

XAFS study of gold catalysts supported on nanostructured alumina and alumina doped with Ce-Zr mixed oxides

V.V. Kriventsov¹, I.L. Simakova¹, A. Simakov^{2*}, E. Smolentseva², F. Castillon², M. Estrada³, E. Vargas⁴, D.P. Ivanov¹, B.N. Novgorodov¹, D.I. Kochubey¹, S. Fuentes²

¹Boriskov Institute of Catalysis, Novosibirsk, 630390 (Russia)

²CNIN-Universidad Nacional Autónoma de México, Ensenada, B.C., 22800 (México)

³Posgrado de Física de Materiales de CICESE, Ensenada, B.C., 22800 (México)

⁴Posgrado de Nanomateriales de UABC, Ensenada, B.C., 22800 (México)

*andrey@cnyn.unam.mx

Introduction

The matrix-trapped finely dispersed gold species are known to be highly catalytically active in some industrially important reactions [1,2]. Thus gold is a potential catalyst for selective isomerization of linoleic acid, a side stream from chemical pulping, to conjugated linoleic acids that are healthy promoting agents in food and pharmaceuticals having anticarcinogenic and antioxidative properties [3].

The present work is devoted to a structural analysis of the nanosized gold species supported on nanostructured alumina and alumina doped with Ce-Zr mixed oxides.

Materials and Methods

The supports based on nano crystalline pure alumina, pure ceria, alumina doped by ceria (10 and 30%), alumina doped with CeZrO₂ (10 and 30%) have been prepared by sol-gel technique using organic precursors [4]. Gold (3wt.%) catalysts were prepared by DP technique. Before test two portions of samples were reduced at 150°C and 300°C, respectively.

The XPS spectra of prepared samples were obtained with the photoelectron spectrometer Riber-Cameka Mac-3 using AlK_α radiation (hν=1486.6 eV). X-ray diffraction (XRD) analysis of samples was carried out with a Philips X'pert diffractometer equipped with a curved graphite monochromator applying CuK_α radiation. Analysis of gold particles distribution in size was performed by transmission electron microscopy (TEM) with a JEOL 2010 microscope. XAFS spectra of the Au-L₃ edges for all the samples studied were measured (transmission mode) and treated by the standard procedure [5] at SSRC, Novosibirsk.

Results and Discussion

The samples reduced at 150°C contain mainly Au³⁺ cations located on the surface of support in a slightly distorted octahedral coordination. The samples reduced at 300°C contain mainly distorted metallic gold species with coordination numbers of the shortest Au-Au bond for these samples: Au-Al₂O₃ catalyst - R_{Au-Au}~2.82 Å, CN~6.3; Au-Al₂O₃-CeZrO₂ catalyst - R_{Au-Au}~2.81 Å, CN~5.8; Au-Al₂O₃-CeO₂ catalyst - R_{Au-Au}~2.84 Å, CN~6.9. The dispersed Al-Ce-Zr-O mixed oxides prepared by sol-gel are highly effective for stabilization of gold metal particles with the characteristic diameter less than 2 nm and particular extra finely dispersed ones with the diameter less than 1 nm.

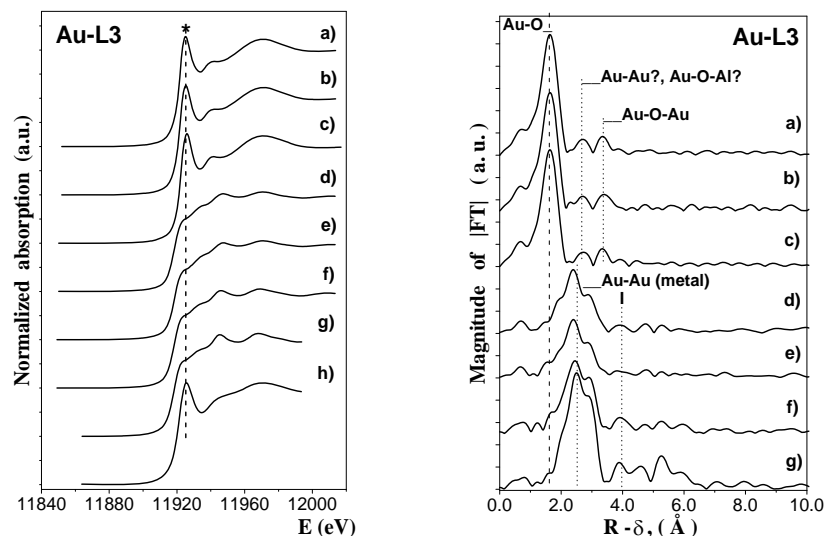


Fig.1 (Left) Normalized XANES spectra (Au-L₃ edge, *- white line) for samples studied: T=150°C - a) Au-Al₂O₃, b) Au-Al₂O₃-CeZrO₂, c) Au-Al₂O₃-CeO₂; T=300°C - d) Au-Al₂O₃, e) Au-Al₂O₃-CeZrO₂, f) Au-Al₂O₃-CeO₂; g) Au-foil; h) Au₂O₃ reference [11]. **(Right)** RDFs describing of Au local arrangement for the same samples.

Significance

Reliable analysis of the nature of gold species is important to design of new effective gold based catalysts for different applications.

Acknowledgements

This research was supported by RFBR (08-03-91758-AF-a), RFBR (06-03-33005-a), RFBR (08-03-01150-a), RFBR-CNRS (08-03-92502a), RFBR (09-03-01012a) grants and by DGAPA -PAPIIT (UNAM, Mexico) through Grant Numbers IN 120706-3, IN 110208 and CONACyT research grant 50547-Q. The authors also acknowledges to J.A. Díaz and E. Flores for their technical help.

References

1. B.Hammer, J.K. Norskov, Nature 238 (1995) 376.
2. V.Aguilar-Guerrero, B.C. Gates, Chemical communications (Cambridge, England), 30 (2007) 3210.
3. M.Haruta, N. Yamada, T. Kobayashi, S. Iijima, J.Catal. 115 (1989) 301.
4. G.Perez, S.Fuentes, V.Petranovskii, A.Simakov, Catalysis Letters, 110 (2006) 53.
5. P.Concepcion, S.Carrettin, A.Corma, Applied Catalysis A, 307(2006)42.