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Title: Promising visible-light driven hydrogen production from water on a highly efficient CuCo2S4

nanosheet photocatalyst†

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CuCo2S4 Nanosheet Photocatalyst

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Abstract:

Here we report the development of CuCo2S4 nanosheets (NSs) as a promising semiconductor photocatalyst for the first time for water splitting reactions under visible light ($\lambda \ge 420$ nm) conditions, without the support of any noble metal co-catalyst. These NSs were produced via a simple hydrothermal route and have desirable properties with a band gap of 2.24 eV, and are photo-catalytically active under visible light with an apparent quantum yield (AQY) of 2.48%. Under visible light, CuCo2S4 NSs exhibit excellent weight-normalized photoactivity that generates ~25 900 µmol h-1 H2 for 1 g of material with sulphide + sulphite as the sacrificial agent under 7.68 mW cm-2 illumination, which is the best evolution reported for any chalcogenide semiconductor material without any co-catalyst to date with unprecedented long-term operational stability (up to 12 h study time). The rate and number of hydrogen gas molecules produced are $8.2855 \times 1015 \text{ s-1} \text{ cm-2}$ which remained constant for three catalytic cycles with a turnover frequency (TOF) value of 0.017 s-1. The effect of Cu substitution on photoactivity was also investigated for comparative studies and it was found that CuCo2S4 NSs show superior activity to Cu0.5Co2.5S4 and Co3S4. These CuCo2S4 NSs absorb the entire visible range of the spectrum from 420 to 800 nm, and have a highly populated density of states at the Fermi level and a high donor concentration of 7.22 × 1018 cm-3 which have been evaluated by Mott-Schottky analysis and favourable adsorption of H+ on S-sites and conversion to H2 corroborate their efficient photocatalytic activity.

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