Co-catalyst assisted mesoporous II-VI metal sulfide nanocrystal assemblies for highly efficient photochemical water-splitting and hydrogen production

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Photocatalytic water splitting for hydrogen (H₂) production has been one of the hot subjects in recent decades as a result of the global energy crisis. Key element of this approach is the development of cost-effective functional catalysts with high activity and long-term stability.[1] To that end, II-VI metal-sulfide colloidal nanocrystals (NCs), such as CdS and ZnS, have attracted enormous research attention because of their interesting optical, electronic and catalytic attributes.[2] However, conventional metal sulfides suffer from low charge-carrier separation and poor chemical stability that restrain their photocatalytic efficiency. A well-established approach to overcome these limitations and achieve improved catalytic performance is to employ functional co-catalysts and create heterostructures with open-pore architecture at the nanoscale.[3,4] Here, we report the design and fabrication of two novel mesoporous materials comprised of CdS and CdS/ZnS nanocrystal assemblies (NCAs) and demonstrate their photocatalytic performance for H₂-generation from water. The resultant materials consist of a highly porous network (BET surface area up to 260 m² g⁻¹) of linked ~5 nm-sized metal-sulfide NCs that is perforated by uniform mesopores (ca. 6 nm in size). When coupled with low-cost active co-catalysts, such as nickel hydroxide or nickel phosphides, the formed mesoporous heterostructures exhibit a remarkable photocatalytic H₂-production activity associated with a quantum yield (QY) up to ~70% at 420 nm. Photocatalytic experiments combined with UV-vis/NIR, photoluminescence and electrochemical impedance spectroscopy studies suggest that the outstanding photocatalytic performance of these catalytic systems manly arises from the accessible 3D open-pore structure and the favorable band-alignment of metal sulfide with the respective co-catalyst, which suppresses carrier recombination and promotes efficient chargetransport at the junction interfaces.

References:

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