Electronic Supplementary Material (ESI) for Nanoscale Advances.

Supporting information for:

Disclosing the Leaching Behaviour of Formic Acid Decomposition in Batch and Fix Bed Reactors

Xiaohui Huang[1,2], Ilaria Barlocco[3], Alberto Villa[3*], Christian Kübel[1,2,4*] and Di Wang[1,4*]

[1] Institute of Nanotechnology, Karlsruhe Institute of Technology, Eggenstein-Leopoldshafen, Germany.

[2] Department of Materials and Earth Sciences, Technical University Darmstadt, Darmstadt, Germany.

[3] Dipartimento di Chimica, Università degli Studi di Milano Via Golgi 19, 20133 Milano, Italy.

[4] Karlsruhe Nano Micro Facility, Karlsruhe Institute of Technology, Eggenstein-Leopoldshafen, Germany.

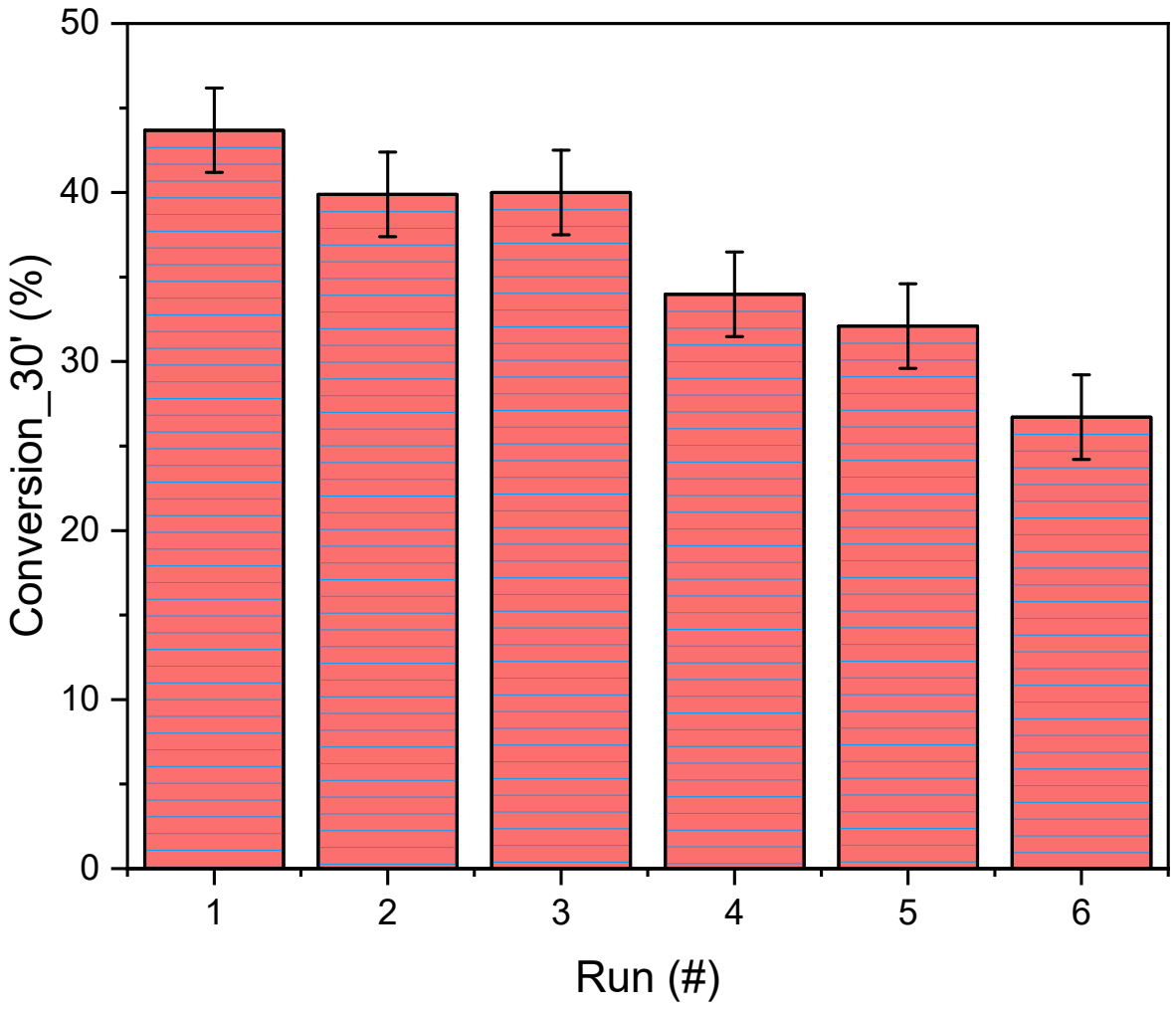

Figure S1. Stability test of Pd@CMK3 after washing the catalyst in batch reactor during FA decomposition after 30 minutes of reaction time. The filtered material was placed in a beaker with 50 mL of water under vigorous stirring for 30 minutes.

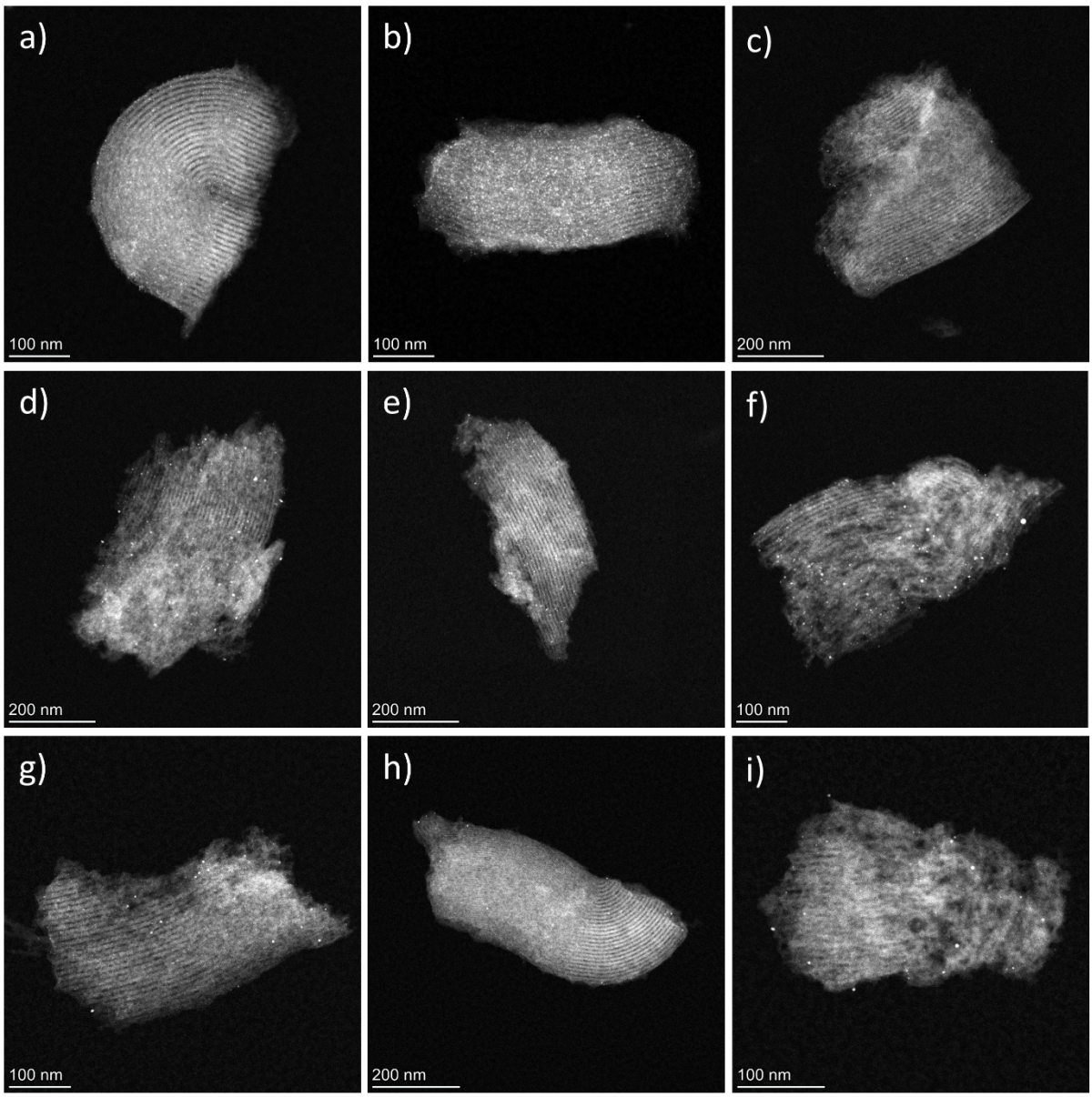

Figure S2. HAADF-STEM images of Pd@CMK3: (a-c) fresh sample and after FA decomposition in (d-f) the batch reactor and (g-i) the fixed bed reactor.

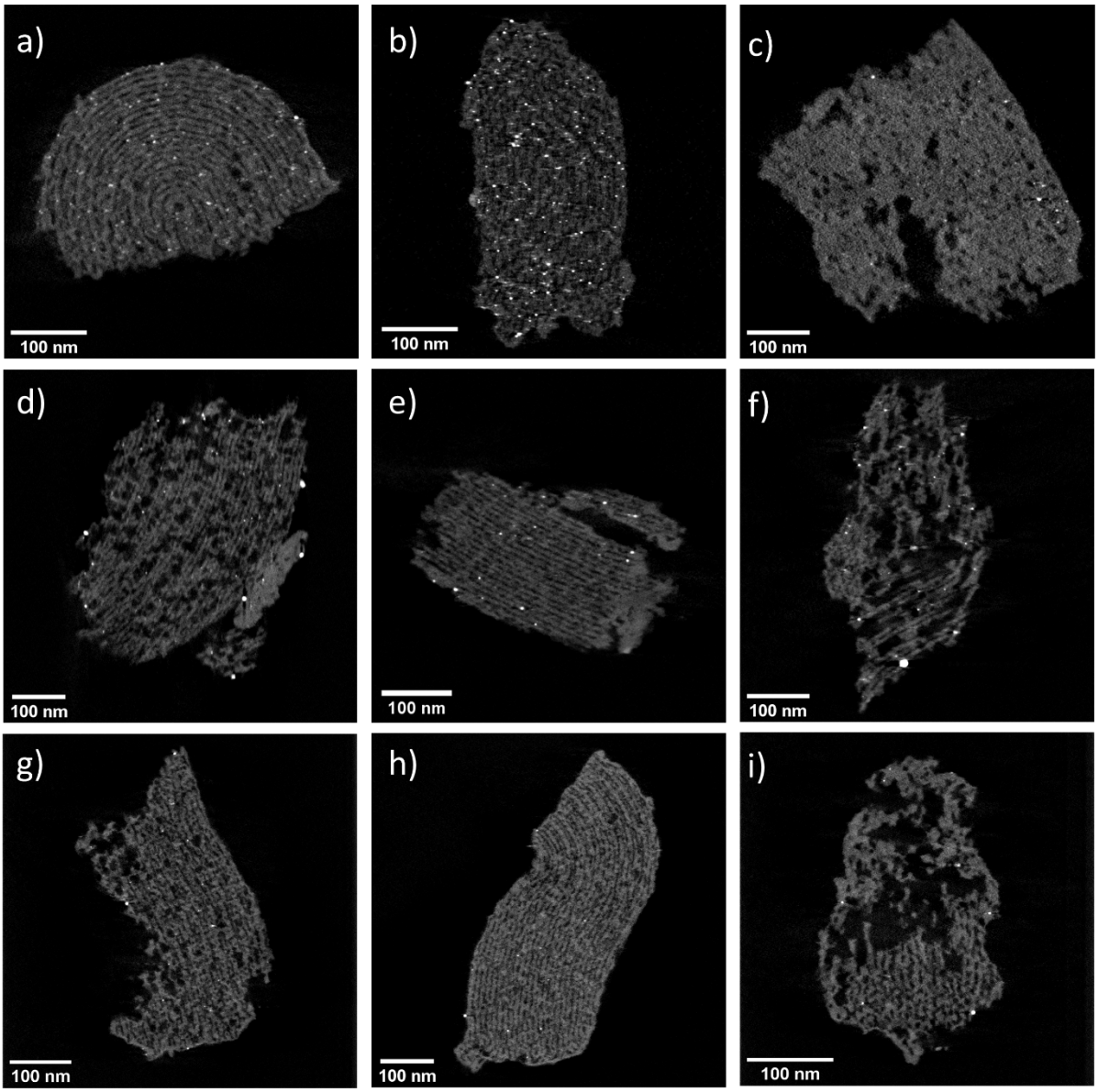

Figure S3. Representative slices from the initial SIRT reconstruction of Pd@CMK3: (a-c) fresh sample and after FA decomposition in (d-f) the batch reactor and (g-i) the fixed bed reactor.

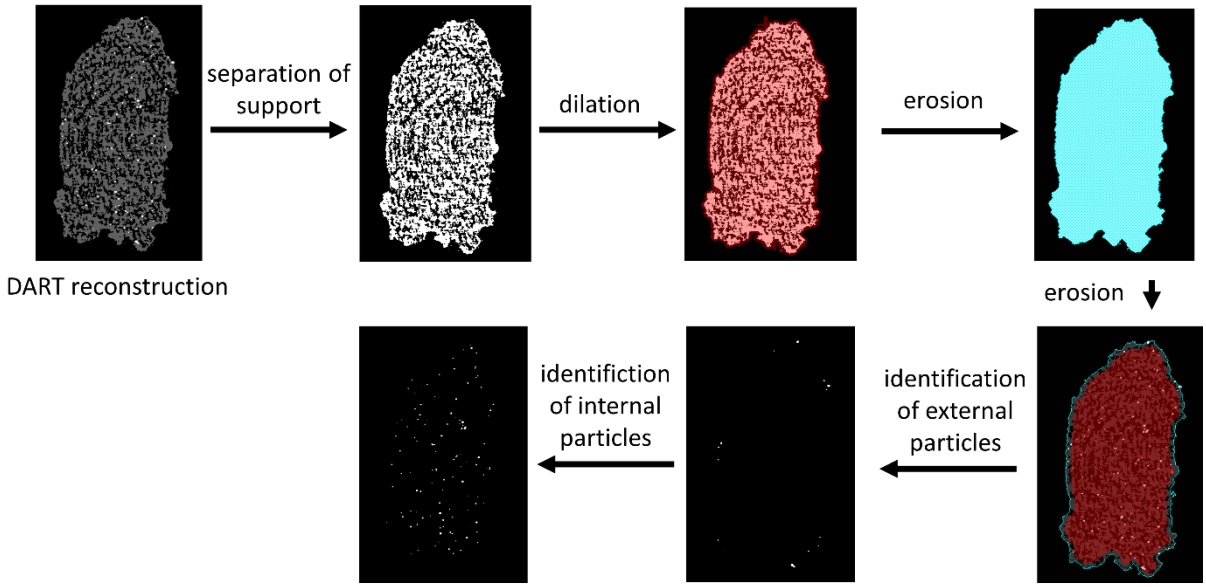

Figure S4. Workflow for determination of the 3D Pd location on Pd@CMK3.

Supplementary Note 1

To identify the 3D location of Pd NPs, a similar but not identical procedure as reported by Wu et al. [1] was used. Based on the DART reconstruction, it is quite straightforward to extract the features of interests, e.g. carbon support and Pd NPs. However, as shown in Fig. S3, it is difficult to determine the internal and external surface of the carbon support by simple edge detection due to the existence of a large number of pores inside the volume. Therefore, the segmented volume was dilated by certain number of voxels (depending on the pore size) using the 'grow' module in Avizo to expand the solid phase and reduce the pore space, thus filling all internal pores. The resulting volume was then eroded by the same number of voxels as previously used during dilation using the 'shrink' module. The resulting volume without pores was further eroded by another certain voxels (depending on the pixel size) to provide a mask for the detection particles on the outer surface. This erosion step is essential to avoid recognizing the intensity gradient between metal particles and vacuum as carbon. In this case, Pd particles located outside the eroded mask are considered as particles on the external surface when they are in contact with vacuum. The others particles located inside the eroded mask are regarded as particles on internal pores.

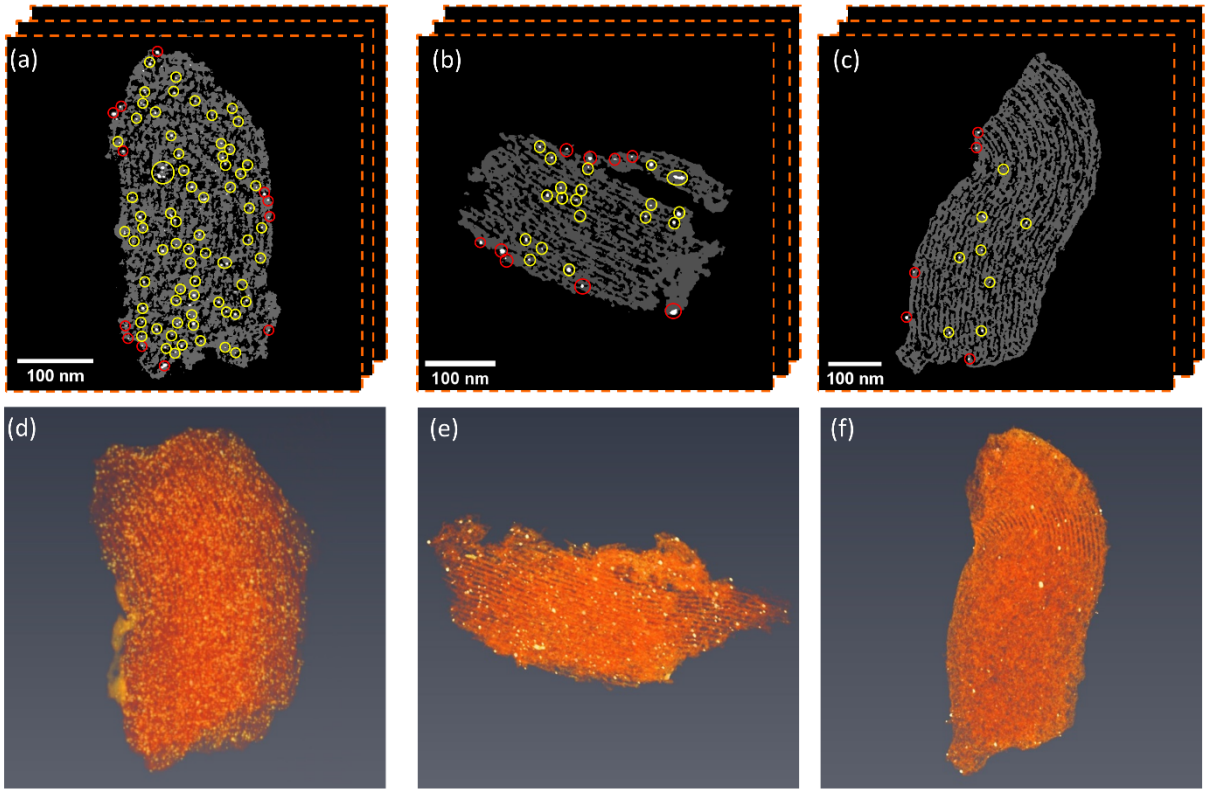

Figure S5. Typical 2D slices and corresponding volume rendering from the reconstructed volume of Pd@CMK3 (a,d) before and after FA decomposition in (b,e) the batch and (c,f) th fixed bed reactor.

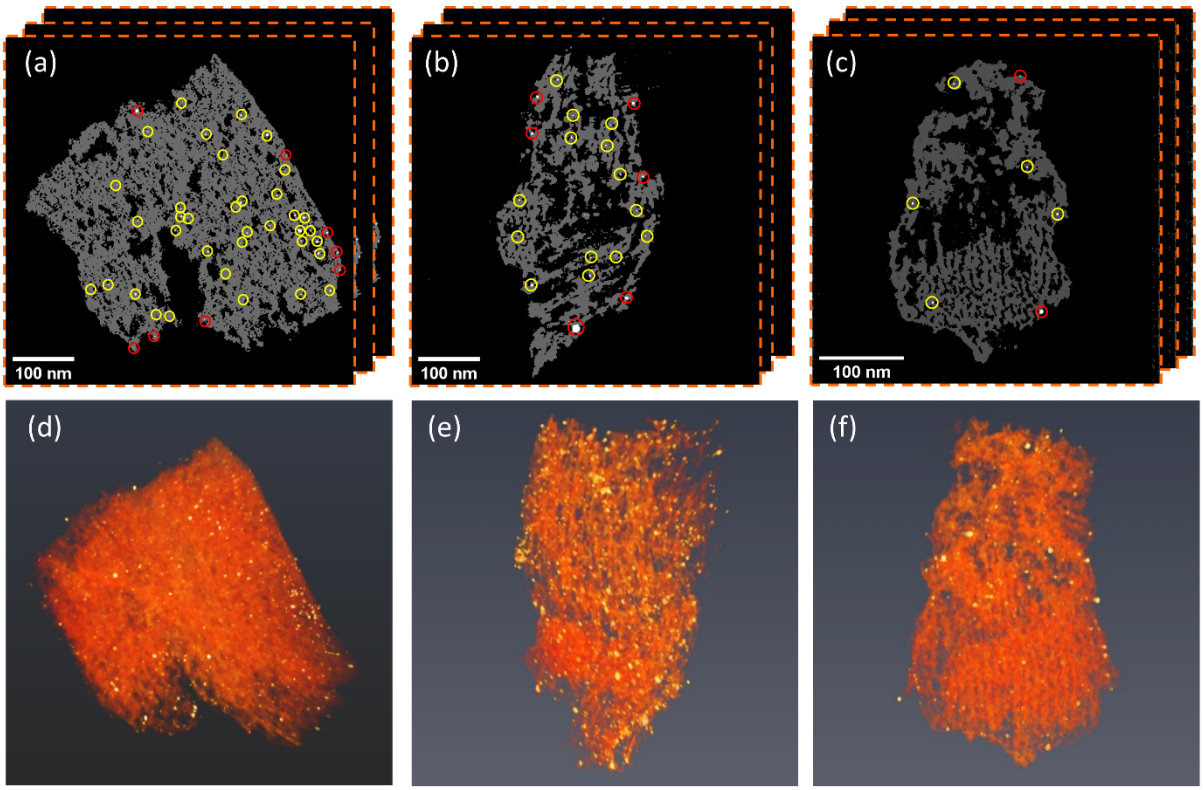

Figure S6. Typical 2D slices and corresponding volume rendering from the reconstructed volume of Pd@CMK3 (a,d) before and after FA decomposition reaction in (b,e) the batch and (c,f) the fixed bed reactor.

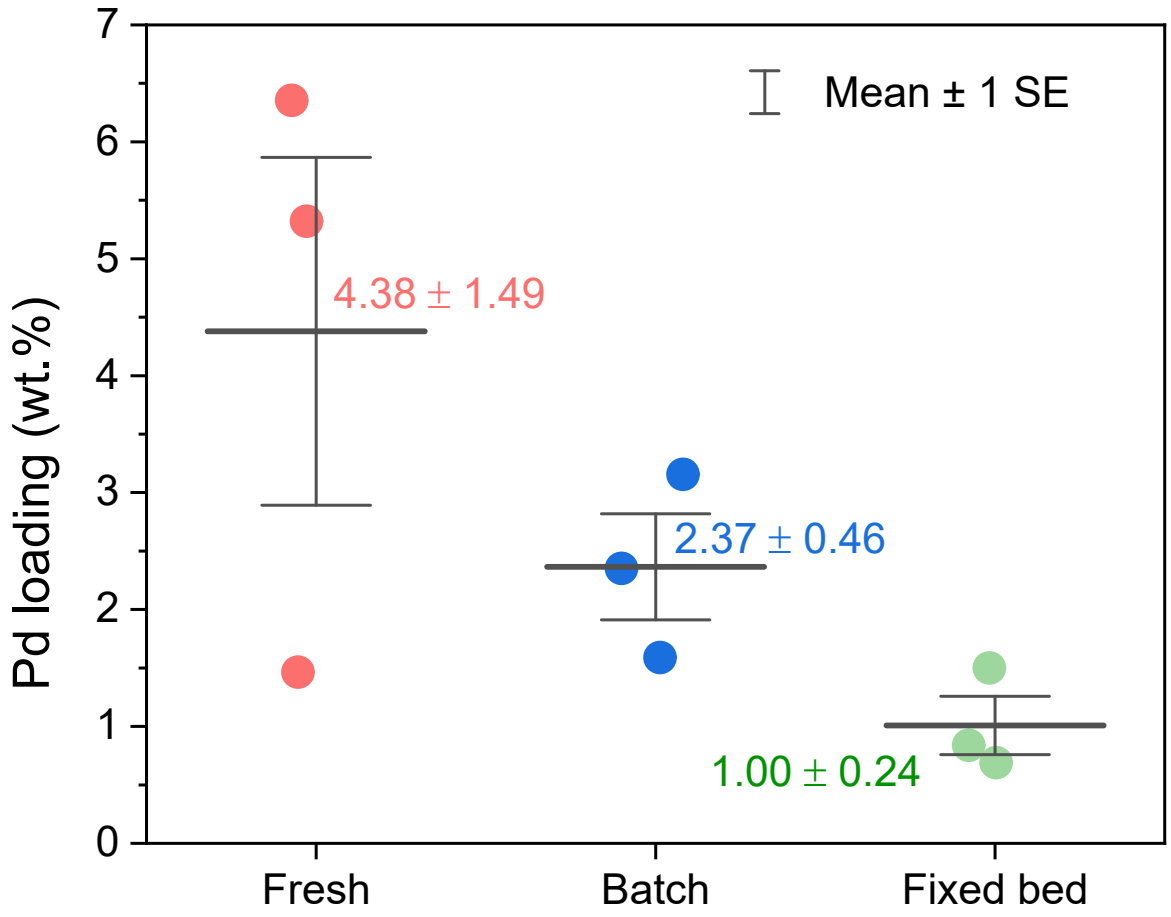

Figure S7. Pd loading calculated from the tomographic reconstructions. The higher loading compared to the EDX and ICP-AES analysis stems probably from an operator bias during selection of loaded catalyst particles.

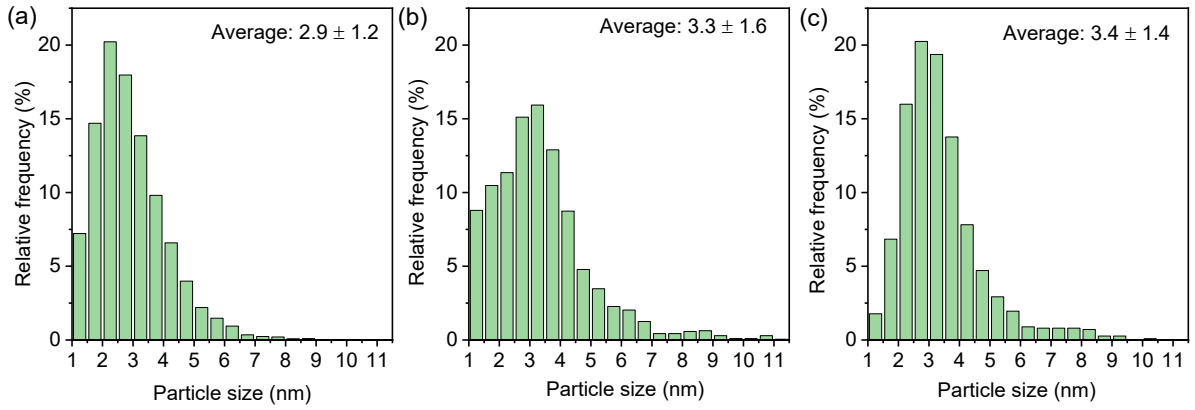

Figure S8. Pd size distribution calculated from the tomographic reconstructions: (a) fresh sample, (b) used sample in batch reactor and (c) used sample in fixed bed reactor.

References

1. Wang, W., et al., Tailoring the 3D Structure of Pd Nanocatalysts Supported on Mesoporous Carbon for Furfural Hydrogenation. ChemNanoMat, 2018. 4(11): p. 1125-1132.