Catalytic Routes for the Conversion of $C_1$ Feedstock

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Abstract:
Non-petroleum resources will play a critical role in supplying the planet with energy carriers in the future. As raw materials for fuels, biomass and light alkanes lie at opposite ends of the chemical spectrum. Light alkanes are inert and their chemical conversion involves the removal of hydrogen and may involve oxygen addition while, biomass-feedstock contains oxygen, the removal of which limits biomass-to-fuels conversion and involves the addition of hydrogen. I will describe our results related to coupling biomass-deoxygenation with alkane-dehydrogenation pathways over zeolite catalysts so that in essence, alkanes serve as a surrogate for molecular hydrogen for biomass deoxygenation while biomass serves as the oxygen carrier for hydrogen removal from alkanes.

The indirect $C_1$ route for conversion of non-petroleum carbon feedstock via methanol as a platform chemical is feedstock agnostic and offers a high degree of flexibility in the choice of products. The methanol-to-hydrocarbons (MTH) process, originally invented by Mobil, is unique in its ability to form carbon chains while concurrently restricting carbon chain length based on the sub-nanometer pore size of the inorganic zeolite catalyst. In the ‘hydrocarbon pool’ mechanism for MTH chemistry, olefin and arene intermediates, contained within the zeolite micropores act as scaffolds for carbon-carbon bond formation. The catalytic behavior of MTH systems is, therefore, determined not only by the structural and compositional features of the zeolite but also by the organic co-catalyst that comprises the hydrocarbon pool. Steady state, transient, and isotopic labeling studies were done to show that the relative contribution of the olefin and arene methylation cycles prevalent in MTH conversion over zeolites can be systematically modulated to control selectivity in MTH and, therefore, provide catalytic routes for converting any gasifiable carbon source to specific fuel or chemical products.