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Photoelectrochemical characterization of lead-based hybrid perovskite semiconductors

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A variety of PbI₂/MAPbI₃ perovskites were prepared and investigated by a rapid screening technique utilizing a modified scanning electrochemical microscope (SECM) in order to determine how excess PbI₂ affects its photoelectrochemical (PEC) properties. An optimum ratio of 2.5% PbI₂/MAPbI₃ was found to enhance photocurrent over pristine MAPbI₃ on a spot array electrode under irradiation. With bulk films of various PbI₂/MAPbI₃ composites prepared by a spin-coating technique of mixed precursors and a one-step annealing process, the 2.5% PbI₂/MAPbI₃ produced an increases photocurrent density compared to pristine MAPbI₃ for 2mM benzoquinone (BQ) reduction at -0.4 V vs Fc/Fc+. As a result of the relatively high quantum yield of MAPbI₃, a time-resolved photoluminescence quenching experiment could be applied to determine electron-hole diffusion coefficients and diffusion lengths of PbI₂/MAPbI₃ composites, respectively. The diffusion coefficients combined with the exciton lifetime of the pristine 2.5% PbI₂/MAPbI₃ ($\tau_{\text{PL}} = 103.3$ ns) give the electron and hole exciton diffusion lengths, ~ 300 nm. Thus, the 2.5% PbI₂/MAPbI₃ led to an approximately 3.0-fold increase in the diffusion length compared to a previous report of ~ 100 nm for the pristine MAPbI₃ perovskite. We then demonstrated that the efficiency of liquid-junction solar cells for 2.5% excess PbI₂ of p-MAPbI₃ was improved from 6.0% to 7.3%.

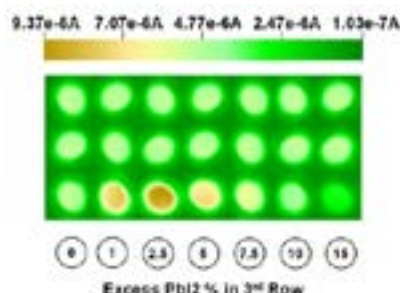


Figure 1. SECM images for a typical photocurrent response of PbI₂/MAPbI₃ composites under irradiation. The color represents the measured photocurrent shown in the scale bar above the SECM image. The first and second seven-spot rows are pure MAPbI₃ perovskites. The third seven-spot row represents the amount of excess PbI₂ in each spot in the array electrode. The photocurrent shown is for 2 mM BQ reduction with 0.1M TBAPF₄ supporting electrolytes in CH₂Cl₂ measured at an applied potential of -0.4 V vs Fc/Fc⁺.

Biography

Sam H. Y. Hsu's research interests involve the material design, synthesis, processing, imaging, spectroscopy and solar energy application, aiming to explore fundamental properties and interactions of hybrid perovskite semiconductors and functional metallopolymer materials for developing efficient solar energy conversion processes. He has keen interests in photoinduced charge transfer processes, interfacial electron transfer, electrochemical hydrogen generation, and photoredox reactions for photovoltaics and solar fuel production. The investigations between material phenomena rely heavily on concepts and techniques of material and physical engineering, consisting of photophysics, electrochemistry, photoelectrochemistry, scanning photoelectrochemical microscopy imaging, ultrafast transient absorption and time-resolved photoluminescence spectra.

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