

Gold Catalyst Supported on Ce_{0.5}Zr_{0.5}O₂-Al₂O₃ Prepared by Sol Gel

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Introduction

While gold has been demonstrated to be chemically most inactive among noble metals due to its electronic configuration [1], matrix-trapped finely dispersed Au species are known to be highly reactive for CO oxidation even at low temperature [2]. The catalytic activity of gold species depends on its electronic state. Gold species being deposited over some oxides undergo electronic transformations in dependence on the nature of supports used [3]. Recently it was found that gold deposited over nanocrystalline ceria is high active in the different reactions [4,5]. The ability of ceria to be reduced or oxidized depends on the presence of structural defects in ceria crystals. The number of structural defects could be controlled by partial replacement of Ce atoms with Zr atoms [6]. The present paper was devoted to study the gold species supported on nanosized ceria-zirconia mixed oxide stabilized in alumina.

Materials and Methods

The Al-Ce-Zr-O mixed oxide was prepared by sol gel technique described elsewhere [6] using organic precursors. Content of Ce_{0.5}Zr_{0.5}O₂ mixed oxide in alumina was about 10 wt.%. Gold (3wt.%) was deposited on calcined supports by incipient wet impregnation using Au(NH₃)₄(NO₃)₃ complex as gold precursor according to method described elsewhere [7]. At the end of preparation samples were dried at room temperature. TPR profiles of fresh and treated samples were obtained using gas mixture 5% H₂+He and in-line analysis with mass spectrometer HPR 20 (Hiden). XPS analysis of catalysts has been carried out using a Cameca-3 (Riber) spectrometer. Catalytic activity test was carried out in a flow micro reactor using gas mixture 1% CO + 1% O₂ (rest helium) and in-line mass spectroscopic analysis (HPR 20, Hiden). Contact time was 12 (g_{CAT}×h×mol_{CO}⁻¹). Before activity test samples were treated in situ with He or He+H₂ flow with temperature ramp 20K/min up to 773K. The same concentrations of CO and O₂ were used in pulse transient experiments.

Results and Discussion

Interaction of gold precursor with alumina doped by nanosized Ce_{0.5}Zr_{0.5}O₂ mixed oxide is stronger than with reference sample of pure alumina which is manifested by different temperature range in desorption of products of gold precursor decomposition and hydrogen consumption with temperature increase. Reduction of gold species by hydrogen proceeds more intensively after partial thermal decomposition of gold precursor.

Deposition of gold over nanosized Ce_{0.5}Zr_{0.5}O₂ mixed oxide permits to stabilize different gold species according to XPS data (Figure 1). The curve fitting of the spectra Au 4f shown in Figure 1 indicates following Au 4f_{7/2} components at BE = 82.7, 84.1, 85.5, 86.5 eV that can be attributed to Au^{-δ} (finely dispersed partly charged gold particles), Au⁰, Au^{+δ} and

Au⁺³ according to [8]. Even after so deep reduction in TPR with H₂ sample being exposed to air at room temperature is characterized by the presence of Au⁺ and Au³⁺ cations (Figure 1,B).

The prepared catalyst is characterized with high activity at 298K. CO conversion was higher than that obtained by Corma [5] for gold supported over nanocrystalline ceria only.

The nature of dynamic changes of activity with time, temperature and sample pretreatment was studied using time resolved kinetic analysis (pulse and transient techniques).

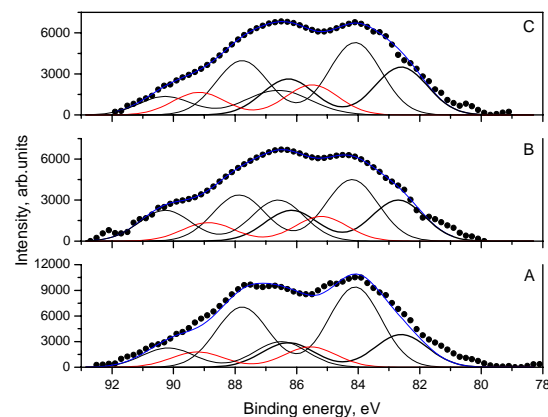


Figure 1. XPS data for Au/Ce_{0.5}Zr_{0.5}O₂-Al₂O₃ catalyst after different pretreatments:

A- fresh, B – after TPR, C – after TPR and catalytic CO oxidation at 353K.

Significance

Data obtained could be applied for the development of new effective gold-based catalysts for environment protection.

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