

学 位 論 文 の 要 旨

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学位論文題目 Development of Advanced Technology for Photocatalytic Degradation of Organic Pollutants in Aqueous Solution with Modified g-C ₃ N ₄ Composites (英訳又は和訳 修飾型 g-C ₃ N ₄ コンポジットによる水溶液中有機汚染物質の光触媒分解の高次技術の開発)			
<p>The pollution of the aquatic environment is becoming a more serious issue due to rapid industrialization, civilization, and other environmental and global changes. Because of the rapid development of the chemical, pharmaceutical, and agricultural industries, various chemical compounds, including pesticides, steroid hormones, pharmaceutical waste, synthetic dyes, and heavy metals, reach the aquatic environment. The main source of contaminated water is thought to be organic contaminants, which are very harmful to aquatic life and human health. Thus, it is crucial to remove organic contaminants from wastewater in order to maintain aquatic life and enhance water quality. Photocatalytic degradation of organic pollutants is to be considered a cost-effective and environmentally friendly technique. Therefore, the objective of the thesis is to prepare different ZnO/g-C₃N₄ composites as visible light-driven photocatalysts and apply them to the degradation of bisphenol E, diclofenac, and orange II dye.</p> <p>In chapter 1, A general introduction about the source and adverse effects of organic pollutants, like bisphenols, diclofenac, and synthetic dyes, has been reported. The possible organic pollutant removal techniques have been mentioned briefly. The advantages of photocatalytic degradation of organic pollutants have been discussed. The uses of g-C₃N₄-based composites as photocatalysts and adsorbents have been briefly addressed.</p> <p>In chapter 2, The ZnO/g-C₃N₄ composite has been prepared by a facile one-step calcination of urea and zinc acetate mixture. The percentage of ZnO content in the composite was determined by TGA. By using FTIR, XPS, SEM, FE-EPMA, and TEM investigations, the formation of the composite was confirmed. UV-Vis DRS analysis revealed the higher visible light absorption ability of the composite. By using PL and EIS analysis of the composite, the reduction of photogenerated electron hole pair recombination was assessed. The photocatalytic efficiency of the synthesized composite was assessed in the degradation of bisphenol E (BPE). Under optimal conditions, the degradation rate of BPE with the ZnO/g-C₃N₄ composite was 8 times higher than that attained with pure g-C₃N₄. The radical scavenger studies specified that the superoxide anion radical ($\cdot\text{O}_2^-$) and photogenerated hole (h^+) were mainly responsible for the degradation of BPE. The stability experiment indicated the chemical and photo stability of the synthesized composite. Two possible photocatalytical mechanisms have been proposed.</p>			

In chapter 3, ZnO was simply deposited on g-C₃N₄ and calcined to generate ZnO/g-C₃N₄ composites. According to structural (XRD, FTIR, and XPS) and morphological (SEM, TEM, and EDS elemental mapping) analysis, the composite was successfully formed. On the other hand, based on PL and EIS analysis, the recombination of photocharges was reduced in the fabricated composite. The photocatalytic activity of the synthesized composite was evaluated in the degradation of diclofenac (DCF). The photocatalytic degradation rate of DCF using the ZnO/g-C₃N₄ composite was 27 times higher than that attained with pure g-C₃N₄ and 11 times higher than that attained with the physical mixture of ZnO and g-C₃N₄. The radical scavenger studies indicated that the superoxide anion radical ($\bullet\text{O}_2^-$) and hydroxyl radical ($\bullet\text{OH}$) act as reactive species during the degradation reaction. Possible photocatalytic degradation mechanisms and corresponding reactions have been proposed.

In chapter 4, ZnO-modified g-C₃N₄ (ZCN) composites were fabricated by a simple calcination of mixtures of urea and zinc acetate. The ZCN composites were utilized as bifunctional adsorptive photocatalysts for orange II removal from aqueous solution through adsorption and photocatalysis processes. The adsorption isotherm data of the g-C₃N₄ (g-CN) and ZCN composites on orange II solution were better fitted with the Langmuir isotherm compared to the Freundlich isotherm. The maximum adsorption capacity for ZCN-2.5 was slightly higher than that of bare g-CN. According to the adsorption thermodynamics investigation of ZCN-2.5 on orange II solution, the positive values of Gibb's free energy change (ΔG^0) suggested a non-spontaneous adsorption process. Furthermore, the negative values of entropy change (ΔS) and enthalpy change (ΔH) indicated the decrement of randomness and exothermic nature during the adsorption process. The adsorption of orange II dye on the ZCN composites is attributed to the π - π interactions, electrostatic interactions, H-bonding, acid-bas interactions between the composite and dye, and pore filling of the composites. The photocatalytic degradation kinetics of g-CN and ZCN composites indicated that the degradation process follows the pseudo-first-order reaction kinetic. The degradation rate of orange II with the ZCN-2.5 composite was 6.67 times higher than that obtained with bare g-CN. The enhanced photocatalytic degradation of orange II dye solution using ZCN composites with visible light irradiation is attributed to the enhanced visible light absorption efficacy and decrement of photogenerated e^-/h^+ pair recombination. Possible adsorption and photocatalytic mechanisms have been proposed.

In chapter 5, the conclusion about the advancement of visible light-induced photocatalytic technology for degradation of BPE, DCF, and orange II using ZnO/g-C₃N₄ composites is described.