

SUPPORTED ORGANOIRIDIUM CATALYSTS FOR ALKANE DEHYDROGENATION

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OBJECTIVES

The goals of the project are to investigate the catalytic activities and selectivities of the supported single-site organoiridium catalysts on high surface area supports for alkane dehydrogenation. Understanding of the structure-property relationship will aid the design of better dehydrogenation catalysts.

RESULTS

The supported organoiridium catalysts were synthesized by reaction of tris(allyl)iridium with fully or partially dehydroxylated γ -alumina (Scheme 1) and subsequently characterized by solid state nuclear magnetic resonance spectroscopy (SSNMR), inductively coupled plasma atomic emission spectroscopy (ICP-AES), X-ray absorption near-edge structure spectroscopy (XANES), and X-ray absorption fine structure spectroscopy (XAFS). Under cyclohexane dehydrogenation conditions, the catalysts remained 3+ at temperatures < 220 °C, however, metallic iridium started to form when the temperature exceeded 230 °C (Figure 1). Both Ir³⁺ and Ir⁰ dehydrogenated cyclohexane to benzene selectively, and the activation energies were calculated to be 70.7 KJ/mol and 41.0 KJ/mol, respectively (Figure 2).

CONCLUSIONS

High surface area γ -alumina-supported organoiridium catalysts dehydrogenate cyclohexane selectively to benzene at ambient temperatures. The tunable nature of these catalysts will allow for substrate-specific optimization and possible development of a variety of tandem processes

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Scheme 1. Synthesis of the supported organoiridium catalysts.

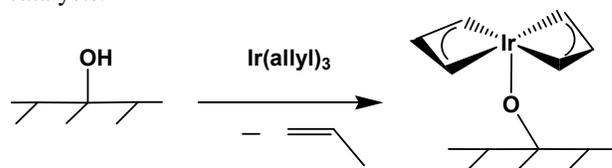


Figure 1. Comparison of the XAFS of Ir(allyl)₂(O-)/Al₂O₃(1000) at different temperatures.

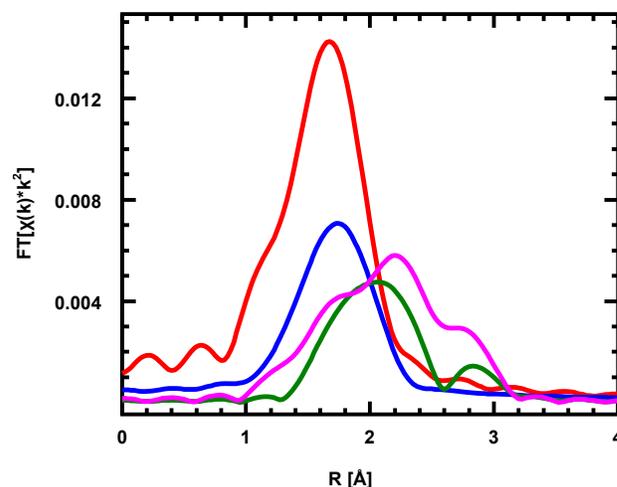


Figure 2. Kinetic plots of Ir³⁺- and Ir⁰-catalyzed cyclohexane dehydrogenation.

