Deposition of Carbon Layers on Pt Surface: Selective Dissociation of H_2 over O_2 on Hindered Pt Surface for Direct Synthesis of Hydrogen Peroxide

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Direct synthesis of hydrogen peroxide (H_2O_2) has been studied extensively over the past decades, however, the efficiency of the catalysts remains unsatisfactory. In order to achieve high catalytic selectivity towards H_2O_2 from an H_2/O_2 mixture, H-H bonds should be dissociated while O-O bonds should be kept unbroken in the course of the catalytic reaction. However, a major dilemma in the catalyst design is that the metal catalysts that dissociate H-H bonds even prefer O-O bonds dissociation thermodynamically. Here we report that selective dissociation of H_2 over O_2 was realized by depositing H_2-selective carbon diffusion layers on the top of a Pt catalyst. Because O_2 cannot access to the carbon-coated Pt surface, O_2 hydrogenation occurs at the carbon surface via spilt-over hydrogen rather than at the Pt surface where O-O dissociation is preferably. Such catalyst using the hydrogen spillover phenomena leads to the great suppression of O-O dissociation, which allows highly selective synthesis of H_2O_2. Notably, nitrogen doping on the carbon diffusion layer could significantly increase the selectivity towards H_2O_2 due to the stabilization of the reaction intermediate hydroperoxy radical on the carbon surface.