Understanding the Pd size effect on formic acid dehydrogenation reaction over Pd/C catalysts

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Formic acid (FA, HCOOH) has gained worldwide attention as a hydrogen-carrier-material for its high hydrogen density, stability and non-toxicitiy. FA can be decomposed into H_2 and CO_2 through a dehydrogenation reaction, which is easily activated in the presence of Pd-based catalysts. Herein, we investigated the Pd size effect on the FA dehydrogenation reaction. The size-controlled Pd/C catalysts were prepared by NaBH4 treatments in the Pd size range from 2.5 nm to 4.8 nm. The catalytic activities continuously decreased with increasing the Pd size. Furthermore, turnover frequency (TOF(h^{-1})) values, which were calculated based on the number of surface active sites, also declined with the Pd size, but their decreasing trend gradually disappeared when ramping the reaction temperature. Thus, we identified that there is a thermodynamic effect of Pd size, which is evidenced by the increasing activation energy of FA dehydrogenation with the Pd size of the Pd/C catalysts. Moreover, through temperature programmed hydrogen decomposition analysis, we could propose that the H_2 desorption step is the rate-determining step for the FA dehydrogenation reaction.