

Experimental section

1. Materials

Ethyl orthosilicate (TEOS, AR, $\geq 99.9\%$), ammonia (NH_4OH , 35%), octadecyltriethoxysilane (C18-TMOS, AR, $\geq 99.9\%$), cyanamide (NH_2CN , 50% aqueous solution), indium chloride ($\text{InCl}_3 \cdot 4\text{H}_2\text{O}$, AR, $\geq 99.9\%$), zinc chloride ($\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, AR, $\geq 99.9\%$), and thioacetamide (TAA, AR, $\geq 99.9\%$) were purchased from Aladdin Reagent (Shanghai) Co. Ltd, China. Hydrochloric acid (HCl , 37%) and ammonium hydrogen fluoride (NH_4HF_2 , CP, $\geq 99.5\%$) were purchased from Shanghai Maclean Biochemical Technology Co. Ltd, China. High-purity nitrogen is supplied by Kunming Deli Gas Co. Ltd, China, were bought from Tianjin Kemiou Chemical Reagent Company. All chemicals were sourced from commercial suppliers and were not further purified before use. Deionized water was used throughout the experiments.

2. Catalyst Characterization

The powder X-ray diffraction (XRD) patterns were acquired on an X-ray diffractometer (Rigaku, Japan) with $\text{Cu-K}\alpha$ irradiation ($\lambda = 1.5418 \text{ \AA}$) to determine the phase structure of the prepared photocatalysts. The transmission electron microscope (TEM, JEM-2100, Tokyo, Japan) and scanning electron microscope (SEM, Nova nano SEM 450, Oregon, USA) were used to examine the microstructure and morphology of the photocatalysts, and the element distribution was detected by TEM equipped with an energy dispersive spectrometer (EDS). X-ray photoelectron spectroscopy (XPS) was performed by a photoelectron spectrometer (Thermo ESCALAB 250Xi, Massachusetts, USA) with an $\text{Al K}\alpha$ radiation source ($h\nu = 1486.6 \text{ eV}$). The ultraviolet-visible (UV-Vis) diffuse reflectance spectra of the catalysts were measured by a spectrophotometer (UV-2600i, Shimadzu, Japan) equipped with an integrating sphere accessory (ISR-2600Plus), using BaSO_4 as the reflectance standard at room temperature, in the wavelength range of 200-900 nm. The photoluminescence (PL) spectra of the photocatalysts were measured with an FLS 1000 fluorescence spectrophotometer (Edinburgh Instruments, Livingston, Scotland, UK). The excitation wavelength was 375 nm, the scanning speed was 1200 nm min^{-1} , and the widths of the excitation slit and emission slit were both 2.5 nm. The work function of the samples was tested by a Scanning Kelvin probe (SKP) (SKP5050 system, Edinburgh, Scotland). The temperature of the sample was measured using Testo 865 infrared thermography.

3. Photocatalysis Experiment

3.1. Photocatalytic degradation

The photocatalytic pollutant degradation reactions were carried out on a multi-purpose photochemical reaction system (CEL-LAB500E4, Education Au-light Co., Ltd., Beijing, China). About 50 mg of the photocatalyst was added to 100 mL of a solution containing OTC (10 mg/L). Before the photocatalytic experiments, the solution containing the pollutant and the photocatalyst was placed in a dark room for 30 min to obtain the adsorption-desorption equilibrium. Then, the solution was irradiated under a 300 W xenon lamp with a circulating water jacket (CEL-HXF300, Education Au-light Co., Ltd., Beijing, China). Every 15 min, 3 mL of each liquid sample was removed from the beaker and filtered with

0.22 μm Millipore filter heads. Concentrations were subsequently tested using a high-performance liquid chromatograph (LC-3100, Dalian, China).

3.2. Photocatalytic H_2 evolution

The photocatalytic hydrogen evolution reaction was performed on the all-glass automatic online trace gas analysis system (Labsolar-6A, Beijing Perfect Light Technology Co., Ltd.). With Labsolar-6A, a 300W Xenon lamp (Microsolar300, Beijing, Perfect Light Technology Co., Ltd., light intensity: 200 mW/cm^2) was used as the simulated sunlight spectral source. The as-prepared catalyst (10 mg) was uniformly dispersed by using a

magnetic stirrer in 120 mL of methanol solution (containing H_2O /methanol, $v/v=90:30$). The temperature of the reaction was kept at 298 K by cool, flowing water. Prior to the

photocatalytic water splitting reaction, the sealed system was completely degassed to remove dissolved air in the aqueous solution by purging with high-purity Ar gas for 0.5 h. During the irradiation process, a hydrogen sample (0.5 mL) was extracted from the reactor at a given interval, and the amount of hydrogen produced was analyzed by an online gas chromatograph (GC7900, Shanghai, China). For the recycling experiment, the photocatalyst powder was dispersed into 120 mL of methanol solution (25 vol.%), then the sealed system was purged with high-purity Ar gas to remove the dissolved air in the solution. During the continuous irradiation of 5 h in each cycle, a gaseous sample (0.5 mL) was extracted from the reactor at a given interval of 1 h to analyze the H₂ yield by using a gas chromatograph with a thermal conductivity detector. After finishing one cycle, the sealed system needed to be degassed to remove the generated H₂. Meanwhile, the cooling water was used to maintain the temperature at 298 K.

3.3. Photoelectrochemical measurement

The electrochemical measurement was carried out by a standard three-electrode system. 50 mg as-prepared sample was dispersed in 35 mL of ethanol and then spread uniformly on indium-tin-oxide (ITO) conductor glass as the work electrode, while Ag/AgCl as the reference electrode, Pt foil as the counter electrode, and 1.0 M Na₂SO₄ aqueous solution as the electrolyte. The transient photocurrent of the sample was obtained by multiple on/off visible light irradiation, with a period of 20 s. Electrochemical impedance spectroscopy was measured with an amplitude of 5 mV and frequencies varying from 0.01 to 10000 Hz.

3.4. Photothermal test

The photothermal test of the as-prepared samples was carried out as follows: About 0.2 g of sample was laid out flat on white weighing paper (**Figure S3**), and the initial temperature was controlled at room temperature. The temperature of the sample was measured using the Testo 865 infrared thermography. A 300 W xenon lamp (CEL-HXF300, Beijing China Education Au-light Co., Ltd., Beijing, China) was used as the light source for all the photothermal experiments, with a distance of approximately 30 cm between the light source and the sample.

Instruction: The photothermal tests were carried out in the winter (2022.08.12-2022.10.13) in Kunming (24°49'N and 102°51'E). The temperature during this time is about 22 °C and the water temperature is about 15 °C.

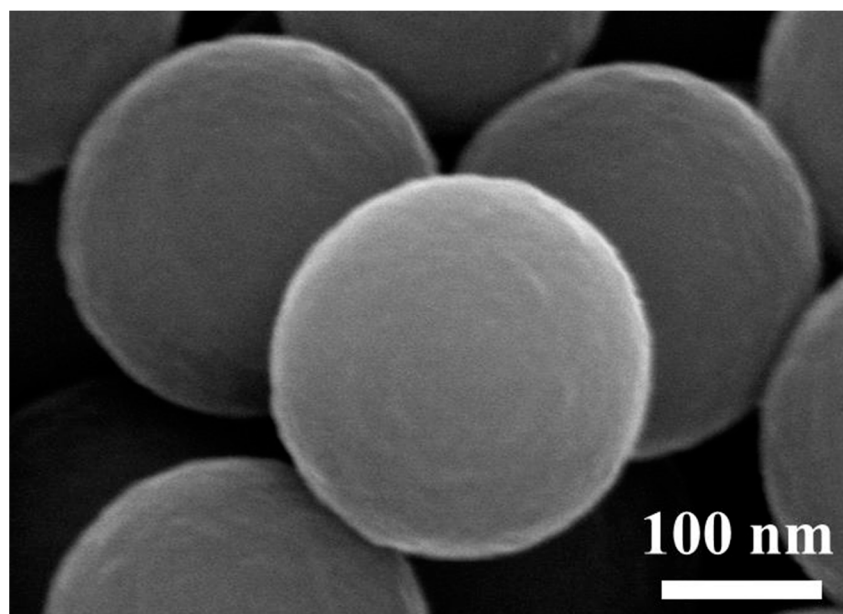


Figure S1. SEM image of the SiO₂ spheres.

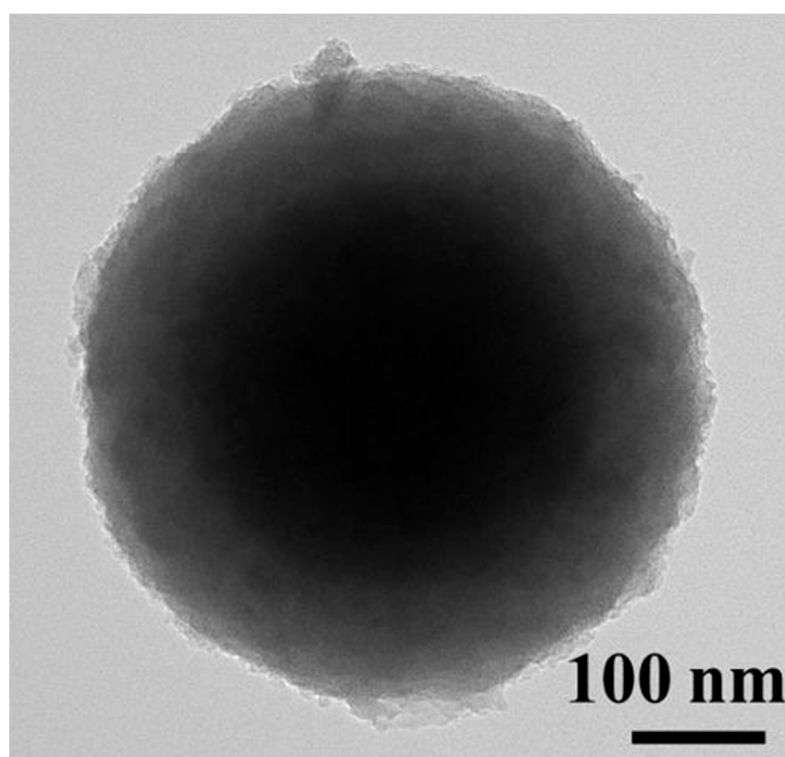


Figure S2. TEM image of SiO₂@g-C₃N₄.

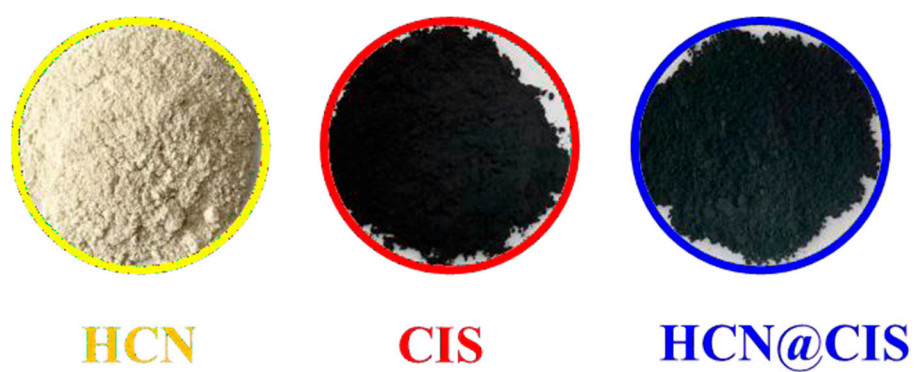


Figure S3. The sample patterns of HCN, CIS, and HCN@CIS.

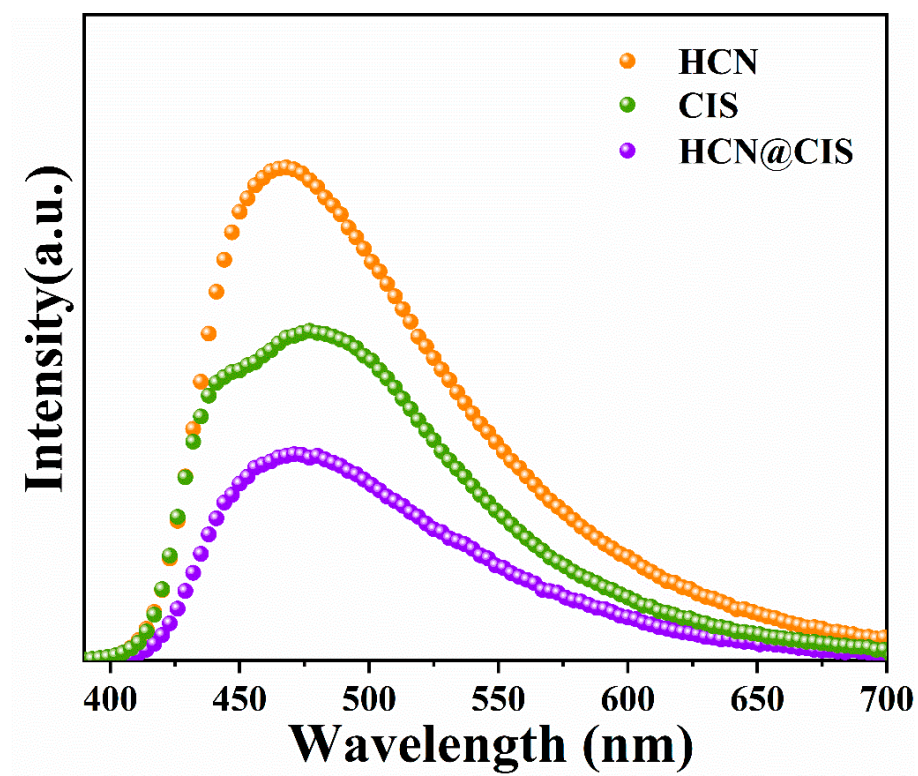


Figure S4. Steady-state PL emission spectra.

Table S1. Comparison of H₂ generation rates of different photocatalysts.

Photocatalysts	Light source	H ₂ evolution rate ($\mu\text{mol g}^{-1} \text{ h}^{-1}$)	Ref.
g-C₃N₄@ Cu_{0.5}In_{0.5}S	Xe lamp (300W, $\lambda > 420$ nm)	2325	This work
MoS ₂ /CdS/g-C ₃ N ₄	300 W Xe lamp ($\lambda > 420$ nm)	956	[62]
KCCN	Xe lamp (300 W)	557	[63]
Pd@TiO ₂ @ZnIn ₂ S ₄	Xe lamp (300 W)	2190	[64]
Au/TiO ₂ (P25)-gC ₃ N ₄	Hg lamp (150W, $\lambda > 420$ nm)	419	[65]
ZnIn ₂ S ₄ /Ti ₃ C ₂	Xe lamp (300 W, $\lambda > 420$ nm)	979	[66]
g-C ₃ N ₄ /WO ₃ -carbon microsphere	Xe lamp (300W, $\lambda > 420$ nm)	2500	[67]
Hollow core-shell TiO ₂ /g-C ₃ N ₄	Xe lamp (300 w)	809	[68]