

Quantifying the 3D Distribution of Pd Nanocatalysts Supported on Mesoporous Carbon for Furfural Hydrogenation

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Supported catalysts play an irreplaceable role in chemical and fuel production as well as in environmental control [1]. Highly porous materials are often used as support because of their high specific surface areas and controllable pore structure [2]. The distribution and location of nanoparticles supported on the porous support has a significant effect on the catalytic performance due to a combination of factors such as confinement effects, transport properties, and interaction with the support surface [3,4]. To understand the effect of the nanoparticle location on catalytic properties as basis to design desired catalyst, the precise determination of the location and distribution of the supported nanoparticles location is essential.

In this work, using quantitative electron tomographic analysis, the 3D distribution of Pd nanoparticles supported on an ordered porous CMK-3 carbon has been systematically investigated to evaluate the influence of the catalyst preparation on the particle distribution and to correlate this with the catalytic performance. Incipient wetness impregnation (Pd_{IW}/CMK-3), wet impregnation (Pd_{IMP}/CMK-3) and immobilization of preformed PVA stabilized nanoparticles (Pd_{PVA}/CMK-3) was used for the preparation. Figure 1 shows representative 2D slices from the reconstructed volume of the three Pd/CMK-3 catalysts. Pd nanoparticles were found both inside pores and on the external surface of the support in the Pd_{IW}/CMK-3 and Pd_{IMP}/CMK-3 catalysts, whereas in case of Pd_{PVA}/CMK-3 Pd nanoparticles were only observed on the external surface. The number of Pd nanoparticles on the external and internal support was quantified based on the identification and segmentation of the particles in the reconstructed volume. The percentage of particles on the external surface is calculated from several (3-7) tomograms of each Pd/CMK-3 catalyst. As shown in Figure 2, about 20%-30% of Pd nanoparticles are located on the external support surface for Pd_{IW}/CMK-3, almost independent of the size of the CMK-3 support, whereas in case of Pd_{PVA}/CMK-3, all Pd nanoparticles are exclusively located on the external CMK-3 surface. For the Pd_{IMP}/CMK-3, the Pd nanoparticle fraction on the external surface is roughly proportional to the external surface of the support, suggesting adequate time and mobility of the precursor to access the anchoring sites on the internal and external surfaces. However, the areal density on the outer surface is a factor of ~2 higher than on the internal surface, presumably due to the different accessibility. Furfural hydrogenation testing indicates that the selectivity of these catalysts is highly related to the particle location. Pd nanoparticles confined inside pores of the support promote the formation of 2-methyl furan while Pd nanoparticles on the external surface favor to form the tetrahydrofurfuryl alcohol. Moreover, the stability of the catalyst could be related to the differences in the preparation [5].

References:

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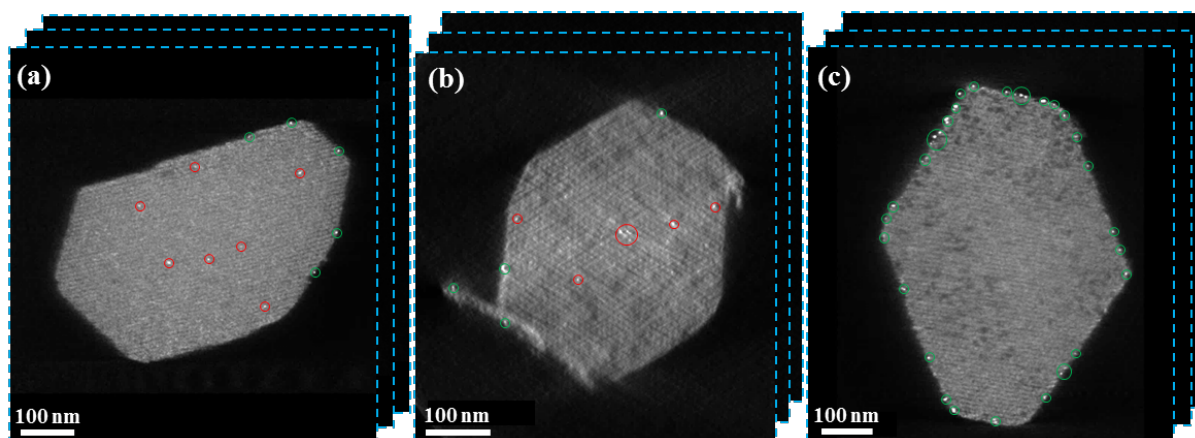


Figure 1. Typical 2D slices from the reconstructed volume of (a) Pd_{IW}/CMK-3, (b) Pd_{IMP}/CMK-3 and (c) Pd_{PVA}/CMK-3. The green and red circles highlight Pd nanoparticles on the external surface and inside of the porous support.

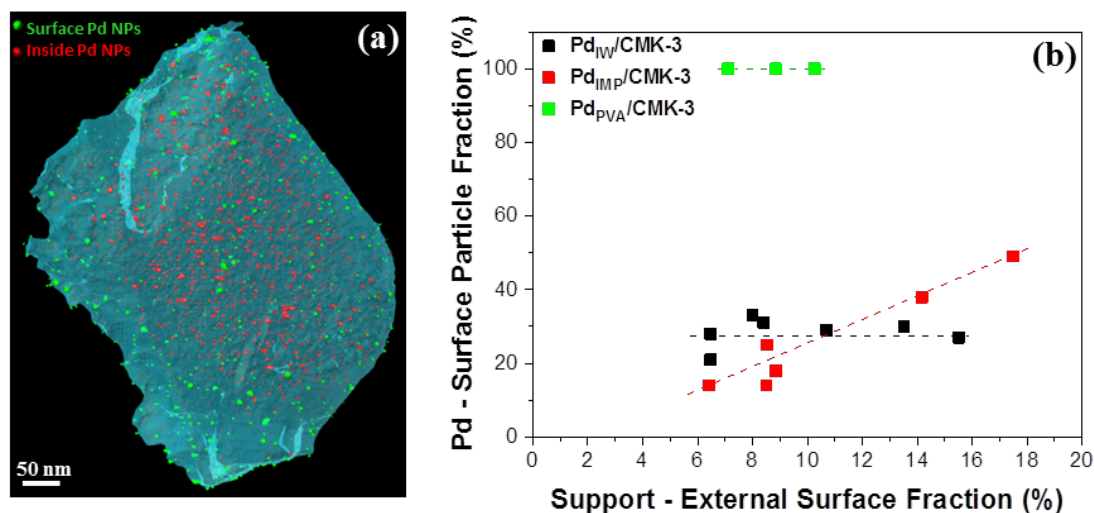


Figure 2. (a) A representative 3D visualization of Pd_{IW}/CMK-3 reconstruction showing Pd nanoparticles on the external and internal surface of support and (b) the fraction of Pd nanoparticles on the external surface as a function of the external surface fraction of the support in Pd/CMK-3 catalysts.