OMICS Group Conferences
Accelerating Scientific Discovery

2nd International Conference and Exhibition on
Materials Science & Engineering

October 07-09, 2013 Hampton Inn Tropicana, Las Vegas, NV, USA

Catalyst surface design by molecular imprinting of oxide-supported Ru complex catalyst for regioselective epoxidation

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Molecular imprinting of a supported metal complex in which a specific ligand serves as a template is contributed to tailor-made design of catalyst surface for selective catalysis. A reaction cavity whose shape is similar to a template ligand can be selectively prepared on an unsaturated metal site supported on an oxide surface.

A novel molecularly-imprinted Ru complex catalyst was designed and prepared on a SiO_2 surface in a controllable manner, and the regulation of regioselective epoxidation of the terminal C=C bond of limonene, the reaction rate of which is much slower than its internal C=C bond, was achieved. Limonen-10-ol, which has a similar shape to an intermediate of terminal C=C bond epoxidation of limonene, was coordinated to a SiO_2-supported Ru complex as a template for molecular imprinting, and SiO_2-matrix overlayers were stacked around the supported Ru complex. Then a shape-selective reaction space with a similar shape to the template was prepared by the removal of the template. The molecularly-imprinted Ru catalyst was fully characterized step-by-step by solid-state NMR, UV/vis, XPS, BET, and Ru K-edge EXAFS.

The molecularly-imprinted Ru catalyst was highly active and exhibited fine shape selectivity for alkene epoxidation; it exhibited high regioselectivity for terminal epoxide (90% selectivity), while internal epoxide (62% selectivity) was the major product on the simple supported Ru catalyst. The terminal C=C bond epoxidation of limonene was significantly promoted on the molecularly-imprinted Ru catalyst, resulting in large decrease in the activation energy of the reaction.

Biography

Satoshi Muratsugu received his B.Sc. (2004), M.Sc. (2006), and Ph.D. (2009) from Department of Chemistry, Graduate School of Science at The University of Tokyo. After graduation, he moved to Institute for Molecular Science in 2009 as an Assistant Professor. He got Dalton Transactions Poster Prize (2009) from The Royal Society of Chemistry, The 26th Inoue Research Award for Young Scientists (2010), Presentation Award for Young Chemists (2010) from Chemical Society of Japan, and The Royal Society of Chemistry Poster Prize (2010).

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