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学 位 論 文 題 名

Study of Advanced TiO₂-based Materials towards Superior Photocatalytic Properties

(高活性酸化チタン光触媒の研究開発)

Semiconductor photocatalysis has attracted considerable attention as a promising solution to the environmental pollution and energy crisis. Among the various photocatalysts, TiO₂ has been proven to be the benchmark photocatalyst because of its superior photoreactivity, nontoxicity, long-term stability, low cost, and environmental friendly features. Nevertheless, there are still some limitations in the aspects of light absorption, charge recombination, and surface reactivity for TiO₂ photocatalysis, which need to be further improved for the practical application.

In this thesis, the topic is design and development of advanced TiO₂ based materials for the superior photocatalytic properties via enhancing the light absorption, charge separation, and surface reactivity. Firstly, via adopting Mie's scattering effect into the light absorption region of TiO₂, the light utilization efficiency is largely enhanced, accompanied with higher activity in H₂ evolution. Next, to enhance the charge separation efficiency, Cu₂O/TiO₂ nanojunction material is fabricated; this material exhibits much higher photocatalytic activity than pure TiO₂ because of its modified nanojunction structure. Furthermore, considering many physical and chemical processes take place on the surface of TiO₂ in the photocatalytic reaction, TiO₂ single crystals exposed with high-active facets are prepared; via optimization of the surface reactivity, the photocatalytic performance is largely enhanced.

Chapter 1 provides an overview of the photocatalysis, photocatalytic water splitting, CO₂ photoreduction. This chapter also summarizes the TiO₂ based photocatalysis, and strategies of achieving high photocatalytic activities for TiO₂.

In Chapter 2, to enhance the light utilization efficiency, a novel strategy of coupling Mie's scattering effect into the light-absorption region of TiO₂ was developed. TiO₂ spheres with different diameters (330-750 nm) were controllably fabricated. It was found that both the scattering effect and the photocatalytic activity vary with the size of the TiO₂ spheres.

Among them, TiO₂ spheres with diameter of 380 nm exhibited the highest photocatalytic activity, which is attributed to its highly efficient scattering happened in the light-absorption region of TiO₂ ($\lambda < 387$ nm).

In Chapter 3, to enhance the charge separation efficiency, porous-structured Cu₂O/TiO₂ nanojunction materials were successfully fabricated. The well-designed nanojunction structure between Cu₂O and TiO₂ is beneficial for the charge separation. In parallel, the porous structure of the material can facilitate the CO₂ adsorption and offer more reaction active sites. Thus, both the charge separation efficiency and the CO₂ adsorption simultaneously enhanced, leading to the excellent photocatalytic performance in the CO₂ photoreduction.

In Chapter 4, anatase TiO₂ nanosheets with 95% active {100} facets exposed and a relatively large surface area of 57.1 m²/g were successfully prepared. This percentage (95%) is the highest among previously reported {100} facet exposed anatase TiO₂. Both the higher percentage of exposed {100} facets and larger surface area can offer more surface active sites in the photocatalytic reaction. On the other hand, the superior electronic band structure which is resulted from the higher percentage of {100} facet is also beneficial for the higher activity in both H₂ evolution and CO₂ photoreduction.

In Chapter 5, for the first time, {111} facet exposed anatase TiO₂ single crystals were prepared via both F⁻ and ammonia as the capping reagents. In comparison with the intensively investigated {001}, {100}, and {101} facets for anatase TiO₂, {111} facet owns a much higher surface energy of 1.61 J/m² and more under-coordinated Ti and O atoms, in addition with a superior electronic structure. Both the superior surface atomic structure and electronic band structure make the great contribute to the marked photocatalytic activity.

Chapter 6 summarizes the importance and originality of this research, and provides future prospects of this work. This thesis revealed that the photocatalytic properties of TiO₂ are largely affected by the light absorption, charge separation, and surface reactivity. Mie's scattering effect happened in the light absorption region of TiO₂ can greatly enhance the light utilization efficiency in the H₂ evolution. Porous-structured nanojunction material is beneficial for both the charge separation efficiency and CO₂ adsorption in the CO₂ photoreduction test. Furthermore, fabrication of TiO₂ single crystals with high-active crystal facet exposed can largely promote the surface reactivity and offer more reaction sites in the photocatalytic reaction, is proved to be an effective approach to achieve advanced and excellent performance over photocatalysts.