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## Development of Hierarchical Zeolite Catalysts for the Synthesis of Renewable Chemicals

再生可能化学品合成のための階層型ゼオライト触媒の開発

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In the last century, energy, environmental and resource problems were recognized separately. However, global environmental problem was the one where former two were combined. In addition, now the resource and other issues have been combined, and it is now considered that it is the time for us to tackle on "sustainability". In this situation, new approaches are required in science and engineering.

Increasing demand for energy and commodity chemicals has led to accelerated research efforts in the conversion of renewable resource into chemicals and fuels for a sustainable economy. The processing of lignocellulosic biomass, an inexpensive, abundant and sustainable source of carbon, offers the promise of sustainable chemicals and carbon-neutral liquid transportation fuels. The International Energy Agency (IEA) laid out a 'roadmap' to ramp up the use of biofuels converted from biomass feedstocks from around 2% of global transport fuel at 2011 to 27% by the year 2050. Zeolite catalysts have shown superior catalytic activity and selectivity for converting lignocellulosic biomass into fuels and chemicals including aromatics and olefins because of the intrinsic ordered micropore structures and unique catalytic activity of zeolite catalysts.

However, the micropore structures and high intrinsic activities frequently lead these materials to be subject to diffusion limitations that restrict reactant accessibility to the active sites on the interior surfaces of zeolites, inhibit the full utilization of zeolite catalysts, and cause fast catalyst deactivation. Synthesis of hierarchical zeolites with mesoporosity is a proven strategy for reducing the diffusion limitation in zeolite catalysts. Current synthesis methods, however, request either utilization of hard templates (e.g. carbon and polymer) or synthesis of complicated surfactants.

In this presentation, a simple and efficient one-step synthesis method for mesoporous zeolites will be introduced. No any secondary organic template is used in the synthesis method. Our concept to generate mesoporosity in zeolites is to precisely control the formation and self-assembly of the zeolite precursors formed in the initial stage of zeolite crystallization. The approach requires comprehensive understanding of the crystallization mechanism of zeolites and ability to control the nucleation and crystal growth. I will first focus on the introduction of synthesis of mesoporous ZSM-5, and highlight the possibility of using the method to a wide range of zeolite catalysts. The mass transport properties and catalytic properties for biomass conversion will be discussed along with the critical future aspect regarding the development of mesoporous zeolites. It was concluded that transport control in the hierarchical zeolites includes the contribution from the configurational diffusion in micropores, diffusion on the external surface and surface pore narrowing or surface pore blockage at the external surface of the crystals. Rational development of hierarchical zeolites requires a comprehensive understanding of their transport phenomena.