（様式６号）「課程博士用」

学 位 論 文 の 要 旨

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学位論文題目

Photocatalytic Activity of Polycyclopentadithiophene Derivatives

（ポリシクロペンタジチオフェン誘導体の光触媒活性）

In the quest for sustainable solutions to urgent environmental challenges and the pursuit of efficient energy utilization, photocatalysis has emerged as a promising avenue. Harnessing the power of sunlight to drive chemical reactions, photocatalysis offers potential applications in environmental remediation, energy conversion and synthesis of valuable chemicals. Among the materials explored for photocatalytic purposes, conjugated polymers have garnered significant interest due to their unique electronic properties, facile synthesis and tunable structures. The present thesis is a collection of studies concerning with Polycyclopentadithiophene derivatives. This thesis consists of three chapters. The first chapter describes the synthesis and photocatalytic activity of poly(4,4'-(((4H-cyclopenta[2,1-b:3,4-b']dithiophene-4,4-diyl)bis(ethane-2,1-diyl))bis(oxy))bis(4-oxobutanoic acid)) (PCPDT-CO_2H). The second chapter describes the synthesis and photocatalytic activity of Poly(4,4-dihexylcyclopenta[2,1-b:3,4-b']dithiophene) (PDiHexCPDT). Finally the last chapter is about photocatalytic fixation of Carbon Dioxide with Polycyclopentadithiophene derivatives.

【 Chapter 1 】

A novel π-conjugated polymer based on cyclopentadithiophene (CPDT), PCPDT-CO_2H, was prepared as a sparingly soluble material. In this study, we presented the synthesis and characterization of PCPDT-CO_2H as a versatile photocatalyst for organic transformations. We investigated its ability to generate hydroxyl radicals in aqueous media, confirmed through the use of coumarin as a hydroxyl radical indicator. Moreover, we explored its catalytic performance in the oxidative hydroxylation of arylboronic acid and the oxidation of benzaldehyde, showcasing its efficacy as a metal-free and entirely organic heterogeneous photocatalyst. Our findings demonstrated that PCPDT-CO_2H efficiently could catalysed the oxidation benzaldehyde under photoirradiation with conversion of 70%. Additionally, we discussed avenues for further research, including optimization of synthesis conditions, exploration of additional photocatalytic reactions, and integration of PCPDT-CO_2H into practical devices for real world implementation. These experiment results indicate that PCPDT-CO_2H is a promising for a metal-free and 100% organic photocatalyst.

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【 Chapter 2 】

This chapter investigated the photocatalytic activity of PDiHexCPDT, a novel conjugated polymer based on CPDT, for the first time. PDiHexCPDT was compared with poly(3-hexylthiophene) (P3HexTh) in terms of its photocatalytic performance. The generation of hydroxyl radicals and the oxidative hydroxylation of boronic acid and oxidative coupling of benzylamine were used as model reactions to evaluate PDiHexCPDT photocatalytic capabilities. Our results demonstrated that PDiHexCPDT exhibits superior photocatalytic activity under fluorescent light irradiation compared to P3HexTh. PDiHexCPDT exhibited better photocatalytic activity than P3HexTh under fluorescent light irradiation. However, the combination of PDiHexCPDT with P3HexTh improved the photocatalytic activity to 84%. The combination improved the photocatalytic reactivity when the light source was a white LED, which is composed of blue and yellow emissions. The enhanced performance of PDiHexCPDT is attributed to its absorption spectrum, which matches well with the emission spectrum of the light source. Moreover, the study explored the potential of PDiHexCPDT/SiO_2 and PDiHexCPDT/TiO_2 composites and highlights the synergistic effects observed in mixed systems. However, composite formation of PDiHexCPDT with SiO_2 or TiO_2 did not give a high conversion rate. This research sheds light on the promising role of organic π-conjugated polymers as efficient and versatile photocatalysts for sustainable energy applications.