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Dinitrogen Activation and Functionalization in Multimetallic Hydride Frameworks

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Abstract

Dinitrogen (N_2) is an abundant and readily accessible nitrogen resource, but its high thermodynamic stability and kinetic inertness render its activation and transformation challenging. In the chemical industry, N_2 is transformed to ammonia (NH_3) by the energy-intensive Haber–Bosch process under quite harsh conditions, and ammonia is currently the only nitrogen source for the synthesis of nitrogen-containing organic compounds which essential components in numerous pharmaceuticals, bioactive molecules, and functional materials. Developing alternative methods for the synthesis of value-added nitrogen-containing organic compounds through N_2 activation and functionalization under mild conditions by bypassing ammonia is of great interest and importance. We have been engaged in the activation and functionalization of N_2 using well-defined multimetallic hydride clusters as a platform. This talk mainly focuses on our recent studies on dinitrogen activation and functionalization based on titanium polyhydride frameworks such as hydroamination of alkenes with N_2 and trititanium polyhydrides[1] and aza-Michael addition of N_2 to α,β -unsaturated carbonyl compounds in a PNP-ligated dititanium framework.[2]

References

- [1] Shima, T.; Zhuo, Q.; Zhou, X.; Wu, P.; Owada, R.; Luo, G.; Hou, Z. Hydroamination of alkenes with dinitrogen and titanium polyhydrides. *Nature* **2024**, 632, 307-312.
- [2] Zhuo, Q.; Yang, J.; Zhou, X.; Shima, T.; Luo, Y.; Hou, Z. Aza-Michael addition of dinitrogen to α,β -unsaturated carbonyl compounds in a dititanium framework. *J. Am. Chem. Soc.* **2023**, 145, 22803-22813.



Takanori Shima is a Senior Research Scientist at the RIKEN Center for Sustainable Resource Science (CSRS). He received his Ph.D. in 2001 from the Tokyo Institute of Technology. In 2002, he conducted postdoctoral research in the group of Prof. John A. Gladysz at the Institute für Organische Chemie, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany, as an Alexander von Humboldt Fellow. He joined RIKEN in 2004 in the group of Prof. Zhaomin Hou as a Special Postdoctoral Researcher and was later appointed as a Research Scientist in 2009 and a Senior Research Scientist in 2011. He is a recipient of the Chemical Society of Japan Award for Creative Work (2025) and the RIKEN EIHO Award (2025). His research focuses on the development of multimetallic polyhydrido clusters for small molecule activation.