Photodecomposition of H_2S to Produce H_2 over CdS/Al-MCM-41 Composite Catalyt

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Introduction

H_2S is a type of by-product obtained from oil refinery, natural gas processing and other chemicals production. Till now, the well-established Claus process has been applied to deal with H_2S. It involves partial oxidation of H_2S to sulfur and water where hydrogen is wasted. If hydrogen can be recycled from H_2S instead of being directly oxidized into water, an immense amount of H_2 would be regenerated. The photocatalytic decomposition H_2S to H_2 over semiconductor photocatalysts is considered as a process to convert solar energy into fuels and the process requires less energy than the photodecomposition of water[1]. CdS is suitable for the visible light and used widely to catalyze photochemical reaction. But CdS is very unstable against photocorrosion in aqueous solutions under irradiation. There are many attempts to overcome the photocorrosive nature of CdS. Loading systems with large surface area , high adsorption capacity and strong cation exchange properties, have been considered as a promising method to offer prevent electron-hole recombination and improve the photocatalysis efficiency and photostability of CdS, Mesoporous Al-MCM-41 (named Al-M41) has the well-defined crystalline structures with these properties. It could provide the place for assembling small semiconductor clusters, thereby producing nanocomposite materials[2~3]. In this paper, a CdS loaded on Al-M41 photocatalyst (CdS/Al-M41) had been prepared and used for photocatalytic H_2 evolution to explore the possibility in improving the photocatalysis efficiency.

Materials and Methods

Al-M41 with the SiO_2/Al_2O_3 ratio of 25 was carried out Cd^{2+} ion-exchange twice in 0.5 mol/l $Cd(NO_3)_2$ solution. As-prepared sample was filtered, washed, and then dired in air. The above sample was sulfurizeda by addition of 0.5 mol/l Na_2S solution at 353K for 2 h. The obtained sample was filtered, washed and calcined at 923K for 1 h under N_2 to get CdS /Al-MCM-41 catalyst (named as CdS/Al-M41). The content of CdS in CdS/Al-M41 is 6.541 wt% by ICP analysis.

UV-Vis diffuse reflectance spectra (UV-Vis) was recorded on a scanning spectrophotometer (UV-2450, Shimadzu, Japan) equipped with an integrating sphere (ISR-240A). The amount of CdS loaded on CdS/Al-M41 catalyst was measured by inductively coupled plasma emission spectrometer (ICP-AES, Thermo Elemental IRIS Intrepid II XSP).

The photocatalytic reaction was carried out in an inner-irradiation type reactor using a 250 W high-pressure lamp as the irradiation source under visible light irradiation by an inner circulating system containing 1 mol/l $NaNO_2$ (cut off λ<400nm). 0.2 g CdS, CdS/Al-M41 or Al-M41 was suspended in 300 ml aqueous solution containing 0.7 mol/l NaOH and 0.25 mol/l Na_2SO_3 with pumping in some hydrogen sulfide gas for preparing different pH value.

Results and Discussion

From the UV-Vis spectroscopy in Fig.1, it is known that the adsorption edge of CdS/Al-M41 shows a blue shift compared to that of bulk CdS, which indicates that CdS nanoparticles have formed inside the Al-M41 pores. The H_2S photocatalytic decomposition reactions were carried in the reaction media with pH value of 9.0 over CdS and CdS/Al-M41 respectively. The results showed that H_2 evolution rate over CdS/Al-M41 (30.55*mmol/h·g*) was about 57 times higher than that over bulk CdS (0.536 *mmol/h·g*). The above improved H_2 production result is ascribed to the increase of the special surface area of CdS/Al-M41 and the nano-structure of CdS with no internal electric field.

Fig.2 was the H_2 evolution rates in reaction media with different pH value over CdS/Al-M41. It was shown from Fig.2 that the highest H_2 evolution rate appears at pH value of 9.0, which providing for the possibility of H_2 evolution from H_2S continuous photocatalytic decomposition reaction.

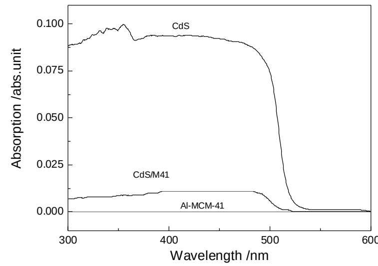

Fig.1 The UV-Vis spectroscopy of CdS, CdS/Al-M41 and Al-M41

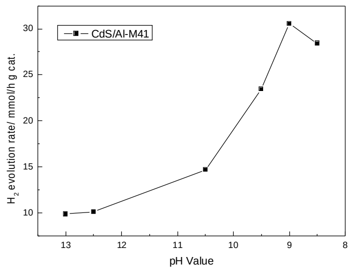

Fig. 2 Effect of pH value on H_2 evolution rate over CdS/Al-M41

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