Sustainable chemistry, Enzyme engineering, Artificial photosynthesis

Unified catalysis concepts from molecular design to sustainable chemicals and materials

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Abstract

This research is conducted in the interdisciplinary field involving chemistry, biotechnology, and computational science and machine learning, with international joint research focusing on the development of substance conversion, catalyst design, and chemical energy production technology. The collaboration between chemistry and biotechnology will lead to the generation of novel catalysts and the development of processes that contribute to sustainable material transformation. Our team's research interests include the following topics; For example, the creation of new biocatalysts by embedding artificial metal complexes in proteins, the catalyst design and enzyme modification using data science, the production of chemical energy sources by artificial photosynthesis from ubiquitous small molecules, the development of new catalysts using microorganisms and chemical catalysts, and the preparation of biocompatible materials using both microorganisms and chemical catalysts (Figure 1).

Background & Results

Catalysts are essential tools for material transformation in a sustainable society. In particular, the conversion of inexpensive raw materials and sustainable resources into highly functional materials and chemical energy sources (hydrogen, methanol, etc.), and the effective utilization of small molecules obtained from the degradation of polymers are important current issues in synthetic chemistry and bioengineering. The development of excellent catalysts and processes that skillfully handle catalysts are crucial to solving these issues. This research started at the end of 2022 by the support of a Grant-in-Aid for Scientific Research on International Leading Research from JSPS, and has already begun with collaborators from both Osaka University and RWTH Aachen, Germany. One of our recent research achievements is the construction of an artificial metalloenzyme consisting of a protein called nitrobindin with a stable β-barrel structure where a rhodium complex is inserted into the cavity via covalent bond with the side chain of an amino acid residue as shown in Figure 2. The obtained artificial metalloenzyme is found to catalyze the synthesis of isoquinoline from acetophenone oxime and alkyne. Furthermore, we constructed a chimeric protein by introducing the helix-loop-helix domain of another lipid-binding protein to the entrance of the nitrobindin cavity by genetic engineering. In collaboration with a group at RWTH Aachen, an expert in protein-directed evolutionary engineering, our group improved the domain and achieved a dramatic increase in the catalytic activity of isoquinoline synthesis. In addition, our group is currently constructing various artificial enzymes by inserting non-natural metal complexes into protein cavities and is challenging to obtain activities that surpass natural enzyme reactions and to develop catalytic systems that can handle reactions not found in nature (Figure 3).

Significance of the research and Future perspective

The collaboration between the two fields, chemistry and biotechnology, is expected to have an even greater impact on catalyst development, which has so far been developed independently in the respective fields. Through interdisciplinary international collaborations, this project aims to vigorously develop material transformations and materials using the latest catalytic technologies as well as to integrate the academic concepts of catalysis (Figure 1). The final goal is to integrate chemical- and bio-catalysis and unify the concepts of catalysis, focusing on developing and utilizing catalysts that contribute to bioeconomical material transformation.

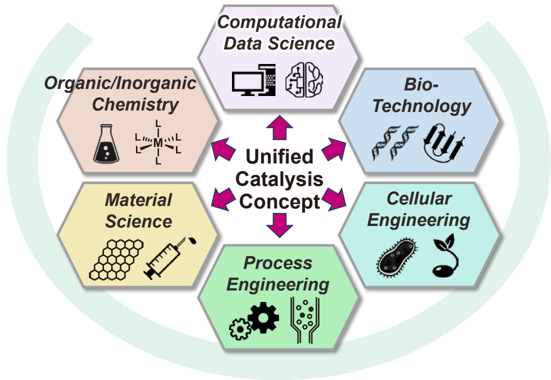

Figure 1 Various research fields related to catalysis and integration of their catalytic concept concepts

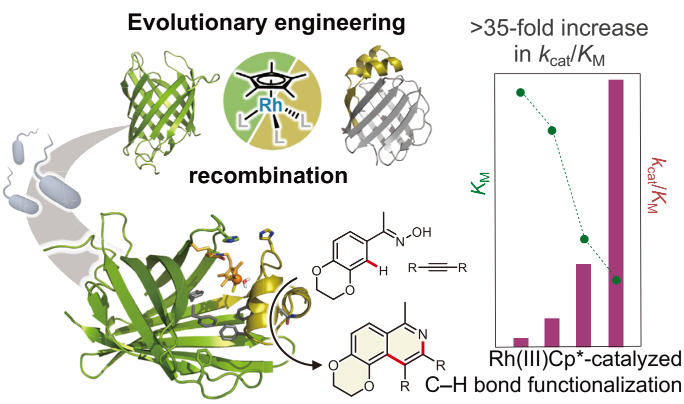

Figure 2 Development of new artificial metalloenzymes combining protein and metal complex

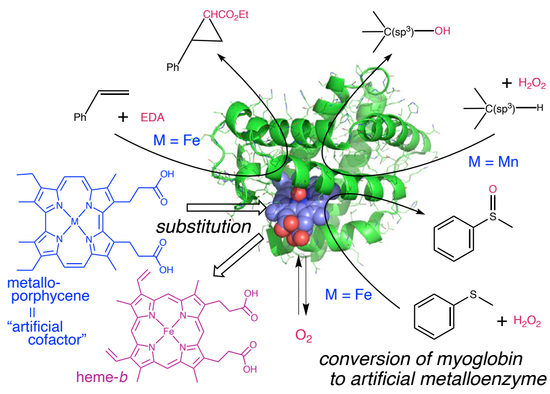

Figure 3 Construction of various catalytic reaction systems of myoglobin reconstituted with artificial metal cofactor

Patent	
Treatise	Kato, Shunsuke; Schwaneberg, Ulrich; Hayashi, Takashi et al. Evolutionary engineering of a Cp*Rh(III) complex-linked artificial metalloenzyme with a chimeric β-barrel protein scaffold. J. Am. Chem. Soc. 2023, 145, 8285-8290. doi: 10.1021/jacs.3c00581 Oohora, Koji; Onoda, Akira; Hayashi, Takashi. Hemoproteins reconstituted with artificial metal complexes as biohybrid catalysts. Acc. Chem. Res. 2019, 52, 945-954. doi: 10.1021/acs.accounts.8b00676 Kato, Shunsuke; Honda, Kohsuke; Hayashi, Takashi et al. Chitin- and streptavidin-mediated affinity purification systems: A screening platform for enzyme discovery. Angew. Chem. Int. Ed. 2023, 62, e20230376. doi: 10.1002/anie.202303764
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