Science and Technology Group
Annual Report FY2022

Satoshi Takebayashi
Science and Technology Associate

1 Introductions

Project 1: Iron-catalyzed olefin metathesis

The olefin metathesis reaction is among the most widely applicable catalytic reactions for carbon-carbon double bond formation. Currently, molybdenum- and ruthenium-carbene catalysts are the most common choices for this reaction. It has been anticipated that base metal catalyst would be a desirable economical and biocompatible substitute of the ruthenium-catalysts. *In this project, we developed such base metal catalyzed olefin metathesis reactions using **iron** organometallic complexes.*

Project 2: Direct observation of bond homolysis

Bond homolysis is among the most common bond cleavage mechanisms. Thus, a fundamental understanding of bond homolysis influences the development of a wide range of chemistry. Photolytic bond homolysis and its reverse process have been observed directly using time-resolved spectroscopy. However, direct observation of reversible homolysis remains elusive. *In this project, we developed a system that enables us to directly observe reversible homolysis.*

2 Activities and Findings

Project 1: Iron-catalyzed olefin metathesis

We discovered iron-catalyzed ring-opening metathesis polymerization of olefins using three-coordinate iron complexes. The detailed mechanistic investigation revealed a possible initiation mechanism of the polymerization reaction. The result of this project was published at Nature Catalysis in 2022.

Project 2: Direct observation of bond homolysis

We investigated the direct observation of reversible Co-Co bond homolysis using two-dimensional nuclear magnetic resonance exchange spectroscopy. The unambiguous characterization of the Co-Co bond homolysis process enabled us to study ligand steric and electronic factors that influence the strength of the Co-Co bond. Understanding of these factors will contribute to rationally designing multimetallic complexes with desired physical property or catalytic activity. The result of this project was published at Chemical Science in 2022.

3 Collaborations

These projects were carried out partly by corroboration with

Project 1: Iron-catalyzed olefin metathesis

- Prof. David Milstein (Weizmann Institute of Science)

Project 2: Direct observation of bond homolysis

- Dr. Robert R. Fayzullin (Arbuzov Institute of Organic and Physical Chemistry, FCR Kazan Scientific Center, Russian Academy of Sciences)

Project at other labs

- Prof. Akimitsu Narita (OIST)

4 Publications and other outputs

Science and Technology Group
Annual Report FY2022

Satoshi Takebayashi
Science and Technology Associate

Publication (*: corresponding authors)

(1) Xu, X.; Takebayashi, S.; Hanayama, H.; Vasylevskyi, S.; Onishi, T.; Ohto, T.;* Tada, H.; Narita, A.*
6,6'-Biindeno[1,2-b]anthracene: an Open-Shell Biaryl with High diradical Character.
J. Am. Chem. Soc. 2023, 145, 3891-3896.

(2) Takebayashi, S.*; Fayzullin, R. R.; Bansal, R.
Direct observation of reversible bond homolysis by 2D EXSY NMR.
Chem. Sci. 2022, 13, 9202-9209.
Selected as a 2022 Chemical Science HOT Article.

(3) Takebayashi, S.*; Iron, M. A.; Feller, M.; Rivada-Wheelaghan, O.; Leitus, G.; Diskin-Posner, Y.; Shimon, L. J. W.; Avram, L.; Carmieli, R.; Wolf, S. G.; Cohen-Ofri, I.; Sanguramath, R. A.; Shenhar, R.; Eisen, M.; Milstein, D.*
Iron-catalysed ring-opening metathesis polymerization of olefins and mechanistic studies.
Nat. Catal. 2022, *5*, 494-502.
Highlighted in Chem-Station (the largest chemistry web portal in Japan)

Presentation (*: corresponding presenter)

(1) Takebayashi, S.*, Milstein, D.
Iron-catalyzed ring opening metathesis polymerization of olefins
103rd Annual Meeting of The Chemical Society of Japan, Chiba, 2023.

(2) Takebayashi, S.*
Base metal catalysis evolved from organometallic chemistry: Fe-catalyzed olefin metathesis and Co-catalyzed H_2 homolysis
University of Urbino, Urbino, Italy, 2022.

(3) Takebayashi, S.* Fayzullin, R. R.; Bansal, R.
HOMOLYTIC H_2 ACTIVATION BY 17-ELECTRON COBALT COMPLEX AND ITS DIMER
44th International Conference of Coordination Chemistry, Rimini, Italy, 2022.

(4) Takebayashi, S.*, Milstein, D.
Iron-catalyzed ring opening metathesis polymerization of olefins and mechanistic investigations
29th International Conference on Organometallic Chemistry, Prague, Czech, 2022.

(5) Takebayashi, S.*, Milstein, D.
Iron-catalyzed ring opening metathesis polymerization of olefins
Gorgon Research Conference in Organometallic Chemistry, Newport, RI, USA, 2022

Patent

(1) Takebayashi, S.
Cobalt catalysts for the semi-hydrogenation of azo compounds
Japanese provisional patent application, 2022, 2022-135091.

(2) Milstein, D.; Takebayashi, S.
Iron-Catalyzed Metathesis Polymerization of Olefins.
PCT patent application, 2022, P-571572-PC.

5 External fundings

KAKENHI Grant-in-Aid for Scientific Research (C), 22K05134, FY2022-FY2024.