

Influence of the Dehydrogenation Function on Propene Aromatization Catalysis over Physical Mixtures of PtZn/SiO₂ and H-ZSM-5

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THESIS ABSTRACT

This work studies propene aromatization reaction on H-ZSM-5 (Si/Al = 40) and physical mixtures of H-ZSM-5 (Si/Al = 40) and PtZn/SiO₂ (2 wt% Pt, 3 wt% Zn) at 723 K - 823 K and 3 kPa C₃H₆. The influence of PtZn alloy (dehydrogenation function) is investigated on the product distribution and selectivity of propene aromatization. Typical product distribution consists of methane, ethane, ethene, propane, C₄-C₆ alkanes and alkenes, benzene, toluene, xylene (BTX). On comparing the BTX carbon selectivity over the two catalysts at first equivalent space velocity and then equivalent propene conversion, higher BTX selectivities are observed on PtZn+H-ZSM-5 than H-ZSM-5 in both cases. The higher BTX selectivities were previously attributed in the literature to the dehydrogenation pathway on the metal function. However, space velocity is an incorrect descriptor of reaction progress because the conversion of reactants can be different at the same space velocity. Similarly, propene conversion is an inaccurate descriptor for aromatization reaction because intermediates such as ethene and C₄-C₆ hydrocarbons react to form higher molecular weight hydrocarbons and subsequent aromatics as the reaction progresses. Such reactive hydrocarbons were lumped together as reactive species, and the remaining hydrocarbons were classified as non-reactive species or products. When BTX selectivities over PtZn+H-ZSM-5 and H-ZSM-5 are compared at equivalent temperature, and equivalent conversion of all the reactive species, both the catalysts exhibit similar BTX selectivities, suggesting that the presence of the dehydrogenation metal function doesn't influence the selectivity towards BTX products. Further, using cyclohexene as a probe reaction at 723 K and 823 K shows the presence of an alternate route of aromatic formation via dehydrogenation of cyclo olefins, and this dehydrogenation pathway has an order of magnitude higher rates than the hydride transfer route on Brønsted acid sites. In summary, these findings reveal previously unknown mechanistic details of metal bifunctionality for propene aromatization catalysis.