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Hydrogenation of Alkyne Using Palladium-Zeolite Coupled with Diphenyldiethoxysilane

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Palladium-zeolite coupled with diphenyldiethoxysilane was prepared by Chemical Vapor Deposition (CVD). The hydrogenation of alkyne was carried out over this catalyst. Alkyne was converted to the corresponding alkene with a high selectivity in a manner similar to that in the case of Lindlar catalyst.

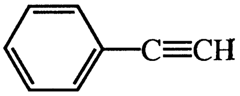
The half hydrogenation of alkynes is difficult, since the rates of the hydrogenation of double and triple bonds are nearly the same. It is known that the rate decreases after the first hydrogenation of triple bond over palladium catalyst.1) Palladium-calcium carbonate deactivated by lead acetate, which is known as Lindlar catalyst, is the most popular catalyst of the half hydrogenation.2)

In previous papers,3,4) we reported the regioselective hydrogenation of unsaturated compounds over palladium-zeolite 4A which was modified with diphenyldiethoxysilane by the CVD-method (catalyst A). It was suggested that steric hindrance should be created around the palladium as the active site in this catalyst system. In this stady, we have found that various alkynes are efficiently converted to the corresponding alkenes by half hydrogenation over catalyst A.

The preparation of this catalyst was described in a previous paper.4) A hexane solution (5 cm^3) of alkyne (0.05 mmol) and undecane, as an internal standard, was stirred with catalyst A (150 mg) at 25 °C under a hydrogen atmosphere. The reactants and products were analyzed by gas chromatography (a capillary column DB-1 30 m and PEG 20M 25 m) and GC-MS (Shimadzu QP-1000). The compounds were assigned and their concentrations were determined using authentic samples and the internal standard.

The hydrogenation of various alkynes were performed over catalyst A. These results are shown in Table 1. All alkynes were converted to the corresponding alkenes with high yields. 3-Nonyne was converted to 3-nonene with a 38.7% yield over non-modified palladium-zeolite 4A (control catalyst), while, the yield of 3-nonene increased by ca. 100% using catalyst A. Thus, catalystA is useful for the half hydrogenation of alkyne.

Table 1. Half Hydrogenation of Acetylene-Compounds[a]

Reactant	Catalyst	Yield/%
$CH_3(CH_2)_4C\equiv CCH_2CH_3$	Catalyst A	97.0
	Control catalyst	38.7
$CH_3(CH_2)_3C\equiv C(CH_2)_2CH_3$	Catalyst A	98.7
	Control catalyst	53.1
$CH_3(CH_2)_6C\equiv CH$	Catalyst A	73.5
	Control catalyst	15.4
	Catalyst A	60.7
	Control catalyst	25.8
$CH_3(CH_2)_4C\equiv C(CH_2)_2OH$	Catalyst A	65.3
	Control catalyst	22.6

a) Catalyst A: palladium-zeolite 4A coupled with diphenyldiethoxysilane, Control catalyst : non-modified palladium-zeolite 4A.

Conditions: catalyst, 30 or 150 mg; alkyne, 0.05 mmol; hexane, 5 cm^3;reaction temperature, 25 °C.

References

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