World Academy of Science, Engineering and Technology
International Journal of Energy and Environmental Engineering
Vol:18, No:04, 2024

Mixed Tetravalent $Cs_2Ru_mPt_{1-m}X_6$ (X = Cl-, Br-) Based Vacancy-Ordered Halide Double Perovskites for Enhanced Solar Water Oxidation

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Abstract : Vacancy ordered double perovskites (VOPs) have been significantly attracting researchers due to their chemical structure diversity and interesting optoelectronic properties. Some VOPs have been recently reported to be suitable photoelectrodes for photoelectrochemical water-splitting reactions due to their high stability and panchromatic absorption. In this work, we systematically synthesized mixed tetravalent VOPs based on $Cs_2Ru_mPt_{1-m}X_6$ (X = Cl-, Br-) and reported their structural, optical, electrochemical and photoelectrochemical properties. The structural characterization confirms that the mixed tetravalent site intermediates formed their own phases. The parent materials, as well as their intermediates, were found to be stable in ambient conditions for over 1 year and also showed incredible stability in harsh pH media ranging from pH 1 to pH 11. Moreover, these materials showed panchromatic absorption with onset up to 1000 nm depending upon the mixture stoichiometry. The extraordinary stability and excellent absorption properties make them suitable materials for photoelectrochemical water-splitting applications. PEC studies of these series of materials showed a high water oxidation photocurrent of 0.56 mA cm^{-2} for $Cs_2Ru_{0.5}Pt_{0.5}Cl_6$. Fundamental investigation from photoelectrochemical reactions revealed that the intrinsic ruthenium-based VOP showed enhanced hole transfer to the electrolyte, while the intrinsic platinum-based VOP showed higher photovoltage. The mix of these end members at the tetravalent site showed a synergic effect of reduced charge transfer resistance from the material to the electrolyte and increased photovoltage, which led to increased PEC performance of the intermediate materials.

Keywords : solar water splitting, photo electrochemistry, photo absorbers, material characterization, device characterization, green hydrogen
Conference Title : ICSHEST 2024 : International Conference on Solar-Hydrogen Energy Systems and Technologies
Conference Location : New York, United States
Conference Dates : April 22-23, 2024