

Structural Dynamics of Pd-Based Catalysts in Acetylene Hydrogenation

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For the sustainable synthesis and conversion of renewable hydrogen into synthetic fuels and as a solution for energy storage, highly efficient catalysts are crucial. Understanding catalytic mechanisms is essential. Within the CatLab framework, we developed Laterally Condensed Catalysts (LCCs) with a functional interface between a thin metal film (Pd, Au) and buffer layer SiO₂. This study centers on the selective hydrogenation of acetylene to ethylene, an industrially significant process, focusing on enhancing ethylene production by selectively converting acetylene in the product stream. We investigated the effect of a second element in bimetallic Pd-based catalysts under acetylene hydrogenation using a few nanometers thick Pd and PdAu LCC catalysts, characterized by dual interfaces: one non-reactive with the support and the other reactive with the gas feed. Our time-on-stream catalytic measurements indicated that, initially (<20h), adding gold to palladium enhanced ethylene selectivity, consistent with literature. However, extended operation (>20h) revealed that the enhanced selectivity by Au diminished due to palladium segregation to the surface (Fig. 1f), which was not predicted by atomistic simulations. Despite this, the Pd LCC doped with Au showed less agglomeration (Fig. 1). Spectroscopy and microscopy studies confirmed that gold minimized atomic carbon incorporation into the catalyst lattice (Pd:C), suggesting that carbon and gold have analogous roles in palladium catalysts for selective acetylene hydrogenation. Overall, the LCC catalysts outperformed standard powder and bulk catalysts.

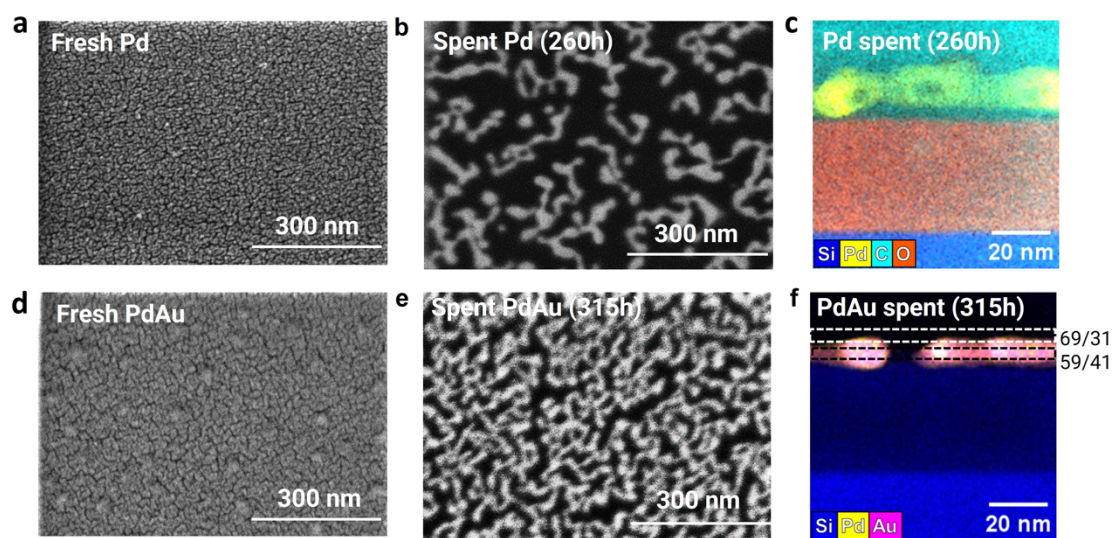


Figure 1. Development of the surface morphology of fresh and spent (a-b) Pd and (d-e) PdAu LCCs. Cross-sectional STEM-EDX elemental maps of spent (c) Pd and (f) PdAu LCCs.