Tuning interfacial energetics for enhanced H_2O_2 generation

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Solar-driven H_2O_2 generation over a particulate photocatalyst has attracted much attention. Yet, the efforts in designing high-performance particulate photocatalysts are largely impeded by inefficient charge separation. Because charge separation in a particulate photocatalyst is driven by asymmetric interfacial energetics between its reduction and oxidation sites, enhancing this process demands nanoscale tuning of interfacial energetics on the prerequisite of not impairing the kinetics and selectivity for H_2O_2 generation. In this study, we realized this target with a general strategy involving the application of a core/shell type cocatalyst that is demonstrated on various photocatalytic systems. Particularly, this strategy was highlighted on a $BiVO_4$ system for overall H_2O_2 photosynthesis. A core/shell type Ag/Pd cocatalyst was selectively deposited on the reduction facets of $BiVO_4$, where the Ag core formed a low Schottky barrier with $BiVO_4$ at its reduction site for enhancing charge separation and Pd shell preserved the surface kinetics and selectivity for H_2O_2 generation (Figures 1a-1d). Time-resolved spectroscopy and numerical simulations suggest the $BiVO_4$/Ag junction enhanced the asymmetric interfacial energetics as expected. With successful interfacial energetics tuning, $BiVO_4$ exhibits high overall H_2O_2 photosynthesis among inorganic photocatalysts, with an apparent quantum yield (AQY) of 3.0% and a solar-to-H_2O_2 conversion (STH) efficiency of 0.73% at full spectrum, as well as an AQY of 13.1% at 420 nm (Figures 1e-1f).

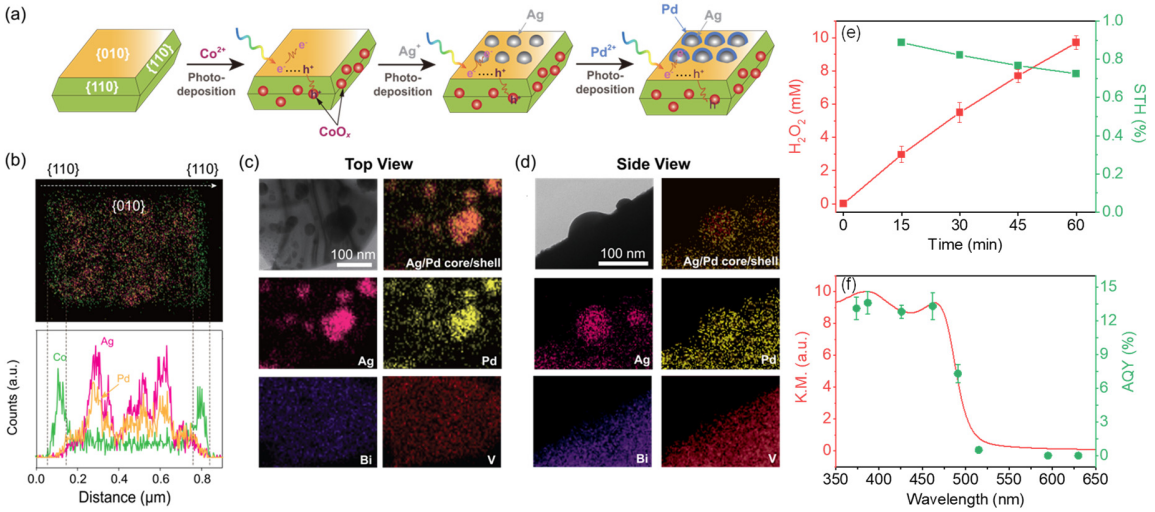

Figure 1 (a) Stepwise and facet-selective photodeposition of Co, Ag and Pd on $BiVO_4$. (b) EDS elemental mapping and line profile of CoO_x/$BiVO_4$/(Ag/Pd). (c)-(d) STEM-EDS elemental mapping of Ag/Pd particles loaded on $BiVO_4$. (e) Time courses of photocatalytic H_2O_2 generation over CoO_x/$BiVO_4$/(Ag/Pd) and the corresponding STH efficiency. (f) AQY of H_2O_2 photosynthesis over CoO_x/$BiVO_4$/(Ag/Pd) as a function of the incident light wavelength.

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[2] T. Liu, Z. Pan*, K. Kato, J.J.M. Vequizo, R. Yanagi, X. Zheng, W. Yu., A. Yamakata, S. Hu, K. Katayama, C. Chu*, A general interfacial-energetics-tuning strategy for enhanced artificial photosynthesis, *Nat. Commun.*, Accepted.