

CO oxidation on gold nanoparticles supported over titanium oxide nanotubes

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Introduction

Titanium oxide nanotubes (TNT) prepared by hydrothermal synthesis have attracted special attention since 1998 [1-3]. This material is interesting because its high specific area and low cost. One of its applications is as support for catalysts [4-5]. Gold is a metal with no reactivity in bulk, but if it is divided into nanoparticles has a high activity for some reactions like CO oxidation [6]. High reactivity of gold is found in nanoparticles lower than 5 nm [6]. An adequate method to obtain small gold nanoparticles is deposition-precipitation with urea, developed by Louis team [7], based in method of Haruta [6] that allow to obtain gold nanoparticles lower than 5 nm necessary for high catalytic activity. In this paper the possibility to obtain highly active catalysts for CO oxidation by means of deposition of gold nanoparticles lower than 5 nm over TNT is analyzed. In this work, the changes in activity that catalysts undergo when the support calcination temperature is varied, the storing time of catalysts, gold charge and pretreatment conditions before reaction (temperature and gas type) are analyzed. To this end, supports and catalysts will be characterized by means of N₂ fisisorption, XRD and TEM, and they will be evaluated in CO oxidation in a programmed temperature reaction system.

Materials and Methods

TNT were prepared as described elsewhere [1], keeping mixture at 110°C during 24 h. Material obtained was calcined in static air at 300-500°C during 4 hours. Gold was deposited on calcined TNT by deposition-precipitation with urea method using as precursor HAuCl₄ · 3 H₂O according to method described elsewhere [7]. TEM images were obtained using a JEOL 2010 transmission electron microscope at 200 kV, samples were taken without dispersion on a holey carbon film supported on a copper grid. Catalytic experiments were performed in a In Situ Research RIG-150 catalyst characterization system, using a fixed bed quartz reactor with 42.5 mg of catalyst and with a gas flow of 100 mL/min; gas mixture (98% of N₂, 1% of CO and 1% of O₂). CO and CO₂ detection was carried out in a 6890N Network GC chromatographic system. Catalysts tests were carried out as oxidation to programmed temperature, from -5 to 300°C with a linear ramp of 2°C/min. Sample is pretreated in situ with air or hydrogen with a flow of 80 mL/min, heating up to 200-400°C during 2 h.

Results and Discussion

Fig. 1 shows activity of catalysts with supports calcined at different temperatures. Catalyst with support calcined at 300°C had low activity compared with catalysts which supports were calcined at 400° and 500°C; as a matter of fact, these two cases had practically

the same activity, reaching 100% of conversion at 200°C for catalyst with support calcined at 400°C, and last catalyst reached 99% at 240°C. TEM images of these catalysts show that sizes of gold nanoparticles are in agreement with catalysts activity, since catalyst with support calcined at 300°C has an average size particle of 4.39 nm, being the less active catalyst, while that one calcined at 400°C has nanoparticles with average size of 2.26 and the calcined at 500°C has 2.42 nm; these two have high activity and very similar, although material calcined at 400°C has the smallest nanoparticles, and therefore is scarcely more active.

Significance

Data could be applied to establish conditions and parameters of preparation of more active catalysts.

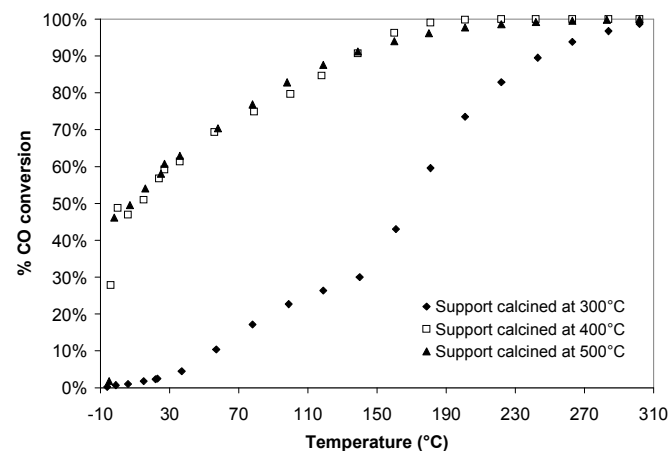


Figure 1. CO oxidation on gold nanoparticles (3 wt. %) over TNT calcined at different temperatures.

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