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Titel des Beitrags: Ethylene Conversion to Ethylidyne over Pd(111): Revisiting the Mechanism with First-Principles Calculations

Abstract:
On Pd(111), thermal activation of ethylene has been reported to yield ethylidyne. Using more approximate models, a plausible three-step mechanism, ethylene -> vinyl -> ethylidene -> ethylidyne, was recently proposed for this process on the basis of DFT calculations. We employed more elaborate computational models and characterized the thermodynamics and kinetics of the mechanism of ethylene conversion to ethylidyne on Pd(111). We carried out density functional slab-model studies for three coverages of the adsorbate, 1/3, 1/4, and 1/9. The resulting refined potential energy landscape turned out to differ notably from that reported previously: our calculated barriers for the various elementary steps are significantly lower than those of previous studies, and we determined the overall process to be exothermic, in contrast to earlier computational results. We show that the three-step mechanism is thermodynamically and kinetically feasible on Pd(111), with the dehydrogenation of ethylene to vinyl being the rate-limiting step at all coverages considered. Direct conversion of ethylene to ethylidene is unlikely due to a very high barrier. Coverage effects have been found important. At high coverage, the rate-limiting first reaction barrier is similar to 50 kJ mol(-1) above the desorption energy of ethylene,
whereas at low coverages the two energies become comparable.

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