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CATL – General Catalysis

Effects of support surface structure and composition on the selectivity of Pd/C for the hydrogenation of multifunctional chemicals

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Palladium supported on carbon (Pd/C) catalyzes a broad range of cellulosic biomass and bio-oil upgrading reactions, including decarbonylation, hydrogenation, and hydrogenolysis reactions. Surprisingly, while Pd/C is a widespread catalyst, its reactivity is not well understood. Differences in selectivity towards C=C and C=O bond hydrogenation were observed for the conversion of cinnamaldehyde, an α,β -unsaturated aldehyde, depending on the nature of the Pd/C catalyst. These variations have been previously attributed to Pd particle size and/or carbon support effects on cinnamaldehyde adsorption. However, the heterogeneous nanoscale structure of Pd supported on activated carbon and carbon black makes the identification of conclusive structure-activity correlations particularly challenging.

Here, we decreased the level of structural complexity of the support in order to decouple effects due to surface chemistry and electronic structure. Specifically, we have successfully supported 3-5 nm Pd particles on nanocarbons with similar morphology and textural properties but different graphitic characters. Significant differences in selectivity were observed for the hydrogenation of cinnamaldehyde. Electron microscopy, X-ray diffraction, and hydrogen chemisorption allowed us to rule out particle size/shape effects. Correlations between the supports' defect structure and the metal's surface chemistry were identified by X-ray photoelectron spectroscopy and aberration-corrected transmission electron microscopy.