

Catalytic Ring Opening of Perhydroindan - Hydrogenolytic and Cationic Reaction Path

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摘要 Perhydroindan (bicyclo[4.3.0]nonane) was converted in a flow-type apparatus under a hydrogen pressure of 5 MPa on six different catalysts, namely on a bifunctional Pd/Na,H-Beta zeolite, on Ir/Na,H-Y and Pt/Na,H-Y zeolites with a low concentration of Brønsted acid sites, and on three catalysts containing the three noble metals on the non-acidic support silica. On the bifunctional zeolite Pd/Na,H-Beta, skeletal isomerization of perhydroindan was the primary reaction followed by opening of one naphthenic ring, the formation of open-chain nonanes in low yields of ca. 6%, and hydrocracked products C₈-. The carbon number distribution of the latter was volcano-shaped with no C₁, C₂, C₇, and C₈ indicating a carbocationic hydrocracking of C₉ precursors with one naphthenic ring. On Ir/Na,H-Y and Pt/Na,H-Y (“high-performance ring-opening catalysts”), ring opening and hydrocracking to C₈- occurred by hydrogenolysis on the respective metal. Opening of the five-membered ring was found to be much faster than opening of the six-membered ring, in agreement with literature reports. The maximal selectivities of open-chain nonanes (OCNs) attained on Ir/Na,H-Y and Pt/Na,H-Y were very high, viz. 49% and 54%, respectively, and significantly better than those of the open-chain decanes observed previously with decalin as model hydrocarbon. The OCNs formed on Pt/Na,H-Y were much less branched than those formed on Ir/Na,H-Y which was interpreted in terms of the different hydrogenolysis mechanisms on both metals. Valuable ancillary mechanistic information was obtained from the selectivities of perhydroindan hydroconversion on the three noble metals on silica. In contrast to Pd/silica, Ir/silica and Pt/silica gave appreciable selectivities of OCNs as well, yet the maximum values of these selectivities were lower than those obtained on the two high-performance zeolite catalysts.

关键词: ring opening hydrogenolysis bifunctional catalysis perhydroindan open-chain nonanes noble metals silica zeolites

Abstract: Perhydroindan (bicyclo[4.3.0]nonane) was converted in a flow-type apparatus under a hydrogen pressure of 5 MPa on six different catalysts, namely on a bifunctional Pd/Na,H-Beta zeolite, on Ir/Na,H-Y and Pt/Na,H-Y zeolites with a low concentration of Brønsted acid sites, and on three catalysts containing the three noble metals on the non-acidic support silica. On the bifunctional zeolite Pd/Na,H-Beta, skeletal isomerization of perhydroindan was the primary reaction followed by opening of one naphthenic ring, the formation of open-chain nonanes in low yields of ca. 6%, and hydrocracked products C₈-. The carbon number distribution of the latter was volcano-shaped with no C₁, C₂, C₇, and C₈ indicating a carbocationic hydrocracking of C₉ precursors with one naphthenic ring. On Ir/Na,H-Y and Pt/Na,H-Y (“high-performance ring-opening catalysts”), ring opening and hydrocracking to C₈- occurred by hydrogenolysis on the respective metal. Opening of the five-membered ring was found to be much faster than opening of the six-membered ring, in agreement with literature reports. The maximal selectivities of open-chain nonanes (OCNs) attained on Ir/Na,H-Y and Pt/Na,H-Y were very high, viz. 49% and 54%, respectively, and significantly better than those of the open-chain decanes observed previously with decalin as model hydrocarbon. The OCNs formed on Pt/Na,H-Y were much less branched than those formed on Ir/Na,H-Y which was interpreted in terms of the different hydrogenolysis mechanisms on both metals. Valuable ancillary mechanistic information was obtained from the selectivities of perhydroindan hydroconversion on the three noble metals on silica. In contrast to Pd/silica, Ir/silica and Pt/silica gave appreciable selectivities of OCNs as well, yet the maximum values of these selectivities were lower than those obtained on the two high-performance zeolite catalysts.

Keywords: ring opening, hydrogenolysis, bifunctional catalysis, perhydroindan, open-chain nonanes, noble metals, silica, zeolites

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