Support Effect in High Activity Gold Catalysts for CO Oxidation

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Abstract: In this work, we present a detailed study concerning the evaluation of the metal—support interaction in high activity gold catalysts for CO oxidation. Using the colloidal deposition method, model catalysts were prepared, which allow the isolation of the effect of the support on the catalytic activity. Prefabricated gold particles were thus deposited on different support materials. Since the deposition process did not change the particle sizes of the gold particles, only the influence of the support could be studied. TiO2, Al2O3, ZrO2, and ZnO were used as support materials. Catalytic tests and high resolution transmission electron microscopy clearly show that the support contributes to the activity. However, our results are not in line with the distinction between active and passive supports based on the semiconducting properties of the oxic material. The most active catalysts were obtained with TiO2 and Al2O3, while ZnO and ZrO2 gave substantially less active catalysts. Furthermore, the effect of other important parameters on the catalytic activity (i.e., particles size distribution, calcination temperature, and aging time for a Au/TiO2 catalyst) has also been studied. Using this preparation route, the catalysts show high-temperature stability, size dependent activity, and a very good long-term stability.

Introduction

Gold had long been disregarded for catalytic applications, due to its inert nature in the bulk state. However, when it is very highly dispersed on different supports, it exhibits a surprisingly high activity for several reactions, both in the liquid and in the gas phase. Thus, since Haruta discovered a remarkable activity of supported gold nanoparticles in CO oxidation, different methods to prepare high activity gold catalysts have been developed, and several potential application fields have been explored.1–8

The availability of efficient gold catalysts was then the driving force of many publications devoted to the analysis of the different factors controlling the activity. Unfortunately, despite numerous studies published on supported gold catalysts, there is still no clear picture with respect to the origin of the catalytic activity, and often, the results reported in the literature concerning gold catalysis are contradictory. The role of the support is still under discussion, and recent reports have also demonstrated that unsupported gold particles can exhibit a comparable activity to the supported ones in the aerobic glucose oxidation.9 In the CO oxidation reaction, highly controversial are the mechanisms for oxygen adsorption and activation, which are required to explain the high activity of gold nanoparticles and that contrast the inhibited oxygen dissociation on single crystals.10 Several models have been proposed to resolve this discrepancy. In the probably most widely accepted of them, oxygen adsorption is believed to occur on the support or at the metal support interface, possibly in oxygen vacancies that should be present on semiconductor materials (e.g., TiO2 and ZnO) as a consequence of the Schottky junction at the metal—support interface. Consequently, the different activities observed for different catalysts were attributed to the different conducting properties of the material used as support.11

To gain further insight in this controversial issue, and to also obtain a complete picture on how a good gold catalyst should be prepared, pretreatment and preparation conditions, reaction conditions, and many other physicochemical parameters have been investigated.12–16 Among them, the most relevant seem to be particle size, oxidation state of the gold species, nature of the material used as support.