

Design and synthesis of highly active supported Au catalysts

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Introduction

Supported Au nanoparticles on oxides have been shown to be active catalysts for a number of reactions [1-3]. Their special catalytic properties have been attributed to effects such as particle thickness and shape, the Au oxidation state, and to support effects [4]. Recently, we have reported that Au completely wets an oxide surface to form (1x1) mono- and (1x3) bilayer structures on a $\text{TiO}_x/\text{Mo}(112)$ [5]. The (1x3) Au bilayer has been shown to be highly active for catalytic CO oxidation. These results demonstrate that a continuous Au film on a reduced oxide surface can be far more active on a Au atom basis than Au nanoparticles on a support. These studies have been extended to address the detailed structures of ordered Au films on $\text{TiO}_x/\text{Mo}(112)$, the wetting of Au on other oxide supports, and the catalytic properties of bilayer Au films compared to bilayer Au particles.

Experiments

The experiments were carried out in an ultrahigh vacuum (UHV) system with a base pressure of 2×10^{-10} Torr equipped with a high-resolution electron energy loss spectroscopy (HREELS), scanning tunneling microscopy (STM), Auger electron spectroscopy (AES), low-energy electron diffraction (LEED) and temperature programmed desorption (TPD). A contiguous elevated pressure reaction was also available for kinetic studies at realistic conditions. The Mo(112) sample was cleaned by repeated cycles of oxidation at 1200 K followed by a flash to ~ 2100 K; cleanliness was verified with AES and LEED. The Mo(112)-(8x2)- TiO_x support was prepared by depositing ~ 1 ML of Ti onto a Mo(112)-c(2x2)-[SiO₄] surface which was then oxidized at 800 K and annealed at 1200, 1400 K in O₂ [5]. The $\text{TiO}_2(110)$ sample was cleaned by repeated cycles of Ar⁺-sputtering/annealing. Gold were evaporated from a gold wire wrapped around a Ta filament that was heated resistively. A CO:O₂ (2:1) mixture was used for the kinetic studies; the CO₂ product was analyzed by mass spectrometry and cross-checked by trapping and measuring the product partial pressure.

Results and discussion

Atomically resolved STM images of Figure 1 are consistent with the structural models of the ordered (1x1) monolayers and (1x3) bilayers proposed previously, as indicated in the insert of Figure 1. The structure, stability, catalytic properties, and sintering mechanism of Au nanoparticles on clean and chemical modified $\text{TiO}_2(110)$ are compared with the corresponding properties of the (1x3) Au bilayer film. Ongoing parallel investigations of high-surface-area TiO_2 supported Au catalysts addressing the wetting of Au to oxide surfaces to enhance of the density of catalytically active sites will also be discussed.

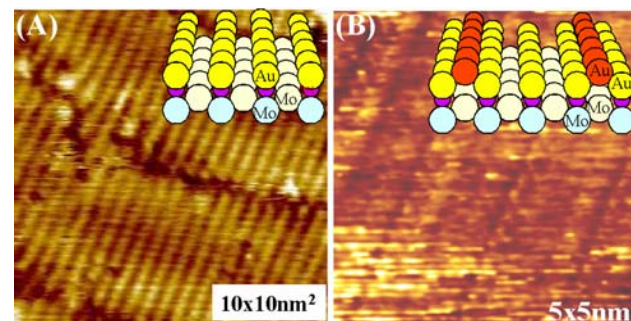


Figure 1. Atomic resolved STM images of (A) (1x1) mono- and (B) (1x3) bilayer on $\text{TiO}_x/\text{Mo}(112)$. The inserts are models of these structures.

Significance

A continuous Au bilayer film is exceptionally active for CO catalytic oxidation. The catalytic activity of this bilayer film is significantly higher on an atom basis than that for oxide-supported Au nanoparticles because the density of catalytically active sites for the Au bilayer structure is several orders higher than that for supported Au nanoparticles.

References

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