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Nanogold and nanosilver catalysts for CO and octanol oxidation

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Setting aside the environmental aspect, the transformation of fatty alcohols as a source of fine chemicals is commercially attractive for pharmaceutical, agrochemical and fragrance industries, among others. Selective oxidation of *n*-octanol (as a model for fatty alkanols C8+) has attracted much less attention as compared with the oxidation of more reactive benzyl or C1-C4 aliphatic alcohols. Although non-activated, primary aliphatic alcohols are the most difficult type of alcohols to oxidize selectively, 1-octanol has been used as reference of alkanols in comparative studies of alcohols reactivity against different catalysts. Currently there is a great interest in the catalytic activity of Au and Ag nanoparticles (NPs) for the selective oxidation of *n*-octanol. CO oxidation is very convenient model reaction for testing catalytic activity of the metals.

The aim of this work is to compare the catalytic activity of Au and Ag NPs supported in titania for liquid phase *n*-octanol oxidation as well as gas phase CO oxidation under two treatments: a) modification with metal oxides and b) treatment with H₂ and O₂ atmospheres. It is worth nothing that those two parameters are systematically compared for the first time in systems containing Au and Ag NPs for *n*-octanol oxidation.

Modifiers (M=Ce, La, Fe or Mg) were deposited on TiO₂ support by impregnation with aqueous solution from nitrates (molar ratio Ti/M = 40). The nominal Au and Ag loadings were 4 and 2.3 wt.%, respectively. Catalytic properties of samples were studied in *n*-octanol oxidation without treatment and also with treatment at 300 °C in H₂ or O₂ atmosphere. Reaction conditions were as follows, 0.1 M *n*-octanol in *n*-heptane, no base added, and at T = 80 °C with a molar ratio *n*-octanol/Au or Ag=100 for 6 h. Catalytic tests in CO oxidation were carried out in a flow reactor using a gas mixture of 5% CO + 5% O₂ + 90% Ar (flow rate 200 ml/min). Characterizations of samples were made with FTIR, CO, XPS, HRTEM, etc.

The results show a significant influence of both titania modifier (Ce, La, Fe or Mg oxides) and pretreatment in hydrogen or oxygen, upon the selectivity and yield of *n*-octanal formation as well as CO conversion. It was found that modifiers influence the structural and electronic properties of gold and silver nanoparticles through a metal-support and metal-modifier interaction. It is interesting that pre-treatment in reactive atmospheres has a different effect on the activity of silver and gold metals despite of their similar chemical nature.

According to our results, Au catalysts showed the higher activity under mild conditions. Moreover, it was revealed that silver catalyst, as an analog of gold catalyst, showed a 12 % of *n*-octanol conversion after treatment. Modified Ag catalyst can be considered as a promising alternative system for *n*-octanol oxidation because the price of Ag is about 20 times lower than Au. Finally, modification of the systems with metal oxide together with processing with reactive atmospheres enhanced up to 6 and 10 times the catalytic activity of silver and gold catalysts, respectively.

Comparison of catalytic and spectroscopic data showed that partly charged metal clusters Au_n^{δ+} and Ag_n^{δ+} are probable active sites of the catalysts in the studied process. Modifying additions of metal oxides influence structural and electronic properties of Au and Ag nanoparticles and stabilize their active forms.