Design Principles of High-Entropy Sulfides: From Phase Selection to Universal Cocatalysis in Semiconductor Photocatalysts

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High-entropy materials have emerged as a new frontier in materials chemistry, where multiple principal elements are incorporated into a single crystal lattice to create unique physicochemical properties. Among them, high-entropy sulfides (HESs) are particularly appealing for applications in energy conversion and catalysis, owing to their tunable electronic structures, rich catalytic active sites, and enhanced structural stability[1–2]. Understanding how phase structures are selected in such complex systems is therefore a fundamental challenge with direct implications for materials design.

While work on HESs have date mostly focused on electrocatalytic applications, we furthermore here explore their performance in photocatalytic hydrogen evolution reaction (HER) for water splitting. In particular, we demonstrate that the DTC-route readily facilitates formation of nanoscaled HES coatings as HER co-catalysts on temperature-sensitive CdS and other semiconductors with different chemical compositions, crystal structures, and electronic properties. These HES@semiconductor hybrids outperform reference sulfide hybrids for extended photocatalytic HER, which we suggest to be related to emerging HES-based cocktail and sluggish diffusion effects.

Combined, our work systematically elucidate the universality of high-entropy sulfides as cocatalysts and phase selection mechanism of them from facile, low-temperature DTC-based synthesis.

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