Wrocław, 15.11.2019 r.

PhD thesis abstract

Synthesis, physicochemical characterization and catalytic properties of the nanostructured Au/SBA-15 catalysts

mgr inż. Piotr Kraszkiewicz

Supervisor: dr hab. Włodzimierz Miśta
This work was done in the Department of Nanomaterials Chemistry and Catalysis

Gold is known for its chemical inertness and for a long time it was thought that it will not find practical application in heterogeneous catalysis. The discoveries of the 1980s have shown that gold exhibits high catalytic activity only when it is in the form of particles smaller than 5 nm. Currently, due to their unique properties, catalysts based on gold clusters – gold particles with a size below 2 nm – are of particular interest. Synthesis of these materials is challenging, but even more problematic is their low thermal stability. The growth of gold particles irreversibly reduces the activity of the gold-based catalysts.

The aim of this work was the synthesis of Au/SBA-15 catalyst, containing gold particles with an average size below 2 nm, characterized by high thermal stability as well as high activity in the low-temperature CO oxidation.

The adsorption of the cationic complex $[Au(en)_2]^{3+}$ (en = ethylenediamine) on the surface of the SBA-15 mesoporous silica support was chosen as the catalyst preparation method. This method allowed the effective introduction of gold into the pores of the support and a high degree of gold dispersion. Placing gold in a system of parallel, few-nanometer pores of the SBA-15 support was aimed at limiting sintering of gold particles.

The synthesized materials were characterized by XRD, TEM, SEM-EDS, UV-Vis and nitrogen adsorption/desorption at 77 K. Much attention was paid to the optimization of the precursor decomposition process and catalyst activation in various atmospheres (H_2, O_2, inert). The analysis of gaseous products formed at different stages of catalyst activation using a mass spectrometer, enabled the proposition of a mechanism of gold particles stabilization on the SBA-15 support. The activity of the obtained catalysts was tested in the CO oxidation and selective CO oxidation in hydrogen rich gas (PROX).

According to the set goal, a Au/SBA-15 catalyst, containing gold particles with an average size below 2 nm, showing high thermal stability and activity in low-temperature CO oxidation (nearly 100% CO conversion at 25 ° C), was obtained. Particularly noteworthy is the unusually high stability of the catalyst at temperatures of 700 – 800 ° C under CO oxidation. It was found that a special procedure for the catalyst precursor decomposition in hydrogen, during which a strong Au-SiO_2 interaction is formed, plays a key role in stabilizing gold particles on the surface of the SBA-15 support. During the precursor decomposition, a thin layer of carbonaceous material forms, covering the surface of gold clusters. The carbonaceous material probably prevents sintering of gold in the initial stage of precursor decomposition.

The high activity of the Au/SBA-15 catalyst in low-temperature CO oxidation was primarily associated with the very small size of the gold particles. It was also found that doping the catalyst with sodium significantly increases its activity in the CO oxidation reaction. Catalytic activity studies in the PROX reaction have shown that the obtained Au/SBA-15 catalyst effectively remove carbon monoxide from a hydrogen-rich gas below 100 °C and, after further optimization, can be used in purification of hydrogen for fuel cells.