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Author(s)	劉, 維
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博士の専攻分野の名称 博士（工学） 氏名 劉 維

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Graphitic Carbon Nitride Based Photocatalysts: Design, Synthesis and Properties
Investigation
(黒鉛相窒化炭素基光触媒の設計・合成と性能研究)

Graphitic carbon nitride ($\text{g-C}_3\text{N}_4$)-based photocatalysts is gathering much attention recently for photo-induced CO_2 reduction, H_2 evolution, decomposition of harmful substances, antibiotics, etc. In this thesis, the main purpose is to explore the effect of structure modulations of $\text{g-C}_3\text{N}_4$ on photocatalysis and to improve the structural and electronic properties such as surface area, carrier separation and transfer as well as to facilitate the proton reduction kinetic towards high photocatalytic activity. I have thoroughly analyzed the structure and electronic structure and compared photocatalytic activities of the materials prepared by newly developed techniques. The results are as follows:

In Chapter 2, few-layered porous $\text{g-C}_3\text{N}_4$ (fl-P-CN) has been prepared by a novel combination of thermal etching and gas driven exfoliation of bulk $\text{g-C}_3\text{N}_4$. The high temperature treatment in ambient condition has an etching effect on $\text{g-C}_3\text{N}_4$, leading to the thermal exfoliation of $\text{g-C}_3\text{N}_4$. Later on, after immersing into liquid N_2 , the $\text{g-C}_3\text{N}_4$ is further peeled off with the immediate drastic gasification of liquid N_2 . As a result, due to the optimized aromatic pi-conjugated system, the enlarged surface area, the increased porosity as well as the quantum confinement effect induced by nanoplates formation, fl-P-CN exhibited impressively improved photocatalytic performance. By adding scavenger of active species, the active species for the RhB decomposition by this photocatalyst was determined to be photogenerated holes. These findings deepen the understanding on the dimensionality-related issues of $\text{g-C}_3\text{N}_4$ -based materials.

In Chapter 3, few-layered $\text{g-C}_3\text{N}_4$ nanoplates were prepared via sugar-assisted mechanical exfoliation method using fructose as the assistant reagent. The few-layered structure rendered the as-prepared $\text{g-C}_3\text{N}_4$ much increased surface area, and also accelerated charge separation and charge transfer efficiency. As a result, such materials exhibited high hydrogen evolution rate ($100 \mu \text{mol/h/g}$), which was about 3 times higher than that of the bulk $\text{g-C}_3\text{N}_4$. By adding scavenger of active species, the active species for the RhB decomposition by this photocatalyst was determined to be super oxides. These findings emphasized the significance of 2D structure of $\text{g-C}_3\text{N}_4$ for the efficient carrier separation and transfer and subsequent photocatalytic reactions, and also suggested that the modulation of nanostructure in $\text{g-C}_3\text{N}_4$ -related organic semiconductors in a sustainable way might open new avenues towards improved photocatalysis.

In Chapter 4, I demonstrated that post-annealing treatment of $\text{g-C}_3\text{N}_4$ nanoplates could lead to a significant enhancement in photocatalytic H_2 production over the bulk $\text{g-C}_3\text{N}_4$ under visible light irradiation. After the fast thermal etching treatment of $\text{g-C}_3\text{N}_4$ nanoplates, a H_2

evolution rate as high as $264.3 \mu \text{ mol/h/g}$ was achieved for the N-deficient g-C₃N₄ nanoplates, which was about 7 times higher over that of bulk g-C₃N₄ and 3-fold greater than that of g-C₃N₄ nanoplates by visible light irradiation. The relative photocatalytic activity of this annealed g-C₃N₄ was different between hydrogen evolution and RhB decomposition. It was confirmed that such improvement in catalytic activity was attributed to the tailored crystal, textural, optical and electronic structures. These discoveries would provide a feasible approach for the design of semiconductor materials with porous few-layered structure for efficient photocatalysis.