In situ gravimetric monitoring of Atomic Layer Deposition on powders using a thermal magnetic suspension balance

Verena E. Strempel, Raoul Naumann d’Alnoncourt, Matthias Driess, Frank Rosowski

Beside the predominantly use as a deposition technique for thin films, Atomic Layer Deposition (ALD) developed to a valuable tool for the synthesis of heterogeneous catalysts. Few ALD cycles (≤ 10-20) can lead to homogeneously distributed precursor deposits, which can act as active sites or promoters in catalysis. Usually the first ALD cycles differ in mechanism and growth from the following cycles since they occur mostly on the surface of the original substrate.

As our main application of ALD is the synthesis of catalysts in a submonolayer regime, a better understanding of the first couple of cycles on our powder substrates is of great importance. On that account we built an ALD setup with an integrated in situ gravimetric monitoring of the deposition using a thermal magnetic suspension balance. As far as we know, this is the first attempt of a gravimetric in situ analysis of the ALD process directly on powders. The commonly used in situ analysis with a Quartz Crystal Microbalance (QCM) can solely give information about the growth thickness on the SiO2 crystal or on preliminary grown films on this crystal. Our setup offers the possibility to analyze the deposition process in real-time directly on our porous, high-surface powder substrates. Due to the special design of the balance, it is easily possible to transfer and scale-up to ALD in a fixed-bed reactor.

The setup has been successfully employed in the modification and synthesis of different catalysts. In the current contribution we focus on in situ analyses of elaborately investigated ALD processes to demonstrate the setups utility. We present the deposition of e.g. Al2O3 or ZnO, even though they feature no relevant catalytic process. The presented figure shows first results of the percentage mass gain in the first three ALD cycles for the deposition of Al2O3 on SiO2 powder at 120 °C (Davisil, grade 636, 35-60 mesh, 611.0 mg). Trimethylaluminium (TMA) and H2O served as precursors and Argon as the constant carrier flow (50 mL min⁻¹). The dose and purge procedure is visualized in the upper part of the graph. Mass gains are around 15 % for each cycle, although it is clearly shown, that the gain portions differently between the TMA and H2O steps from cycle to cycle. Saturation points during the precursor dosing are well-defined. By combining our gravimetric data with online mass spectrometry, QCM measurements and further chemical analysis of the samples, we can gain new insights into ALD growth on powders.
