

# International Research Center for Elements Science – Synthetic Organotransformation –

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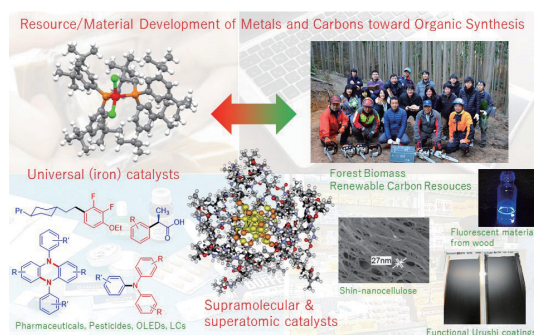
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## Scope of Research

Our research activity focuses on the development of new molecular transformations, which can contribute to better or ideal synthesis of functional molecules as well as to exploitation of new chemical (metal and carbon) resources. The present research subjects are (1) metal-catalyzed carbon–carbon and carbon–heteroatom bond forming reactions by using universal metals such as iron (2) development of small metallic nanocluster catalysts based on supramolecular approaches (3) utilization of woody biomass as the renewable carbon resources by mild catalytic transformations of wood molecules into useful compounds and materials.



## KEYWORDS

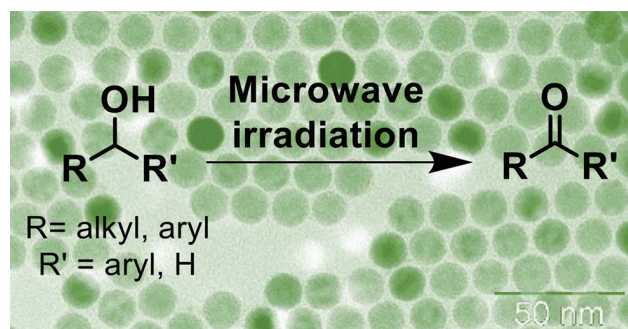
Iron Catalysis    Organotransformation    Woody Molecular Transformation    Supramolecular & Superatomic Catalysis

## Recent Selected Publications

Isozaki, K.; Iseri, K.; Saito, R.; Ueda, K.; Nakamura, M., Dual Catalysis of Gold Nanoclusters: Photocatalytic Cross-Dehydrogenative Coupling by Cooperation of Superatomic Core and Molecularly Modified Staples, *Angew. Chem. Int. Ed.* (in press).  
Nakajima, S.; Hashimoto, T.; Lu, S.; Hashizume, D.; Matsuda, H.; Hatakeyama, T.; Isozaki, K.; Takaya, H.; Nakamura, M., SciPROP-R: An Effective Bisphosphine Ligand for the Chemo-selective Iron-Catalyzed Suzuki–Miyaura Coupling of Alkyl Chlorides, *Bull. Chem. Soc. Jpn.* (in press).  
Isozaki, K.; Ueno, R.; Ishibashi, K.; Nakano, G.; Yin, H.; Iseri, K.; Sakamoto, M.; Takaya, H.; Teranishi, T.; Nakamura, M., Gold Nanocluster Functionalized with Peptide Dendron Thiolates: Acceleration of the Photocatalytic Oxidation of an Amino Alcohol in a Supramolecular Reaction Field, *ACS Catal.*, **11**, 13180–13187 (2021).  
Adak, L.; Jin, M.; Saito, S.; Kawabata, T.; Itoh, T.; Ito, S.; Sharma, A. K.; Gower, N. J.; Cogswell, P.; Geldsetzer, J.; Takaya, H.; Isozaki, K.; Nakamura, M., Iron-Catalysed Enantioselective Carbometallation of Azabicycloalkenes, *Chem. Commun.*, **57**, 6975–6978 (2021).  
Agata, R.; Takaya, H.; Matsuda, H.; Nakatani, N.; Takeuchi, K.; Iwamoto, T.; Hatakeyama, T.; Nakamura, M., Iron-Catalyzed Cross Coupling of Aryl Chlorides with Alkyl Grignard Reagents: Synthetic Scope and FeII/FeIV Mechanism Supported by X-Ray Absorption Spectroscopy and Density Functional Theory Calculations, *Bull. Chem. Soc. Jpn.*, **92**, 381–390 (2019).

## Iron-Catalyzed Organic Synthesis

Development of organic transformations using earth abundant transition-metals is becoming more and more important in recent years. Our group focuses on use of iron as a catalyst, and so far developed a variety of iron-catalyzed cross-coupling reactions, some of which can't be achieved by conventional transition-metal catalysts such as palladium. As a recent representative example, we developed a new bisphosphine ligand, SciPROP-R which enables facile iron-catalyzed Suzuki-type cross-coupling between alkyl chlorides and arylboronates. Based on the mechanistic studies, we proposed a catalytic cycle in which SciPROP-R ligand facilitates the smooth halogen abstraction from alkyl chlorides. Also, we have developed a magnetite nanoparticle-catalyzed oxidation reaction of primary and secondary alcohols assisted by microwave irradiation (Figure 1). Magnetite nanoparticles have the additional advantage of being easily recoverable from the reaction mixture by magneto-separation, which allows them to be successively recycled several times with negligible loss in activity.

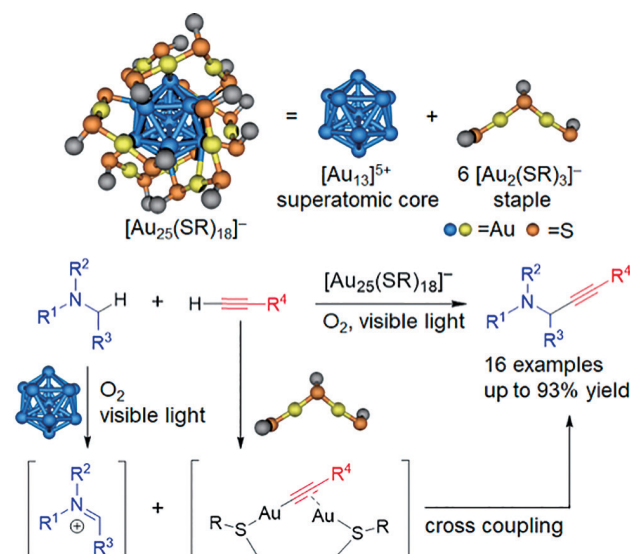


**Figure 1.** Microwave-assisted oxidation of alcohols catalyzed by iron-oxide nanoparticles.

## Supramolecular & Superatomic Catalysis

This project focuses on the development of highly active and selective metallic nanoparticle and cluster catalysts. To achieve this purpose, four key methodologies have been developed: 1) creation of reaction field with advanced self-assembly, 2) design of selective molecular transformation with multi-points intermolecular interaction, 3) size selective synthesis of metallic nanoparticles and clusters,

and 4) utilization of plasmonic resonance with light. As a recent representative example, we reported a new finding of dual catalytic property of gold nanoclusters originating from two structural components, superatomic core and surrounding staples. The cooperative catalysis enables the cross coupling between photooxidized iminium intermediates and alkynyl-gold species, providing propargylamine derivatives.



**Figure 2.** Dual catalysis of gold nanoclusters toward photocatalytic cross-dehydrogenative coupling between tertiary amines and terminal alkynes.

## Wood Molecular Transformation

Woody biomass is renewable and the most abundant carbon resource on the earth, thus expected to utilize as a feedstock to chemicals and energy toward a sustainable society. This project focuses on the chemical transformation of woody biomass into useful materials under mild catalytic conditions. To achieve this purpose, we have developed two key technologies; 1) direct transformation of woody lignin to fluorescent molecules, 2) one-pot disintegration of wood powder into nanocellulose. As a recent representative example, we reported the development of artificial urushi-coating materials by installing trienyl side chains from natural linolenic acid to the bio-renewable phenoxypropanone compounds obtained from enzymatic degradation of woody lignin.