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Preparation of Cu-doped $Cd_{0.1}Zn_{0.9}S$ solid solution by hydrothermal method and its enhanced activity for hydrogen production under visible light irradiation

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ABSTRACT

A series of Cu-doped $Cd_{0.1}Zn_{0.9}S$ solid solution with various amounts of Cu dopant was successfully prepared by hydrothermal method. The properties and the photocatalytic activity of the prepared samples for hydrogen production under visible light irradiation were compared to those prepared by co-precipitation method. The Cu-doped $Cd_{0.1}Zn_{0.9}S$ samples prepared by hydrothermal method showed both improved crystallinity and photoabsorption ability as compared to the undoped sample. On the other hand, even though Cu-doped $Cd_{0.1}Zn_{0.9}S$ prepared by co-precipitation method also showed improved photoabsorption ability in the visible light region, the samples showed poor crystallinity compared to the undoped one. With the same amount of Cu dopant, all samples prepared by hydrothermal method were found to exhibit higher photocatalytic activity for hydrogen production than the samples prepared by co-precipitation method. It was revealed that the amount of Cu dopant, crystallinity and narrow band gap energy are important factors to obtain highly active and stable photocatalysts.

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1. Introduction

Semiconductor photochemistry is one research field that plays important role in various photocatalytic applications. As example, photocatalytic hydrogen production from water on a semiconductor has been a great interesting challenge for researchers to convert solar energy into clean and renewable hydrogen energy. In order to utilize solar energy efficiently, development of visible light active semiconductor is crucial. For such reason, photochemistry of the semiconductor such as band structure must be controlled. Proper conduction band position is important to provide high potential energy for reduction of water to hydrogen. Two possible methods which are most commonly used to control band structure are doping with foreign elements and forming solid solution between wide and narrow band gap semiconductors [1–9].

One of the most extensively studied sulfides solid solutions is $Cd_xZn_{1-x}S$ due to its controllable band structure and excellent performance under visible light irradiation [1–5]. Although $Cd_xZn_{1-x}S$ was successful to overcome shortcomings of photocorrosion from CdS and wide band gap energy of ZnS, the band gap energy of this solid solution was still large, thus limited the utilization of photon energy for hydrogen evolution under visible light irradiation. Recently, doping of $Cd_xZn_{1-x}S$ solid solution with various metal ions have been reported, such as Cu [10–13] and Ni [14,15] to further improve the efficiency of $Cd_xZn_{1-x}S$. It was reported that Cu^{2+} and Ni^{2+} can contribute to the enhancement in the visible light response of the $Cd_xZn_{1-x}S$ photocatalyst by reducing the band gap energy [10–15].

In addition to the band gap engineering, the method to prepare a photocatalyst itself is very significant since it can affect the properties, thus affect the photocatalytic activity. Some studies revealed that different preparation methods lead to different photocatalytic activity due to the differences in the physical chemical properties [16,17]. Even though prepared by the same co-precipitation method, $Cd_{1-x}Zn_xS$ annealed at different temperatures showed different photocatalytic activities for hydrogen evolution [16]. Recently, we found that $Cd_{0.1}Sn_xZn_{0.9-x}S$ photocatalysts prepared by hydrothermal method showed higher photocatalytic activity and stability compared to the ones prepared by co-precipitation method [17].

On the other hand, some research groups have reported the preparation of Cu-doped $Cd_{0.1}Zn_{0.9}S$ series by co-precipitation method [10–13]. However, other method to prepare Cu-doped $Cd_{0.1}Zn_{0.9}S$ series has never been reported yet. Therefore, there are lack of alternative preparation methods as well as comparison studies on properties and photocatalytic performance of the photocatalysts prepared by different methods. In the present paper, we demonstrate that Cu-doped $Cd_{0.1}Zn_{0.9}S$ can be prepared by hydrothermal method for the first time, and the prepared samples have much higher activity than those prepared by co-precipitation method for hydrogen production under visible light irradiation. Based on the comparison study between properties

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