343a Deactivation of Supported Nanocrystalline Gold Water Gas Shift Catalysts

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Nanocrystalline gold catalysts have recently been demonstrated to be highly active for reactions including the water gas shift (WGS)[1, 2], and their high activities have been linked to the size and structure of gold nano particles. The WGS is a key reaction in the production of hydrogen for a number of processes including fuel cell applications, however, commercial WGS catalysts are not suitable for portable and vehicular applications due to insufficient durabilities and activities. Consequently there is substantial interest in the development of better performing and more durable WGS catalysts. We prepared a series of Au/CeO_2 catalysts that were initially as much as four times more active than commercial Cu-Zn-Al catalyst. These materials deactivated quickly in a simulated reformate mixture containing CO, H_2O, CO_2, H_2, and N_2 [3].

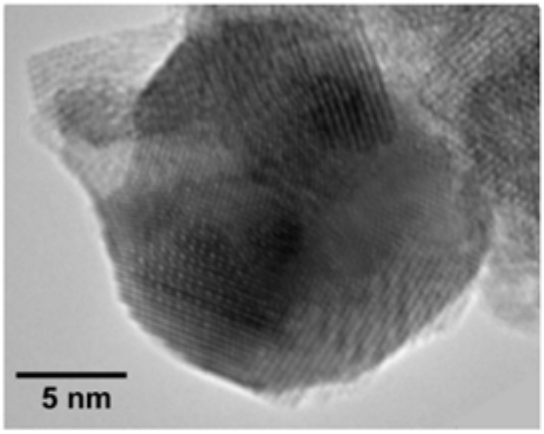

Figure 1. Micrograph of fresh Au/CeO_2 catalyst

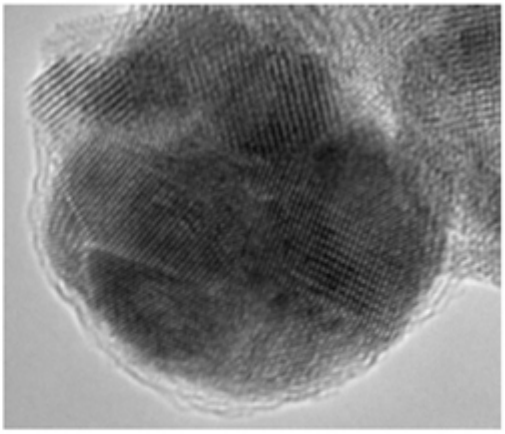

Figure 2. Micrograph of deactivated Au/CeO_2

Characterization of the physical and chemical properties of the fresh and used catalysts helped define the deactivation mechanism. Figure 1 and 2 illustrate morphological changes that occurred during deactivation. The nano-sized gold particles appeared to be covered by a low density, amorphous layer. Based on characterization using transmission electron microscopy, x-ray photoelectron spectroscopy and infrared spectroscopy, we propose that deactivation of the Au/CeO_2 catalyst was caused by poisoning of the active sites by formates and/or carbonates. These and other results will be discussed.

[1] M. Haruta, S. Tsubota, T. Kobayashi, H. Kageyama, M. J. Genet and B. Delmon, J. Catal. 144 (1993) 175.

[2] D. Andreeva, V. Idakiev, T. Tabakova and A. Andreev, J. Catal. 158 (1996) 354.

[3] C. H. Kim and L. T. Thompson, J. Catal, 230 (2005) 66.