Unraveling the reaction pathway and rate-determining step of aqueous formic acid dehydrogenation reaction over Pd/C

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Formic acid (HCOOH), which can be decomposed to H_2 and CO_2, is considered a promising hydrogen source/hydrogen-carrier-material. Thus, many studies have elaborated to develop highly active catalysts for dehydrogenation of formic acid to produce hydrogen. However, there is still a lack of understanding formic acid dehydrogenation (FAD) reaction itself (e.g. reaction pathway and rate-determining step).

Herein, we prepared Pd/C catalysts with ultra-fine Pd particles (~ 2nm) by using trisodium citrate as a stabilizing agent. A series of aqueous FAD reaction and DFT calculation could demonstrate that the reaction pathway of FAD is a formate anion dehydrogenation pathway, not a formic acid dehydrogenation pathway. In addition, further computational prediction combined with kinetic isotope effect experiments verified that combinative desorption of hydrogen is the rate-determining step of FAD. These results would provide the mechanistic insights for developing advanced catalysts for the aqueous FAD reaction.