Efficient charge separations in optimally oxidized  $Cu_xZn_{1-x}S$  photocatalysts for enhanced solar  $H_2$  production

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Solar water splitting is a attractive way of producing hydrogen from the renewable natural resources, and heterostructure photocatalysts have been widely investigated in photocatalytic applications. In this work, flower-shaped  $Cu_x Zn_{1-x}S$  composite photocatalysts were prepared with various copper contents and then the composites were further treated under controlled oxygen concentrations in high temperature furnace. The efficient photo-generated charge transfer was conducted by introduction of oxide materials which constructed the Z-scheme assisted type-II heterosystem. The maximum hydrogen production rate was acheived as 595  $\mu$ mol/g/h from the optimally oxidized  $Cu_{0.05}Zn_{0.95}S$  photocatalyst. This could be mainly attributed to the highest  $Cu_2O$  crystalline phase fraction as confirmed by XRD measurement. High light absorption and low charge recombination in hetero-system were also crucial. Therefore, overall photocatalytic efficiency of the oxidized composite photocatalysts can be enhanced by optimizing their atomic contents and crystal phase fractions.