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### リンク

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Abstract of thesis

Poor visible light absorption efficiency and high recombination rate of electron-hole are still challenging the commercial application of photocatalytic technology. Bismuth-based photocatalysts (BiOX, X=Cl, Br, I), a kind of typical photocatalysts with narrow band gaps, can absorb light in the visible region, while the photogenerated carriers are easy to recombine. This doctoral research attempted four approaches to enhance the visible light photocatalytic activity of BiOX, including (1) breaking the layered structure of BiOX, (2) introduction of nonstoichiometric crystal defects, (3) increasing photocatalyst surface area, and (4) formation of semiconductor composite. Results from this study are expected to provide new solutions for producing efficient visible light photocatalysts which can be applied in practice.

The thesis is divided into 6 chapters. In Chapter 1, the author introduced the research background, general photocatalytic technology and its basic principles in addition to the applicable photocatalytic materials. The author paid more attention to BiOX, its major bottleneck issues, and strategies for enhanced photocatalytic activity. Finally, The author arrived at the objectives and framework of the thesis. In Chapter 2, the author prepared the photocatalyst, Bi4O5Br2 by adjusting the alkalinity of synthesis system with polyethylene glycol (PEG)-10000 as directing agent, achieving a bismuth-based photocatalyst with multiple facets and a uniform sheet-like nanostructure. The prepared composite exhibited a high visible-light activity with the maximum light absorption at 500 nm. By using Bi4O5Br2, 100% decolorization of 10 mg/L Rhodamine B (RhB) solution was realized within 20 min, which also remained after recycling the used Bi4O5Br2 for 5 times. In addition, the prepared Bi4O5Br2 exhibited good detoxification effect on chromium (Cr(VI)). In Chapter 3, the author
synthesized a new mesoporous photocatalyst, namely Bi$_4$O$_5$Br$_2$/SBA-15 by loading Bi$_4$O$_5$Br$_2$ onto the mesoporous silica material (SBA-15). The synthesized composite was confirmed to have mesoporous structure with a high visible-light absorption intensity and a low recombination rate of photo-generated electrons and holes. At a mass Bi/SiO$_2$ ratio of 30/100, the synthesized photocatalyst (Bi$_{30}$/SBA-15) reflected the fastest RhB decolorization, achieving 100% decolorization of 10 mg/L RhB solution within 20 min. This photocatalyst also exhibited high ammonium nitrogen removal and Cr(VI) detoxification. The author attributed the enhanced visible-light photocatalytic activity of the synthesized composite to the existing active sites both inside and outside of SBA-15. In Chapter 4, the author further introduced ZrO$_2$ and WO$_3$ into the structure of mesoporous Bi$_4$O$_5$Br$_2$/SBA-15 to form the narrow band gap catalyst Bi$_4$O$_5$Br$_2$/ZrO$_2$-WO$_3$/SBA-15. This new Bi$_4$O$_5$Br$_2$/ZrO$_2$-WO$_3$/SBA-15 composite could decolor 10 mg/L of RhB within 10 min, exhibiting higher visible light photocatalytic activity than Bi$_4$O$_5$Br$_2$/SBA-15. The high photocatalytic performance might be contributed by the n-p type heterojunction of Bi$_4$O$_5$Br$_2$/ZrO$_2$-WO$_3$/SBA-15, which is beneficial for the effective separation of photo-generated electron-hole pairs. This new composite was also found to have better decontamination effects on organic/inorganic matters under visible light. In Chapter 5, the author attempted to enhance the visible-light photocatalytic activity of Bi$_4$O$_5$Br$_2$ by coupling graphitic carbon nitride (g-C$_3$N$_4$) with Bi$_4$O$_5$Br$_2$ composite using in-situ synthesis method. The synthesized g-C$_3$N$_4$/Bi$_4$O$_5$Br$_2$ was found to have strong light absorption in the visible region due to its narrower band gap than g-C$_3$N$_4$. 100% decolorization of 10 mg/L RhB and 80% of phenol removal were achieved by g-C$_3$N$_4$/Bi$_4$O$_5$Br$_2$ within 15 and 120 min, respectively, in which the highest photocatalytic activity was obtained at a g-C$_3$N$_4$/Bi$_4$O$_5$Br$_2$ mass ratio of 5:5. Meanwhile, the g-C$_3$N$_4$/Bi$_4$O$_5$Br$_2$ composite possessed a large specific surface area and highly stable photocatalytic activity, which also exhibited detoxification effect on Cr(VI) in wastewater under visible light. Finally, in Chapter 6 the author summarized the major conclusions of the thesis, and proposed the future research directions.

審査の要旨

Abstract of assessment result

This research explored four approaches to enhance the visible light photocatalytic activity of bismuth-based photocatalysts (BiOX). The newly synthesized composites, namely Bi$_4$O$_5$Br$_2$, Bi$_4$O$_5$Br$_2$/SBA-15, Bi$_4$O$_5$Br$_2$/ZrO$_2$-WO$_3$/SBA-15, and g-C$_3$N$_4$/Bi$_4$O$_5$Br$_2$ were found to have strong visible light absorption and photocatalytic activity. These composites showed highly efficient RhB decolorization, ammonium removal, and Cr (VI) detoxification when treating the synthetic wastewater. The newly synthesized composites were also characterized with the mechanisms being proposed. The author also prospected the future application fields of the prepared photocatalysts in the real world of wastewater treatment. In order to realize the practical application of these photocatalysts, the impact of other characteristics of wastewater should be investigated like turbidity and co-existence of various organics and metal ions in addition to the recovery of used photocatalysts.

The final examination committee conducted a meeting as a final examination on 18 January, 2019. The applicant provided an overview of the dissertation, addressed questions and comments raised during Q & A session. All of the committee members reached a final decision that the applicant has passed the final examination.

Therefore, the final examination committee approved that the applicant is qualified to be awarded the degree of Doctor of Philosophy in Environmental Studies.