

論文 / 著書情報
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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

論文要旨

THE S I S S U M M A R Y

系・コース：	材料	系
Department of, Graduate major in	材料	コース
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Student's Name		

申請学位 (専攻分野)：	博士	(学術)
Academic Degree Requested	Doctor of	
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要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

This thesis, entitled "Temperature dependence of photocatalysis," consists of eight chapters.

Chapter 1, "Introduction," describes today's environmental and energy problems and the potential of photocatalysis as a promising tool to solve these problems. It then raises the issue that the effect of temperature, an important factor in photocatalytic performance, has not been adequately studied, and at the same time explains the difficulties and importance of studying the effect of temperature.

Chapter 2, "Temperature dependence of bandgap," discusses the temperature dependence of photoexcitation, the first step in photocatalysis. A system that can measure UV-visible spectra at variable temperatures was constructed, and the bandgaps of eight representative oxides used for photocatalysis were measured in the temperature range from 22 to 500 °C. In all oxides, a significant narrowing of the band gap was observed upon heating, indicating the importance of studying the temperature dependence of the band gap, especially for photocatalytic reactions at high temperatures in the gas phase. Furthermore, we found that the degree of band gap narrowing correlates with the bonding distance between the nearest metal and oxygen. These results are related to the interatomic distance that can be changed by phonons and the interaction between the electron orbitals when the distance is changed.

In Chapter 3, "Temperature dependence of action spectra," it is experimentally verified whether carriers excited by additionally absorbed photons can induce photocatalytic reactions when the band gap is narrowed due to temperature increase. The wavelength dependence of the irradiation light for water oxidation by titanium dioxide, IPA oxidation in the gas phase by titanium dioxide powder, and hydrogen production by Pt/TiO₂ was investigated, showing that the additionally absorbed photons can drive photocatalytic reactions in all reaction systems. The results show that heat can be used as a strategy for photocatalytic band engineering.

Chapter 4, "Kinetics of excited carriers in accordance with applied bias," takes the viewpoint of the surface accumulation layer and investigates the temperature dependence of the excited carriers that are separated into them, their reactions, and their back electron recombination (BER) kinetics. In this chapter, a photoelectrochemical system is constructed and the oxidation of water by titanium dioxide is treated as a model reaction. First, the potential dependence is investigated, and it is found that the BER occurs only below 0.8 V (vs. reversible hydrogen electrode potential). The temperature dependence was found to be more significant in the region where BER occurs and less pronounced in the region where BER does not occur. Although the reaction rate of the excited carriers was increased in all potential regions, the temperature dependence was more pronounced in the low-potential region where BER occurred, because the surface-accumulated holes that had caused BER at low temperatures contributed to the water oxidation reaction when the temperature increased. From these results, we summarize the temperature dependence of the excitation carrier transfer upon temperature increase.

Chapter 5, "Establishment a Theory for Utilizing Rotating Disk Electrodes in Photocatalysis," introduces the application of the rotating disk electrode method to photoelectrochemistry, which has been mainly used for electrochemical evaluation. Theory for Utilizing Rotating Disk Electrodes in Photocatalysis I found that the current increases with increasing rotation speed in electrochemistry, but decreases in photoelectrochemistry. After discussing various possibilities, I concluded that the reason for this is that holes have a lifetime, and the probability of encountering a reaction substrate decreases with increasing rotation speed. Based on this discussion, a theoretical equation is constructed that reproduces the experimental results well. Since the decrease in photocurrent is attributed to the lifetime of holes, fitting this theoretical equation to the experimental results means that the lifetime of holes on the surface can be evaluated. Since the evaluation of recombination is a very difficult technique, this method can be a new technique.

In Chapter 6, "Temperature dependence study of photocatalysis using rotating disk electrode," the reaction temperature dependence of photocatalysis is evaluated using the method established in Chapter 5. The dependence of photocurrent on the number of rotations was measured on the same sample under controlled light intensity and reaction temperature, and the results were fitted to a theoretical equation. The theoretical equation successfully fits all of the results, and the validity of the theoretical equation is guaranteed to a certain degree. Furthermore, the experimental results can be interpreted from an industrial point of view, meaning that reaction conditions can be proposed that maximize the use of a certain amount of photon irradiation. In other words, he states that a higher reaction temperature has an advantage in terms of activity, but that

the number of rotations should be optimized depending on the light intensity. In other words, he points out that the degree of agitation in the actual reaction system should be optimized according to conditions such as light intensity.

Chapter 7, "Scientific contribution of this work," describes the contribution of this work from a scientific point of view. It shows that the reaction temperature can be redefined as one of the methods to control BER and reaction, which is a competition on photocatalytic surfaces. The results of this study also suggest an interpretation of the activation energy in photoelectrochemistry using the Arrhenius equation. Furthermore, we show that the rotating disk electrode method is applicable to photocatalysis.

Finally, Chapter 8, "Conclusion," summarizes the overall conclusions and provides design guidelines for improving photocatalytic performance when heat is taken into account.

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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