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Dissertation Summary (approx. 800 words in English)

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（要　旨）
(Summary)

Air and water pollution has been one of the challenging environmental issues in modern society. Semiconductor-based photocatalysis has been regarded as one of the most green and economical processes for potential application in environmental remediation. Bismuth-based semiconductors are considered as a promising group of photocatalytic materials due to their suitable band gap for visible light response, an increased mobility of photo-generated charge carriers because of well-dispersed Bi 6s orbital, non-toxicity, and easy tailoring of their morphologies. To utilize its full potential in environmental remediation, the present study was carried out to enhance the photocatalytic performance of layered Bi_2WO_6, one of the simplest members of the Aurivillius oxide family of layered perovskites, through cerium doping and compositing it with other semiconductors, such as $ZnWO_4$, BiOI, $CeVO_4$, and reduced graphene oxide.

Chapter 1 introduces past achievements, the state of the art, and future perspectives in heterogeneous photocatalysis and its application in environmental remediation, particularly in photocatalytic air and water purification. The layered compounds, bismuth-based photocatalysts, and composite photocatalysts and their application in environmental remediation are discussed.

Chapter 2 describes the influence of pH and Bi_2WO_6 content on the formation of the $ZnWO_4/Bi_2WO_6$ composite photocatalyst synthesized by a one-step hydrothermal method and the effect of Bi_2WO_6 content on the performance for the photocatalytic degradation of acetaldehyde under UV light. An increase in the Bi_2WO_6 content of the $ZnWO_4/Bi_2WO_6$ composite photocatalyst led to a monotonic shift of the absorption edge from about 355 nm to longer wavelength up to about 450 nm and an enhancement of photocatalytic activity for the conversion of acetaldehyde into CO_2 under UV light in comparison to individual $ZnWO_4$ and Bi_2WO_6. The enhanced photocatalytic activity of $ZnWO_4/Bi_2WO_6$ composite photocatalyst is attributed to the *n-n* isotype junctions formed between the two semiconductors and to the improved charge separation and transfer.

Chapter 3 emphasizes the involvement of $CeVO_4$ in improving the photocatalytic activity of a Bi_2WO_6/allophane composite for the degradation of gaseous acetaldehyde under visible light irradiation. The $Bi_2WO_6/CeVO_4$/allophane composites (BW/CV/A) with different amounts of $CeVO_4$ nanocrystals were prepared by mechanical mixing and hydrothermal synthesis. The adsorption capacity for acetaldehyde was strongly dependent on the $CeVO_4$ content in the prepared composites. The hydrothermally synthesized BW1/CV1/A (Bi_2WO_6:$CeVO_4$ = 1:1 mass ratio) composite photocatalyst exhibited higher photocatalytic activity than the mechanically mixed BW1/CV1/A composite. The enhanced photocatalytic activity of BW1/CV1/A is related to the extended light absorption, unique structure that improved the transport of the reactant and the final product molecules, and the effective separation of electron-hole

pairs.

Chapter 4 presents the effect of different Bi_2WO_6:BiOI molar ratios on the adsorption capacity and photocatalytic activity of the Bi_2WO_6/BiOI/allophane (BW/BI/A) composites prepared by mechanical mixing and hydrothermal synthesis. The overall morphology of the composite photocatalyst was changed with increasing the BiOI content. The BW/BI/A composites exhibited absorption edges in the wavelength range of 640–650 nm. The specific surface area, adsorption capacity, and photocatalytic activity of the BW/BI/A composite gradually decreased as the BiOI content increased. The BW/0.5BI/A composite exhibited high adsorption capacity, excellent photocatalytic performance and stability owing to its large specific surface area, greater number of easily accessible active sites, facilitated diffusion of reactants, multiple scattering of incident light, and *p*–*n* heterojunction.

Chapter 5 underlines the important role of cerium doping on tuning the morphological structure, light absorption, and photocatalytic activity of Bi_2WO_6 and Bi_2WO_6-BiOCl synthesized by a hydrothermal method using ethylene glycol and dilute HCl as solvent, respectively. The formation of flower-like morphologies was governed by ethylene glycol forming a chain-like structure on the crystallite surface and the selective adsorption of excessive Cl^- ions on the specific surface of crystallite, leading to a morphological variation. Cerium doping extended the absorption edges of Bi_2WO_6 and Bi_2WO_6-BiOCl up to 460 and > 700 nm, respectively. The results from molecular dynamics calculation confirmed that cerium doping could enhance the interaction of the Bi_2WO_6 surface with the salicylic acid molecules in Ce-doped Bi_2WO_6-BiOCl. The 1 mol% Ce-doped Bi_2WO_6-BiOCl composite photocatalyst showed the highest apparent rate constant (k = -0.360 min^{-1}), which is more than three times higher than that of non-doped Bi_2WO_6 (k = -0.105 min^{-1}), due to the synergistic effect of cerium doping and the formed *p*–*n* heterojunction.

Chapter 6 highlights the modification of the Bi_2WO_6/BiOI composite with reduced graphene oxide in a varying content for enhancing the photocatalytic performance towards the degradation of colorless organic pollutants: acetaldehyde and chloramphenicol under visible light irradiation. The 1 wt% rGO/Bi_2WO_6/BiOI composite exhibited a remarkable improvement in the photocatalytic activity for the degradation of acetaldehyde and chloramphenicol molecules due to the improved adsorption, efficient separation and transfer of photogenerated electrons, and the formation of an additional *p*-*n* heterojunction. The results from molecular dynamics calculation revealed that rGO and BiOI can significantly increase the adsorption of organic molecules over the rGO/BiOI surface in the rGO/Bi_2WO_6/BiOI.

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