2+} etc.), X is the halide (Cl^- , Br^- , I^-) and n denotes the number of inorganic layers between the organic spacer cations. In order to study these properties, a wide range of samples and devices are fabricated ranging from photodetectors, Li-Ion batteries (LIBs) to PRBs. These 2D MHPs are found to be an interesting candidate for PRB applications due to their flexible structural properties, improved stability in electrolyte medium, tunable optical properties, compatibility with carbon additives and binders, matching energy levels to the charge transport materials such as rGO, PCBM, MoS_2 , MXene etc. as compared to their bulk counterpart.

A hybrid material using RP perovskite $(BA)_2(MA)_{n-1}Pb_nI_{3n+1}$ ($n=1-4$) and a few layers of MoS_2 nanoflakes is developed to investigate the optoelectronic and photo electrochemical properties. This hybrid material has shown improved transient photocurrent response than pristine material. Such improvement in the photocurrent leads to responsivity of $\sim 97.7 \mu A W^{-1}$ and detectivity of $\sim 6.93 \times 10^8$ jones at 0.0 V bias under 1 sun illumination, which are ~ 16 and ~ 10 times higher respectively as compared to pristine device. In continuation of prior work, this RP perovskite- MoS_2 hybrid material based LIB exhibits an average discharge specific capacity of 129.17 mAhg^{-1} for 50 cycles at 294 mAg^{-1} current density. Whereas pristine RP perovskite LIB has shown a specific capacity of 25.60 mAhg^{-1} under similar conditions. Furthermore, the hybrid material-based Li-PRB has shown an average dark discharge specific capacity of 128.66 mAhg^{-1} at a current density of 64 mAg^{-1} which enhanced to 180.67 mAhg^{-1} (photo-enhancement: 40.42 %) under illumination. In addition, demonstrated hybrid Li-PRB has shown photo conversion and storage efficiency of 0.52 % under standard 1 Sun illumination, which outperforms other perovskite-based PRBs. An aromatic spacer cation-based RP perovskite of type $(CH)_2(MA)_3Pb_4Br_{13}$ and MXene nanoflakes based hybrid materials is fabricated and these RP perovskite-Mxene hybrid photodetector ($n=4 + 1.5 \text{ mM MXene}$) has shown responsivity of $\sim 1.51 \times 10^2 \text{ AW}^{-1}$ at 2.0 V bias under laser illumination ($\lambda_{ex} \sim 405 \text{ nm}$, $P_{in} \sim 0.62 \text{ mWcm}^{-2}$) which is ~ 8.39 times

higher than pristine RP Perovskite photodetector. Furthermore, the effect of smaller organic cation (A = MA and FA) on the optoelectronic properties of aromatic spacer cation-based RP perovskite of type $(\text{CH})_2(\text{A})\text{Pb}_2\text{I}_7$ is studied. Where perovskite with FA as a smaller cation has shown stable transient photocurrent response due to its high molecular weight which has resulted in responsivity of $\sim 2.6 \text{ AW}^{-1}$ and detectivity of $\sim 3.6 \times 10^{10}$ jones at 2.0 V bias under CW-laser illumination ($\lambda_{\text{ex}} \sim 405 \text{ nm}$, $P_{\text{in}} \sim 5.87 \text{ mW/cm}^2$). This thesis provides a novel approach to improve the optoelectronic, electrochemical as well as photo-electrochemical properties of 2D metal halide perovskites.

...