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Formic acid dehydrogenation over activated carbon supported Pd-based catalysts: Insight into the cold plasma treatment

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Controllable synthesis mechanism of plasma still remains obscure, although it has been proved to be an efficient method for synthesizing high-performance catalysts.[1] Herein, we investigate formic acid dehydrogenation (FAD) over Pd/C catalysts synthesized using four different methods: thermal treatment (Pd/C-C), plasma treatment (Pd/CP), thermal treatment followed by plasma treatment (Pd/C-CP), and plasma treatment followed by thermal treatment (Pd/C-PC).[2] The activity of Pd/C FAD catalysts follows the order: Pd/C-CP > Pd/C-C > Pd/C-P > Pd/C-PC. The $TOF_{initial}$ for Pd/C-CP is 1.4, 2.9, 1.4 times higher than that over Pd/C-C, Pd/C-P and Pd/C-PC, respectively. The excellent performance of Pd/C-CP can be attributed to the small size of Pd nanoparticles, the high concentration of metallic Pd, and the high Pd/C atomic ratio resulting from the migration of the electroneutral Pd species under the Coulomb repulsion effect in the plasma. In addition, we also investigated and disclose the influence of working gas on the structure and performance of Pd/C catalysts.[3] Formic acid (HCOOH) dehydrogenation activity over the Pd/C catalysts follows the order: Pd/C-H_2P > Pd/C-ArP > Pd/C-AirP ≈ Pd/C-O_2P. The Pd/C-H_2P catalyst possessed the highest activity, and the $TOF_{initial}$ was as high as 552.4 h^{-1}. The discharge parameters indicated that few micro discharge channels were generated in H_2 plasma, and the power of a single-channel discharge is high. It facilitates the migration of the Pd active species from the pores to the outer surface of the support, and the mean size of the Pd nanoparticles were 2.6±1.0 nm. In addition, the atomic ratio of Pd/C and the content of metallic Pd in Pd/C-H_2P, determined by XPS, were as high as 0.0229 and 50.8%, respectively. These are beneficial to the reaction, and ensured the highest catalytic activity of the Pd/C-H_2P catalyst.

Based on the above researches, cold plasma has also been applied to prepare PdAu and PdRu catalysts. The catalytic activity and stability of the Pd-based catalysts have been greatly enhanced, and the influence mechanism of cold plasma treatment has been discussed. This work is supported by the National Natural Science Foundation of China (Grant No. 52077024, 21773020, 21673026, 11505019), and the State Key Laboratory of Structural Analysis for Industrial Equipment at Dalian University of Technology (Grant No. GZ21105).

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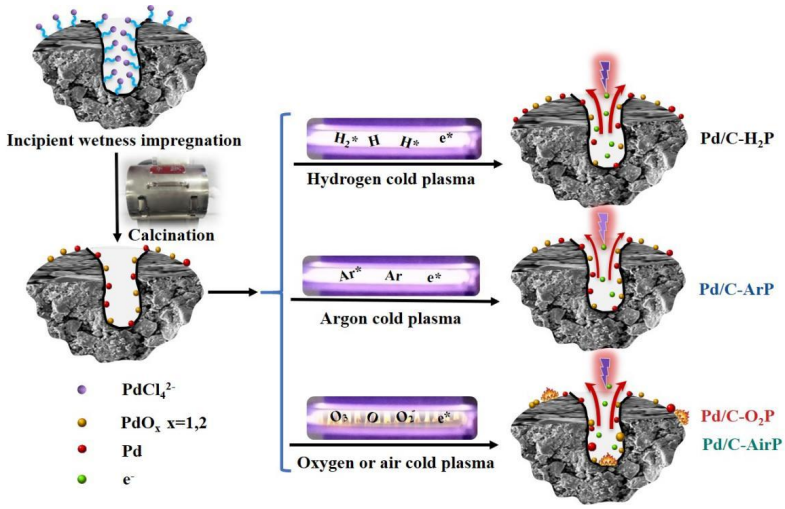

Figure 1. Schematic diagram of the proposed preparation mechanism of Pd/C-AirP, Pd/C-ArP, Pd/C-O_2P and Pd/C-H_2P.[3]