

Investigation of Chalcogenide and Pnictide Supported Gold Catalysts

Gabriel M. Veith^{1,*}, Andrew R. Lupini, Stephen J. Pennycook, and Nancy J. Dudney
Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN
37831 (United States)

*veithgm@ornl.gov

Introduction

Interest in supported gold catalysts has exploded since the discovery that gold nanoparticles catalyze the low temperature oxidation of carbon monoxide [1]. Factors such as particle size, particle shape, and preparation method are critical to obtain significant catalytic activity; however, the reaction mechanism and the origin of the catalytic activity are not understood. The research reported thus far has revolved around a limited number of oxide (TiO_2 , MgO , Al_2O_3 , etc.) and carbon support materials. Studies indicate that the support material plays a critical role in the observed catalytic activity, with reducible oxides, such as TiO_2 , leading to more active catalysts than stable oxides like SiO_2 and Al_2O_3 [2]. However, several studies report $\text{Au}/\text{Al}_2\text{O}_3$ catalysts that are more active than Au/TiO_2 catalysts [3,4].

It is not clear if the oxide support plays an active role in the catalytic process or merely prevents the catalyst particles from coarsening together. This research investigates non-classical support materials and determines the affect these supports have on the catalytic activity of the supported gold catalysts. Several systems have been studied where the support has the same cation, but the anion differs. For example, a series of gold catalysts were prepared on boron oxide, boron nitride, and boron carbide and evaluated for their catalytic activity.

Materials and Methods

Bulk gold catalysts were prepared on a variety of oxide, nitride, and carbide support materials using the physical vapor deposition technique of magnetron sputtering. Particle size and distribution was determined using an aberration corrected Scanning Transmission Electron Microscope (STEM). The electronic properties of the supported catalysts were investigated using X-ray photoelectron spectroscopy. The activity of these catalysts was determined from the oxidation of carbon monoxide.

Results and Discussion

Since the synthesis technique is independent of the support material's isoelectric point, we can synthesize a series of supported catalysts particles without changing the synthesis procedure. Microscopy results indicate that the magnetron sputtering technique successfully prepared gold nanoparticles that are less than 5 nm in diameter, which is the critical particle size to observe significant catalytic activity for the oxidation of carbon monoxide [5]. An example image of gold nanoparticles supported on boron nitride is given in Figure 1. However, preliminary catalytic activity measurements indicate that the catalysts supported on nitrides are not active for the oxidation of carbon monoxide despite the high fraction of sub-5 nm gold clusters. Mechanistic insights gained from these studies will be used to further understand the role the support plays in the oxidation of carbon monoxide.

Significance

This work addresses the fundamental role of the support material during the oxidation of carbon monoxide by supported gold catalysts. This study allows us to gain insights into the reaction mechanism for the oxidation of carbon monoxide. These studies are critical for the development of new, low-temperature oxidation catalysts for environmental and fuel cell applications.

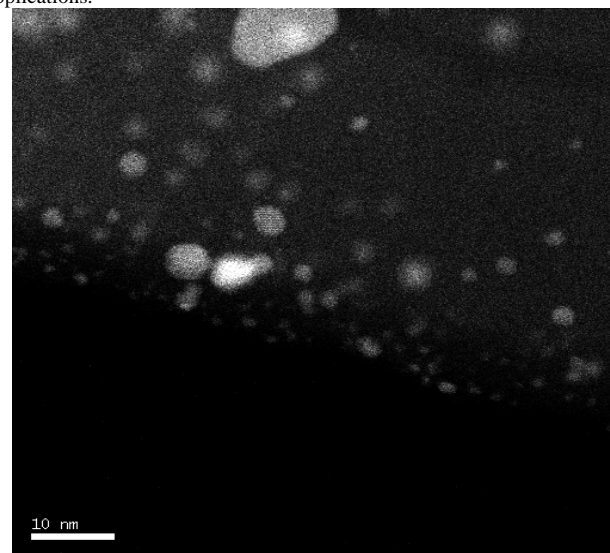


Figure 1. Z-contrast STEM image of gold nanoparticles supported on boron nitride. Many particles have diameters less than 3 nm.

References

- [1] M. Haruta, N. Yamada, H. Kobayashi, S. Iijima, *J. Catal.* 115 (1989) 301.
- [2] M.M. Schubert, S. Hackenberg, A.C. van Veen, M. Muhler, V. Plzak, R.J. Behm, *J. Catal.* 197 (2001) 113.
- [3] M. Comotti, W.-C. Li, B. Spliethoff, F. Schüth, *J. Am. Chem. Soc.* 128 (2006) 917-924.
- [4] N. Weiher, E. Bus, L. Delannoy, C. Louis, D.E. Ramaker, J.T. Miller, J.A. van Bokhoven, *J. Catal.* 240 (2006) 100-107.
- [5] M. Haruta, *J. New Mater. Electrochem. Systems* 7 (2004) 163.

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