Development of gold nanoparticle-decorated graphitic carbon nitride photocatalysts for solar hydrogen production

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WHY HYDROGEN?

- High specific energy
- Chemical fuel with no CO₂ emissions
- Water is the only by-product
- Easy storage
- On-demand energy supply
- Transportable
- Through pipelines, tanks
- Current hydrogen production is mostly fossil-fuel based, leading to large amounts of greenhouse gas emissions.

MOTIVATION

CONCEPT

Photocatalytic water splitting is a method for producing hydrogen from water. It uses the free and renewable energy of sunlight to split water molecules into hydrogen and oxygen, with the help of a semiconductor photocatalyst. Graphitic carbon nitride (g-C₃N₄) is a low cost photocatalyst, which when loaded with gold nanoparticles, is very promising for H₂ generation.

GOAL

Study the effect of gold loading on g-C₃N₄ performance for H₂ production

RESULTS

- Intense photoluminescence peak for pure g-C₃N₄
- Strong radiative relaxation of photoexcited electrons from the conduction band to the valence band of the semiconductor
- Peak intensity decreases as a function of Au loading
- Reduction of electron-hole pair recombination and enhancement of charge separation

OPTICAL PROPERTIES

- Graphitic carbon nitride is yellow, resulting from strong light absorption (photoexcitation) at wavelengths below 450 nm
- Spherical Au nanoparticles are characterized by a localized surface plasmon resonance (LSPR) feature at ~550 nm
- The LSPR is a collective oscillation of valence electrons on the metal nanoparticles
- The LSPR feature intensified with the gold loading, validating our synthesis method

ELECTRONIC PROPERTIES

- The size of the gold nanoparticles increases in function of the Au loading
- Good dispersion of the gold nanoparticles over the g-C₃N₄ nanosheets

TRANSMISSION ELECTRONIC MICROSCOPY

- The gold nanoparticle loading strongly influences the performance of Au/g-C₃N₄ photocatalysts for H₂ production. A trade-off exists between light absorption by g-C₃N₄ and electron capture by Au for H₂ evolution. A gold nanoparticle loading of 2 wt.% is near optimal.

HYDROGEN PRODUCTION ACTIVITY

- 2 wt.% Au was found to be the optimum gold loading for photocatalytic hydrogen production in 10 vol.% TEOA under visible light excitation (λ > 420 nm)
- 43 times more efficient than a g-C₃N₄ sample that does not contain gold

Why does the hydrogen production rate decrease at gold loadings above 2 wt.%?

At higher loadings, a greater portion of g-C₃N₄ surface is covered by gold, thus inhibiting light absorption and creation of electron-hole pairs in the underlying semiconductor. As a result, less charges are available to react with water and TEOA.

 /** References: */

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