OAK RIDGE NATIONAL LABORATORY
U. S. DEPARTMENT OF ENERGY

FWP KCC053 Fundamentals of Heterogeneous Catalysis on Surfaces and Nanostructures

Modification of Au Catalysts with Metal Oxide for CO Oxidation Reactions

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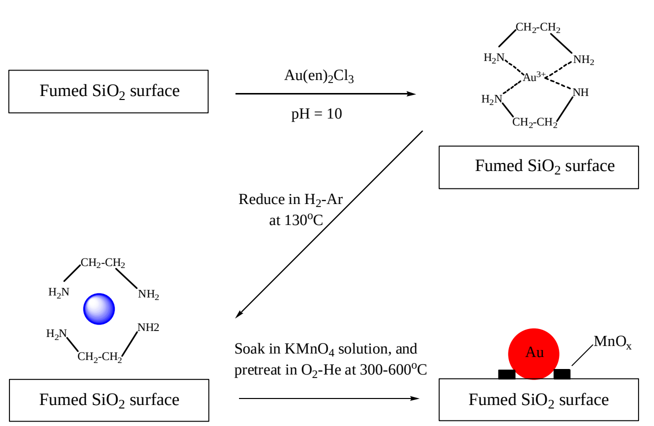
Synopsis: Supported gold catalysts with unique gold-metal oxide interfaces were prepared through oxidation treatment of gold nanoparticles protected by organic ligands with $KMnO_4$. The method can lead to a significant enhancement in activity and stability for CO oxidation.

Background

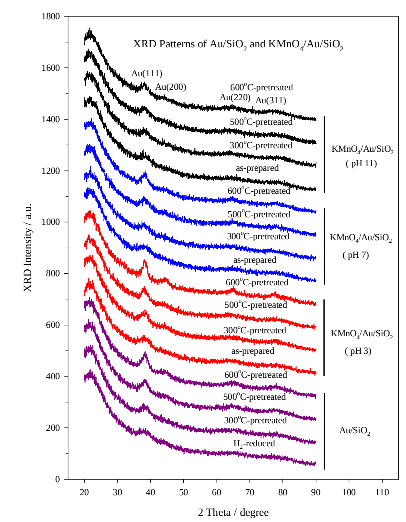
- **Critical factors affecting Au catalysts in CO oxidation**
  - **Size of Au nanoparticles (sintering)**
  - **Support Property (interface property between Au nanoparticles and support)**
  - **Available active sites (organic surfactant coverage)**
- **Calcination under high-temperature conditions is widely used so far to remove these organic ligands and further to activate the nanoparticles for catalysis applications**
- **The key drawback associated with calcination strategy is the facile sintering of nanoparticle catalysts under high-temperature conditions**

Synthesis

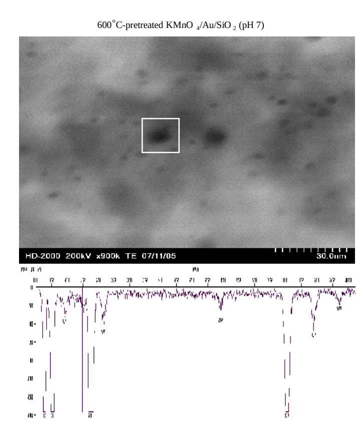


Yin, H.; Ma, Z.; Overbury, S. H.; Dai, S. *J. Phys. Chem. C.*, **2008**, in press.

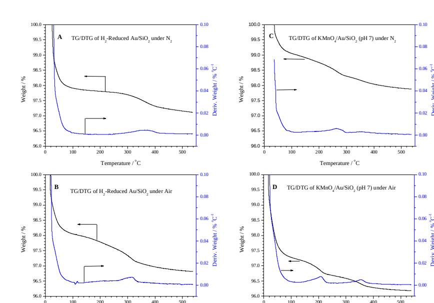
Results



- **XRD patterns of Au/SiO_2, $KMnO_4/Au/SiO_2$ (pH 3), $KMnO_4/Au/SiO_2$ (pH 7), and $KMnO_4/Au/SiO_2$ (pH 11)**



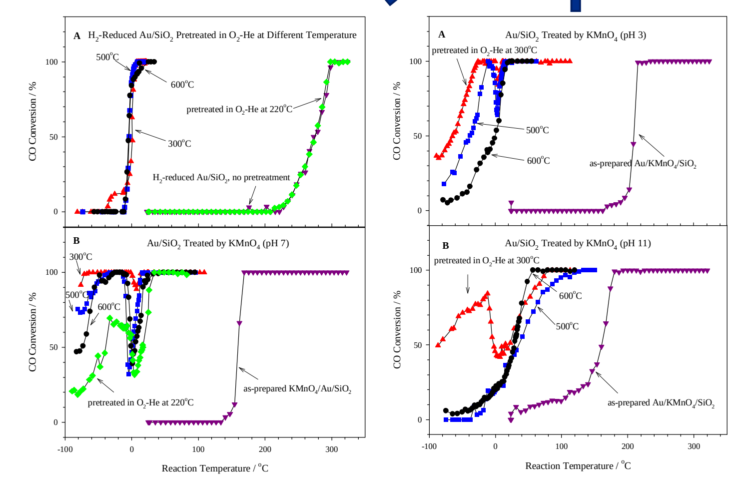
- **TEM image of 600°C-pretreated $KMnO_4/Au/SiO_2$ (pH 7) and EDX analysis indicate the presence of MnO_x near Au active sites via conformal redox precipitation, while no Mn observed outside of Au surroundings**



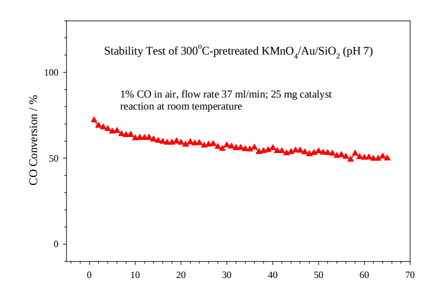
- **TG/DTG analysis of H_2-reduced Au/SiO_2 and $KMnO_4/Au/SiO_2$ (pH 7) under N_2 or air**

	As reduced	300°C	500°C	600°C
As prepared	2.1	3.3	4.5	4.8
pH = 3	2.5	3.0	4.4	7.0
pH = 7	2.5	3.0	3.6	4.4
pH = 11	2.8	2.5	3.4	4.1

- **Introducing metal oxide stabilizes Au nanoparticles against sintering. The presence of MnO_x promoter can activate oxygen at very low temperatures**



- **CO conversions on H_2-reduced Au/SiO_2 and $KMnO_4/Au/SiO_2$ (pH 7)**
- **CO conversions on $KMnO_4/Au/SiO_2$ (pH 3) and $KMnO_4/Au/SiO_2$ (pH 11)**
- **Au particle sizes (nm) of $KMnO_4/Au/SiO_2$**



- **Stability test of 300°C-pretreated $KMnO_4/Au/SiO_2$ (pH 7)**

Conclusion

- **The treatment of $Au(en)_2Cl_3$-derived Au/SiO_2 with $KMnO_4$ promotes the catalysis activity of Au/SiO_2 for low-temperature (-10°C) CO oxidation**
- **A new methodology to remove organics on catalytic nanoparticles with minimum sintering is developed**