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Probing and controlling carrier concentration in complex semiconductor powders for solar water splitting - optical and structural studies

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The $(GaN)_{1-x}(ZnO)_x$ system is one of the most promising semiconductor systems discovered to date for the renewable production of hydrogen fuel through solar water splitting driven by visible light. However, the maximum $(GaN)_{1-x}(ZnO)_x$ quantum efficiency observed for this process is still about an order of magnitude lower than expected for a fully optimized system. Optimization has been hindered by the lack of the fundamental information about this complex semiconductor system, which has almost exclusively been studied in powder form due to the severe challenges involved in preparing thin films or single crystals of $(GaN)_{1-x}(ZnO)_x$. Two thrusts to better understand this material will be described. First, we have developed a new general self-referenced methodology for determining the refractive index and then the absolute absorption coefficient through diffuse reflectance measurements on loose powder samples, as opposed to the crystalline materials or dense pellets required for conventional approaches. These methods suggest that despite having a direct gap optical transition, the absolute absorption coefficient of $(GaN)_{1-x}(ZnO)_x$ powders is about an order of magnitude lower than the best visible light absorbing systems. Second, we have attacked the challenge of controlling the semiconductor concentration in this complex semiconductor system. We demonstrate that the infrared optical response of loose powder samples can be used to very effectively monitor the carrier concentration of this semiconductor system. Using this probe, we demonstrate suitable protocols for tuning the carrier concentration and provide insights into the mechanism by the carrier concentration is changed.