

PREPARATION OF NOVEL PT-, PD- AND PT-PD/AL₂O₃ MODEL CATALYSTS BY MEANS OF SUPERCRITICAL FLUID REACTIVE DEPOSITION

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Noble metal (e.g. Pt and Pd) nanoparticles with high surface area are currently used extensively as efficient catalysts for chemical reactions such as hydrogenation, hydration and oxidation [1-3]. Such catalysts are often prepared by wet impregnation of a porous support with a metal-containing solution, followed by reductive treatment and drying. Catalysts prepared by this procedure yield to metal particles with broad size distribution and to large volumes of waste water.

A promising environment-friendly alternative to this conventional preparation route is the use of the Supercritical Fluid Reactive Deposition (SFRD) to achieve highly dispersed monometallic (e.g. Pt or Pd) or bimetallic (e.g. Pd-Pt) nanoparticles ($d_{50} < 10$ nm) on diverse supports (e.g. TiO₂, Al₂O₃, Black Pearls, Carbon nanotubes, ...) [4,5]. In the SFRD process, the supercritical fluid (SCF) acts as a solvent, reaction and separation media. The SFRD technique involves the dissolution of the organometallic precursor in a SCF (e.g. scCO₂) and the exposure of a support to the solution. After sufficient time for impregnation, the precursor will be transformed to its metal form with a reducing agent, such as hydrogen. Thereafter, the system is slowly depressurized and cooled down to ambient conditions. Since both, CO₂ and the reaction products are in the gaseous state, phase separation can be easily realized and a solvent free product is obtained. Thus, the SFRD technique represents an integrated process that enables process improvement because it combines several steps of the conventional process.

In our experiments, the influence of various process conditions such as precursor, substrate and reduction conditions on particle size, size distribution, and metal loading was investigated. The product was characterized by Scanning and (High Resolution) Transmission Electron Microscopy (SEM and HRTEM), Energy-Dispersive X-ray Spectroscopy (EDXS), Powder X-ray Diffraction (PXRD), and Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) analyses.

Among others, additional experiments show that the catalysts prepared by SFRD possess a superior activity towards oxidation of CO and NO. Furthermore SFRD prepared Pt catalysts lead to improved results compared to catalysts prepared by conventional methods. The talk will give an introduction into the basics of the SFRD process. Based on this, typical results obtained from SFRD experiments are presented and discussed in detail. At the end of the talk the main conclusions and further perspectives are summarized and discussed.

In conclusion, the results of our investigations show that:

- 1) The SFRD process enables the deposition of uniform Pt, Pd and bimetallic (Pt-Pd) nanoparticles on Al₂O₃.
- 2) The average particle size and size distribution can be affected by type and amount of the precursor in the system, precursor reduction method and condition, surface properties (surface area and chemical nature) of the support [5].
- 3) Furthermore, the results obtained from CO oxidation experiments show that the product characteristics of the catalysts (light-off temperature and conversion) are strongly influenced by the preparation method (i.e. flame spray pyrolysis, wet impregnation, SFRD).
- 4) The catalysts prepared by SFRD exhibit an activity higher than reference samples prepared by conventional methods.
- 5) The lowest “light-off temperature” was shown by the Pt/Al₂O₃ sample prepared by SFRD.
- 6) The highest NO conversion during NO/CO oxidation was obtained for the Pt-Pd/Al₂O₃ sample prepared by SFRD.

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