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学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　博士（理学）　氏名　郭　建君

学 位 論 文 題 名

Design and Construction of Ag_3PO_4-based Composite Photocatalysts for Environmental Purification
（環境浄化のための Ag_3PO_4 を基盤とする複合光触媒の設計と構築）

Photocatalytic degradation of environmental pollutants has attracted increasing attention because it is a promising, environmental, and cost-effective technology. For the better utilization of visible light accounting for about 43% of solar energy, visible-light-sensitive photocatalysts are urgently anticipated. Silver orthophosphate (Ag_3PO_4) as a novel photocatalyst exhibits surprisingly high efficiency under visible light irradiation. The quantum yield of Ag_3PO_4 in water oxidation is nearly 90% at wavelength around 420 nm. It has a promising future in photocatalytic applications. However, pure Ag_3PO_4 exhibits low efficiency in photocatalytic degradation of gaseous pollutants and superior activity in photodecomposition of organic dyes is still needed. The recombination of photo-generated electron-hole pairs, the redox potential of generated electron, and the surface status of Ag_3PO_4 are considered as factors that affect its photocatalytic performances. In this regard, the research target of this work is to understand the effect of electronic structures and surface properties on photocatalysis and to improve the generation and separation of photo-generated electron-hole pairs towards developing highly efficient Ag_3PO_4—based photocatalysts for environmental purifications. To achieve this target, three novel composite photocatalysts based on Ag_3PO_4 were successfully synthesized and investigations of the relationships among electronic structures, surface properties and photocatalytic performances were carried out. This dissertation is divided into five chapters:
In chapter 1, an overview of the photocatalysis and the semiconductor photocatalysts was introduced firstly. The synthesis and utilization of composite photocatalysts were summarized with details to provide useful information for developing efficient composite photocatalysts for environmental purification. More importantly, the synthesis of Ag_3PO_4 and Ag_3PO_4-based composite photocatalysts, and strategies of achieving high photocatalytic activities were also summarized.
In chapter 2, a novel composite photocatalysts $Ag_3PO_4/In(OH)_3$ with adjustable surface-electric property for efficient photodegradation of organic dyes was obtained via an in-situ precipitation method. The surface-electric property of the composite photocatalyst was continually adjusted and introduced into the photocatalytic degradation of Rhodamine B (Rh B) in water. It was found that more negative-charged surface of the composite photocatalysts is advantageous for adsorption of organic dyes. Therefore, the surface-electric property is closely related to the photocatalytic activity. The apparent rate constant of the $Ag_3PO_4/In(OH)_3$ (molar ratio=1.65:1) is 5 times higher than that of pure Ag_3PO_4. Moreover, the photosensitization and the intrinsic photocatalytic degradation of Rh B were also investigated under 540 ± 13nm and 420 ± 13nm monochromatic irradiations, respectively. It was revealed that the

intrinsic photocatalytic pathway dominates the whole degradation process. Here, the composite photocatalysts with adjustable surface-electric property and suitable band structure reveal a novel material design-concept of exploiting a new photocatalyst based on the reaction kinetics and thermodynamics.

In chapter 3, a new heterojunction Ag_3PO_4/Cr-$SrTiO_3$ photocatalyst towards efficient elimination of gaseous organic pollutants under visible light irradiation was successfully synthesized. Iso-propanol (IPA) photodegradation over the heterojunctions under visible-light irradiation was employed for evaluating their photocatalytic properties. The highest activity was observed in the Ag_3PO_4/Cr-$SrTiO_3$ heterojunction with the mass ratio of 1:4 (Ag_3PO_4: Cr-$SrTiO_3$) sintered at 500 $^\circ$ C. The evolution rate of CO_2 over the composite is about 33 times higher than that of pure Ag_3PO_4. Moreover, the valence-band state of Ag_3PO_4 and Cr doped $SrTiO_3$ were analyzed by X-ray photoelectron spectroscopy (XPS) to deduce the band structures, which is helpful for understanding the transfer of photocarriers between the two semiconductors. Results reveal that electronic structures of composite photocatalyst promote the generation and separation of photo-generated electron-hole pairs and the multi-photon reactions. Therefore, composite photocatalyst showed much higher activity than the individual materials.

In chapter 4, another new composite photocatalyst Ag_3PO_4/$Sr_2Nb_2O_7$-xNx with adjustable band structures towards efficient elimination of gaseous organic pollutants under visible light irradiation was obtained. The N 2p orbitals have higher potential energies than that of O 2p orbitals, and their hybridization lifts up the top of the valence band of $Sr_2Nb_2O_7$. Therefore, the N-doped $Sr_2Nb_2O_7$ can absorb visible light. More importantly, the electronic structure of $Sr_2Nb_2O_7$-xNx could be adjusted continually by nitridation with NH_3 under different temperatures. The relationship between electronic structure and photocatalytic activity was also systematically investigated. The best activity was obtained over the Ag_3PO_4/$Sr_2Nb_2O_7$-xNx (x = 0.82) nitrided at 750 $^\circ$ C with band gap is about 2.15 eV. The evolution rate of CO_2 over the composite is about 40 times higher than that of pure Ag_3PO_4. Investigations of energy-band structure via valence-band X-ray photoelectron spectrum (VB-XPS) were also carried out for understanding the separation and transfer of photogenerated electrons and holes between the two photocatalysts. It was found that band-gap narrowing can enlarge the visible light absorption, but lower the redox potential of generated electrons and holes. Therefore, an optimal band structure for best photocatalytic activity can be obtained by continually adjusting the electronic structures.

In chapter 5, an overall summary and conclusions of this dissertation work were provided. This thesis provided three new Ag_3PO_4-based composite photocatalysts with high efficiency for environmental purrifications and revealed that the surface-electric property, electronic structure, and multi-photon reaction are closely related to the photocatalytic activity. More importantly, this thesis revealed a novel material design-concept for exploiting new photocatalyst based on the reaction kinetics and thermodynamics. The prospects for further work were also presented in this chapter.