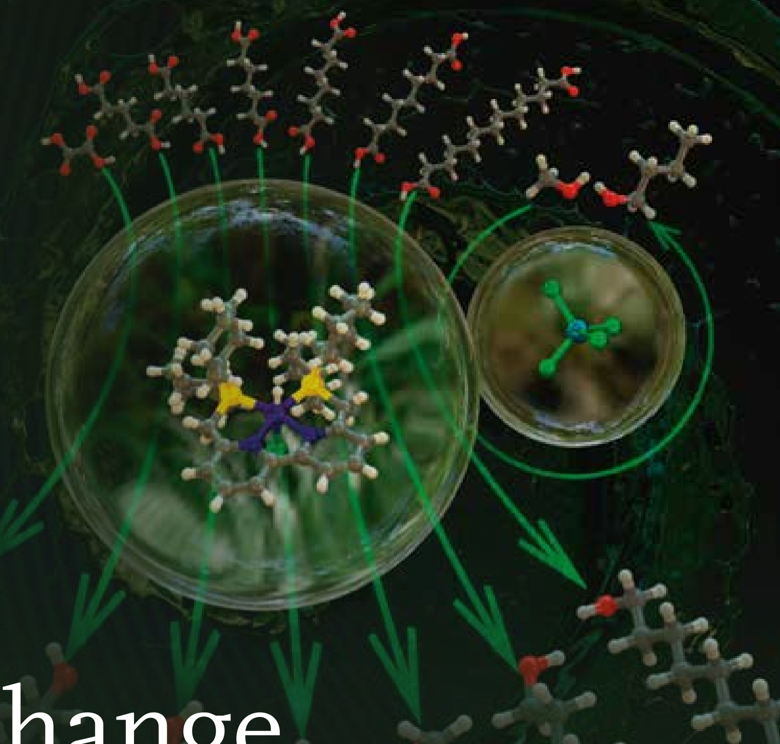


Impact Objectives

- Explore synthesising bioactive organic compounds in a greener fashion
- Develop novel reductive systems to tackle challenging upcycling transformations of highly oxidised or oxygenated compounds

A catalyst for change

***Professor Susumu Saito** shares how his work has progressed into groundbreaking research involving a prototypical structure with the potential to make significant contributions to renewability*

What inspired you to become involved in organic chemistry?

When I was a high school student, I was fascinated to learn that many medicines are made of complex but beautifully arranged organic frameworks. My father told me about prominent scientists Professor Ryoji Noyori and Professor Hisashi Yamamoto, both from Nagoya University, who are leaders in the field of organic chemistry in Japan. My father's friend, Professor Kohei Tamao, who won the Frederic Stanley Kipping Award in Silicon Chemistry in 2002, strongly recommended I study organic chemistry at Nagoya University. When I was an undergraduate freshman student at the University, Professor Yamamoto was teaching a class lecturing on prostaglandins and their biological functions. I joined and enjoyed the class, where I became interested in synthesising bioactive organic compounds in an asymmetric fashion.

Can you discuss your work in the design and synthesis of unknown organic compounds and molecular catalysts?

Renewable carbon feedstock, including biomass, carbon dioxide, wasteful plastics, etc., exist in high oxidation and/or highly oxygenated states, and thus, current state-of-the-art oxidation catalysts must be substantially modified to achieve the reduction (and dehydration) of such bio-renewable resources. We are developing tailored molecular (PNNP)M catalysts (PNNP = tetradentate ligand consisting of two-nitrogen and two-phosphine coordinative atoms, M = metal) that are responsive to diverse energies (heat, photo and electric), for the reduction of highly oxygenated/oxidised renewable carbon feedstock (biomass, CO_2, wasteful plastics, etc.) to create energy-rich useful fuels/chemicals focused on the carbon-neutral social system.

What is the anticipated impact of your work?

Our prototypical structure, (PNNP)M, could operate as a reduction catalyst responsive to diverse energies. The novel molecular design of our (PNNP)M catalysts was originally devised by us, and the reduction catalyst derived therefrom is highly robust and its catalytic activity could be sustained for a long period of time (at least not less than 144 h) under both visible light and heat energy (~200°C). Additionally, the structural robustness and high catalytic activity of (PNNP)M catalysts are a prerequisite for future implementation in society.

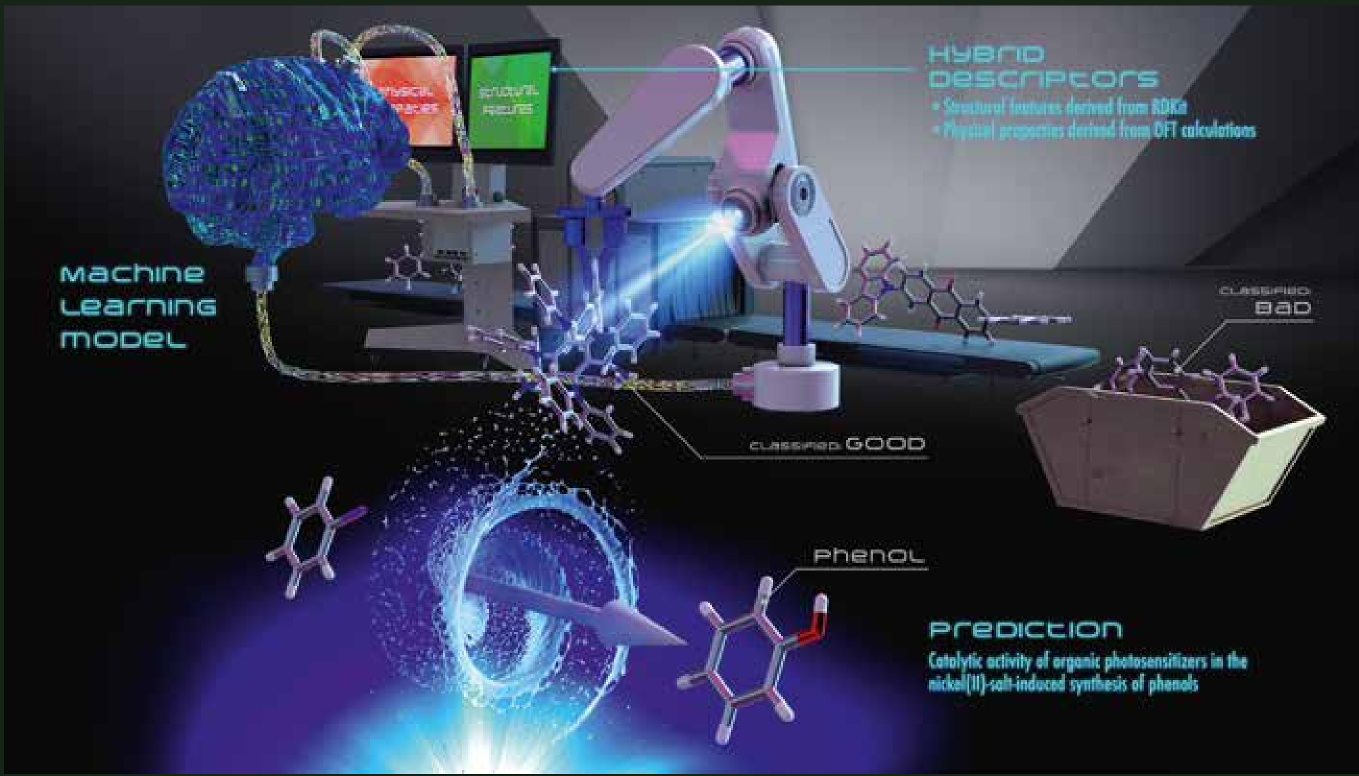
How important is collaboration for your work?

One of my collaborators is inventing a new wave of Raman spectroscopy techniques. This short-time resolution (ps~fs: pico second~femto second) system provides an unprecedented operando analysis (this combines the spectroscopic characterisation of a catalytic material during reaction with the simultaneous measurement of catalytic activity) that could give profound insights into ultra-short reaction steps or courses. Signals of different static and dynamic behaviours of different hydrogen bonds involving water and alcohols could be separated. Signals of many different real time reacting C-H bonds may also be separated. We have the potential to be able to see more effectively such short-time chemical events spectroscopically using a combination of lasers and Induced Raman measurements.

Have you had any exciting discoveries?

I am proud of the discovery of the amazing robustness of (PNNP)M complexes. We clearly demonstrated a prototypical molecular framework (PNNP)M that can be used as many distinct catalysts over different energy inputs including heat, light and electric energies while keeping its robust structure, tetradentate complexation. Furthermore, when we found (PNNP)Ru complexes that catalysed effective hydrogenation of inactivated amides including nylons, this was a real lightbulb moment. I was confident with this discovery that (PNNP)M frameworks could be applied to many different systems for the reduction of biorenewable carbon feedstock and CO_2 to alcohols and of wasteful commodity plastics to monomer units, as well as the reduction of side chains with esters/amides/nitriles of polymers, which have been sequentially demonstrated up to date. ▶

A machine can do better than a human for the discovery of a new photocatalysis?



Sustainable societies built with catalysts

***The Saito Research Group, Nagoya University**, is taking a range of novel approaches to create greener societies through using catalysis to convert biomass, CO_2 and wasteful plastics*

In the race towards a more sustainable future in which fossil fuels are replaced with renewable energy sources, biorenewable energy is an exciting concept that holds great potential for a greener tomorrow. Biorenewable resources, also known as biomass, are organic materials such as living and waste plant matter and animal matter and waste products. These organic materials can be used to generate electricity and gas in the form of bioenergy. But to make this bioenergy truly effective and efficient, catalysis is required to convert the biomass into its useful form. Professor Susumu Saito heads up the Saito Research Group in the Noyori Laboratory at the Graduate School of Science, Nagoya University in Japan, where research is underway to design catalysts that can facilitate such molecular transformations.

In one project, the researchers are developing upcycling catalysts for highly oxidised chemical compounds. Saito explains that vast quantities of carbon dioxide (CO_2) are produced every year, posing significant issues when it comes to achieving a carbon-neutral society. 'Game-changing catalysts are required to chemically transform those highly oxidised or oxygenated compounds (HOCs) which are thermodynamically and kinetically highly stable,' he describes. 'We develop novel reductive systems involving hydrogen (H_2) and H_2O as hydrogen and electron sources for tackling those extremely challenging upcycling transformations of HOCs.' The goal is that using these catalysts makes it possible to quickly and efficiently synthesise high-value-added organic molecules from carbon resources. This research could be transformative in regards to the development of self-degradable (self-immolative) plastics and therefore has great potential to contribute to advancing sustainable societies.

MAGNIFICENT METAL CATALYSIS

In another study, Saito and the team are exploring organic synthesis based on one electron transfer from H_2 or H_2O using molecular and semiconductor photocatalysis. 'Visible light energy can induce photo-excited states of tailored homogeneous and heterogeneous catalysts which promote the homolytic cleavage of chemical bonds of H_2 or H_2O, which gives very active one electron (radical) species (OES) such as hydrogen atom (H•) and hydroxyl radical (HO•),' highlights Saito. 'Using these OESs in addition reactions and H-abstraction reactions, carbon-centred radical species could be readily generated, by which we will achieve artificial photosynthesis directed toward selective organic synthesis (APOS).'

An important focus for the researchers is on molecular metal catalysis. 'Many people have an impression that organic molecules or materials are vulnerable and can be easily decomposed upon exposure for a while to light and high temperatures (>200°C),' observes Saito. 'For example, pigments that colour your hair decompose gradually under sunlight, and their original colour fades. Historically heterogeneous commercialised systems involving solid catalysts for CO_2 reduction typically operate at temperatures higher than 240°C but molecular catalysts frequently degrade and deactivate under similar conditions,' he says. They designed novel (PNNP)M catalysts, with the M representing metals, and were able to derive from these robust reduction catalysts with catalytic activity that can be sustained for a long period of time under visible light, electric and heat energy.

Different ways of thinking, and diverse skills and knowledge, when they are tangible, hit and fuse with one another, resulting in unexpected novel ideas and unpredictable outcomes

EVOLVING RESEARCH

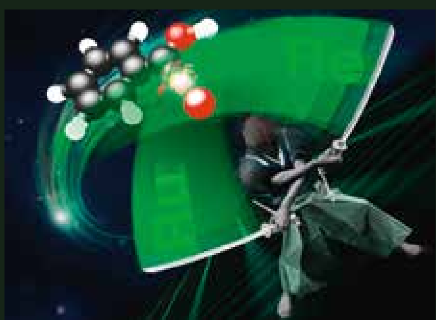
The team's focus is now evolving. 'Chemical and physical interactions of CO_2 with transition metal complexes (in our cases, hydrogen bonds are provided by the metal complexes additionally to stabilise transition states) or organic compounds in a diverse fashion,' outlines Saito. 'The interactions by which CO_2 is activated are important to convert CO_2 into valuable/energetically richer substances, which are more useful. Additionally, our interest is now moving over to the multiple interactions of nH_2O (n = any number) and semiconductor surface that are responsible for sequentially generating HO• from nH_2O and H abstraction from C-H bonds by HO•, which subsequently produces a carbon-centred radical for C-C bond formation reactions.'

Looking ahead, the team has exciting plans in the pipeline, including improving their current state-of-the-art reduction catalysts. 'We want to transform them into the utmost point of robust catalyst systems that have enough potential to be implemented by collaborating industry sectors,' says Saito. Another future line of investigation concerns their work with (PNNP)M. 'We plan to improve the (PNNP)M complexes to make them adaptable to a more effective polymer degradation and polymer functional group conversion.'

CONTRIBUTIONS TO FUTURE SUSTAINABILITY

Saito is also passionate about supporting young researchers in his field, particularly given the need for future generations to take the sustainability baton. 'To achieve the SDGs including carbon-neutral systems for energy and resources, CO_2-reducing systems, and changeover of the present polymer-based materials society, all should need much longer-term efforts and dedication,' highlights Saito. 'I have three young staff in my research group collaborating with me, offering them chances to independently steer on-site management of experiments and education of students. I give them big blueprints on steering strategy for research and which direction we should go in. I also partially support their research financially.' He says that the provision of multifaceted opportunities and resources for early-stage researchers to help them jump into new or interdisciplinary fields is among the most important.

Key to the researchers' success is interdisciplinary collaboration. 'Different ways of thinking, and diverse skills and knowledge, when they are tangible, hit and fuse with one another, resulting in unexpected novel ideas and unpredictable outcomes,' Saito enthuses. He has many academic collaborators, including different theoreticians who are good at inventing new algorithms, and the quantum calculation of both ground-states and excited states (TD-DFT) of organic molecules, a surface physicist, experts in machine learning, and a polymer chemist and physicist. 'They always bring me new wisdom and value in research and practical techniques,' he says. In addition, collaborators within industry are an important part of his work. 'Our collaborators are interested in the Sustainable Development Goals (SDGs) through their contribution to the future of society, so we are always talking about how my innovative results could be embedded into their originally developed systems and further implemented in industry.'



Chopping off the C=O of CO_2H by Ru and Re catalysts' swords

Project Insights

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BIO

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