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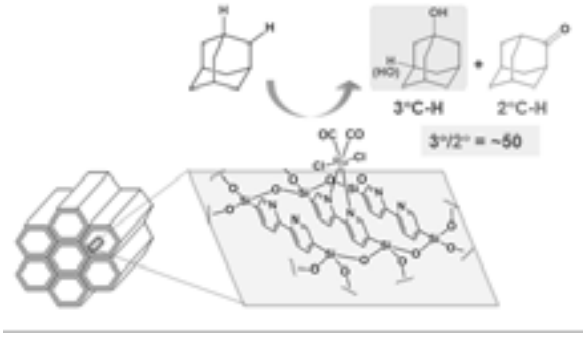
Catalysis with high density molecular monolayers for organic transformation

Kenji Hara
Tokyo University of Technology, Japan

Catalyst is one of the key materials to realize sustainable society. However, we may encounter problematic cases, where conventional catalyst systems cannot provide effective solutions. In order to establish novel methods in catalyst preparation, we investigated utilization of high-density molecular monolayers, expecting that this methodology enables facile and systematic screening of unique and efficient catalysts. It is expected that adjacent multi-metal centers immobilized on the top of a monolayer can synergistically exhibit unique catalytic activity and selectivity. In fact, uniquely high catalyst turnover numbers and high chemoselectivities were observed with high-density monolayer of Rh-phosphine complex anchored on gold surface. Another system of high turnover numbers and chemoselectivities was realized with high-density monolayer of Rh-diisocyanide on gold surface. Catalytic application of high-density Pd-bisoxazoline complex prepared on single crystal silicon surface led to oxidation of benzylic alcohols with high turnover numbers. We also utilized periodic mesoporous organosilica (PMO) as unique and promising catalyst support material. Crystal-like ordered array of the organic moiety forms the surface of PMO, giving an opportunity for its utilization as a high density ligand layer for metal immobilization. The catalytic transformation using metal-immobilized PMO will be presented. One example includes Ru-catalyzed selective and recyclable alkane C-H bond oxidation using inexpensive NaClO as oxidizing reagent. Another example is Fe-catalyzed aminoalcohol synthesis by ring-opening reaction of epoxide with amines, where unique substrate selectivities were observed.



Biography

Kenji Hara received his Bachelor's and Master's degrees from the University of Tokyo under the guidance of Prof. Eiichi Nakamura in the year in 1998 and 2000, respectively. During the Doctoral program, he has worked as JSPS Research Fellow for Young Researchers (DC 1), and moved to Faculty of Science, Hokkaido University as an Assistant Professor in 2001. He received his PhD from Hokkaido University in 2006. He was promoted to Associate Professor at Catalysis Research Center, Hokkaido University in 2007. He became the Research Leader of Research Cluster for Precise Design and Preparation of Molecular Assembly on Surface, Catalysis Research Center, Hokkaido University in 2009. He moved to School of Engineering, Tokyo University of Technology in 2015 and currently he is a Professor at the Tokyo University of Technology in the year 2016. His research focuses on the combination of molecular organometallic chemistry and solid surface science aiming for unique catalysis.

haraknj@stf.teu.ac.jp

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