【Grant-in-Aid for Scientific Research(S)】
Science and Engineering (Chemistry)

# Title of Project : Single-molecule chemistry of nanocatalysis for light energy conversion

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Research Area : Photochemistry
Keyword : Solar light energy, Photocatalysis, Single-molecule fluorescence imaging

**【Purpose and Background of the Research】**

To design an efficient light energy conversion system, it is important to reveal and understand the molecular interactions and the mechanism of chemical reactions at heterogeneous interfaces. In this project, we will investigate the light energy conversion processes occurring on a variety of nanocatalysts using single-molecule fluorescence imaging techniques and gain information related to spatial and temporal heterogeneities in reactions, which are always masked by ensemble averaging.

**【Research Methods】**

Single-molecule fluorescence imaging

Single-molecule fluorescence microscopy will be used to investigate photocatalytic reactions at the heterogeneous interface. We will synthesize novel fluorogenic probes to selectively observe the catalytic reactions. Such probes are designed to become fluorescent upon the reaction with target species under photoirradiation. The position of individual fluorescent products can be determined with several tens nanometers spatial resolution by two-dimensional Gaussian fitting. In addition, the quantitative analysis of fluorescence intensity trajectory or fluctuation can reveal the underlying properties of individual catalysts.

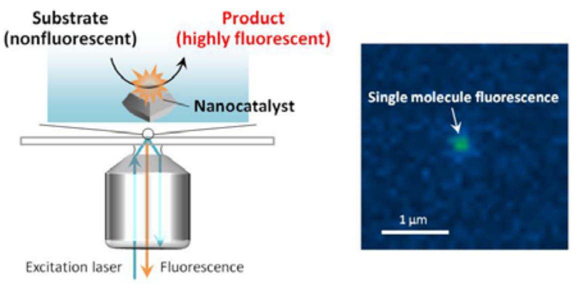


Figure 1 Single-molecule fluorescence imaging of photocatalytic reactions.

Mechanism of light energy conversion processes

We will prepare nanometer- and micrometer-sized crystals of photoactive metal oxide semiconductors, such as titanium dioxide and bismuth vanadium oxide, and explore the photocatalytic reactions on individual catalysts by single-molecule fluorescence microscopy with newly developed redox-responsive fluorogenic probes. The effects of probe concentration, solvent, pH, and light intensity will be examined to optimize the experimental conditions. From the analysis of spatial distribution of reactive sites, the relationship between surface structures and chemical reactivity will be elucidated. From the quantitative analysis of on/off duration times, we will further determine the turnover frequency of individual catalysts, adsorption and dissociation rates, interfacial electron transfer rates, and temporal fluctuation of reaction efficiency.

The photochemical reaction dynamics of metal nanoparticles, supramolecular materials, DNA, and proteins will be studied at the ensemble and single-molecule levels with the aid of quantum calculations to examine their possible use as nanocatalysts.

**【Expected Research Achievements and Scientific Significance】**

The proper understanding of structures and reactions at heterogeneous interfaces will develop the general concept of chemistry and help advance the emerging applications of nanocatalysts for environmentally and economically sustainable uses.

**【Publications Relevant to the Project】**

・T. Tachikawa, S. Yamashita, T. Majima, *J. Am. Chem. Soc.*, 2011, *133*, 7197–7204.
・T. Tachikawa, T. Yonezawa, T. Majima, *ACS Nano*, 2013, *7*, 263–275.

**【Term of Project】** FY2013-2017

**【Budget Allocation】** 145, 900 Thousand Yen

**【Homepage Address and Other Contact Information】**

http://www.sanken.osaka-u.ac.jp/labs/mec/index.html