

論文 / 著書情報
Article / Book Information

題目(和文)	半導体ヘテロ構造体の精密設計とマイクロ波照射による人工光合成系の反応ダイナミクス制御
Title(English)	
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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

論文要旨

THESIS SUMMARY

専攻 : Department of	応用化学	専攻	申請学位 (専攻分野) : Academic Degree Requested	博士 Doctor of	(工学)
学生氏名 : Student's Name	岸本 史直		指導教員 (主) : Academic Supervisor (main)	和田 雄二	
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要旨 (英文 800 語程度)

Thesis Summary (approx. 800 English Words)

This thesis is composed of 6 chapters.

In chapter 1 "Introduction", the development of artificial photosynthetic systems was outlined. The promising system of the artificial photosynthesis is Z-scheme electron transfer system based on semiconductor photocatalysts. The most of researchers are focused on designing the materials to get semiconductor photocatalysts with a high catalytic activity. However, I considered that the efficiency of the system will be limited by only enhancement of the catalytic activity. In this study, we pointed out a construction of a vector electron transfer system by precise design of heterostructure of semiconductor nanosheets synthesized via thiol-ene click reaction, to suppress reverse electron transfer reactions inhibiting Z-scheme electron transfer system. In addition, from the viewpoint of the solid physics, we proposed an irradiation of microwaves from the outside to artificial photosynthesis system to accelerate the material conversion reaction on the photocatalyst surface, which is the bottleneck of the Z-scheme electron transfer system.

In chapter 2 "Electronic Structural Changes of n-Type Semiconductor Nanosheet by Heterostacking", the change of electronic structure caused by heterojunction of semiconductor nanosheets were understood. In an alternate stacked structure of niobate nanosheets and tungstate nanosheets, the absorption wavelength corresponding to the bandgap transition of the tungstate nanosheets were shifted shorter as the distance between the neighboring nanosheets. This short wavelength shift was understood by a model based on the steady electron filling to a conduction band of the tungstate nanosheets by static electron transfer from donor levels of the niobate nanosheets.

In chapter 3 "Dynamics of Ultraviolet Light Induced Charge Separation between Alternately Stacked Nanosheets", I demonstrated that the rate of dynamic ultraviolet light induced electron transfer reaction from the negative conduction band to the positive conduction band in the alternate stacked structure can be controlled by manipulating the interlayer distance. It was revealed that this electron transfer reaction is due to the tunneling electron transfer mechanism of the through space type depending on the distance between the nanosheets.

In chapter 4 "Construction of Z-scheme Electron Transfer System in Alternate Stacked Nanosheets induced by Position Selectively Modification of Molecules", I constructed a Z-scheme electron transfer system in alternate stacked structures driven by visible light, where dye molecules were introduced into the interlayer space of titanate nanosheets and tungstate nanosheets. This electron transfer system enables electrons to be accumulated in the conduction band with a more negative potential. Therefore, the ultraviolet light induced electron transfer reaction demonstrated in Chapter 3 works as a reverse electron transfer reaction inhibiting this Z-scheme electron transfer system. By designing the interlayer distance to suppress this reverse electron transfer process, a vectorial Z-scheme electron transfer system was constructed. In summary of the above chapters 2 to 4, I proposed that arranging the distance of the photocatalytic materials in Z-scheme electron transfer system suppress the reverse electron transfer reaction to create a vectorial electron transfer system. Such a method of exactly separating the catalytic materials to control the electron transfer system proposed a new guideline of the design of the semiconductor photocatalytic systems.

In Chapter 5 "Demonstration of Microwave Effect to Promote Material Conversion Reaction in Artificial Photosynthesis", in order to propose a new methodology to accelerate the slow substance conversion reaction on the solid catalyst surface, I demonstrated an acceleration of reactions by irradiating 2.45 GHz microwaves from outside the reaction system. First, the acceleration of a reduction reaction of water by Pt disk electrode by irradiation of 2.45 GHz microwave was demonstrated. By transient simulation of the temperature distribution under microwaves, this reaction acceleration was due to the temperature alternation of the reaction field by the microwaves. Next, the oxidation current of water by the α -Fe₂O₃ electrode was steadily enhanced by the irradiation of 2.45 GHz microwaves. The behavior of the enhancement of the oxidation current was fundamentally different from the acceleration behavior of the reduction reaction of water by the Pt disk electrode under the microwaves. I concluded that the acceleration of the oxidation current of water by the α -Fe₂O₃ electrode was not caused by the temperature alternation of the reaction field. Moreover, I found that the photoreduce reaction of the bipyridine derivatives on the surface of CdS quantum dots was also accelerated by microwave irradiation. This acceleration was also not caused by the temperature alternation of the reaction field. Based on these results, the microwave effect on the oxidation reaction of water by α -Fe₂O₃ electrode and the photoreduce reaction of bipyridine derivatives by CdS quantum dot were due to the interaction of electron groups at the surface level of solid catalysts with the microwaves.

In Chapter 6 "Summary", the results in this thesis was summarized. The vector electron transfer system suppressing reverse electron transfer in artificial photosynthetic system was successfully constructed by the precise design of the alternate stacked structure. The acceleration of the material conversion reaction on the solid surface induced by the microwaves was demonstrated. Based on these findings, this thesis proposed a new strategy to improve the energy conversion efficiency of the artificial photosynthetic system, by controlling the dynamics of electron transfer reactions and materials conversion reaction by the precise design of nanostructures and the irradiation of the microwaves from the outside of reaction system, respectively.

備考 : 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。

Note : Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1 copy of 800 Words (English).

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