Catalysis Club of Philadelphia

Wednesday October 22, 2008
Holiday Inn Select Hotel
Naamans Road and I-95, Claymont, DE

Joint meeting with the Wilmington area section of AIChE

Supported Gold Catalysts for CO Oxidation: Synthesis, Activation, Deactivation, and Identification of Catalytically Active Species

Prof. Bruce C. Gates
University of California, Davis

Social Hour: **5:30 PM**
Dinner: **6:30 PM**
Meeting: **7:30 PM**

Members: $30.00
Walk Ins & Non-members: $35.00
Student & Retired Members: $15.00

Menu - *Dinner Buffet*
Chicken Francaise *- Egg Battered in a Light Lemon Buerre Blanc*
Broiled Flounder *- Topped with Rock Shrimp in a light Lobster Sauce*
Penne Pommodoro

Meeting registration - Please notify your company representative or Alan Lee Stottlemyer (alan@udel.edu, phone: 302.831.6915, fax: 302.831.1048) by ***Wednesday, October 15***.
No meal reservation is required for this meeting, since the dinner is buffet-style.

Company Representatives – We would like to encourage you to make meal/meeting reservations to your company representative.

Membership - Dues for the 2008-09 season will be $10.00 ($5.00 for the local chapter and $5.00 for the national club). Dues for students and post-docs will be $6.00 ($5.00 for local club and $1.00 for national club). Please send your payment to Steve Harris, Lyondell Chemical Co., 3801 West Chester Pike, Newtown Square, PA 19073-2387.

SPONSORS

PLATINUM
Johnson Matthey

GOLD
The Catalyst Group
PQ/Zeolyst
Colonial Metals
Arkema

SILVER
Lummus Technologies Inc
Air Products
Millennium

CLUB OFFICERS

CHAIR
Hai-Ying Chen
Johnson Matthey

CHAIR-ELECT
Michael A. Smith
Villanova University

PAST CHAIR
Edrick Morales
LyondellBasell Industries

TREASURER
Stephen H. Harris
LyondellBasell Industries

SECRETARY
Bjorn Moden
Zeolyst International

PROGRAM CHAIR
Qi Sun
LyondellBasell Industries

ARRANGEMENTS CHAIR
Alan Lee Stottlemyer
University of Delaware

DIRECTORS
Haiming Liu
Arkema Inc.

Joseph Fedeyko
Johnson Matthey

Elizabeth Ross-Medgaarden
LyondellBasell Industries

NATIONAL REPRESENTATIVE
Anne Gaffney
Lummus Technology, a CB&I Company

Catalysis Club of Philadelphia

Wednesday October 22, 2008

Holiday Inn Select Hotel
Naamans Road and I-95, Claymont, DE

Joint meeting with the Wilmington area section of AIChE

Supported Gold Catalysts for CO Oxidation: Synthesis, Activation, Deactivation, and Identification of Catalytically Active Species

Prof. Bruce C. Gates
Dept. of Chemical Engineering & Materials Science
University of California, Davis

Abstract

Notwithstanding the inertness of bulk gold, the chemistry of gold compounds is rich and varied; in most of its known complexes, gold is cationic, Au(I) or Au(III). Some gold complexes are highly reactive, and many are catalysts in solution. Numerous reactions of potential industrial value, including hydrogenations, oxidations, and coupling reactions, are now known to be catalyzed by highly dispersed supported gold, but identifications of the catalytically active species remain controversial. We have addressed the subject by preparing well-defined gold complexes on supports as starting materials for our catalysts.

Site-isolated, mononuclear (single-metal-atom) dimethyl gold complexes on oxides including CeO_2 and MgO were synthesized by chemisorption of $Au(CH_3)_2(acac)$ (acac is acetylacetonate, $C_5H_7O_2$). Characterization by EXAFS, XANES, and IR spectroscopies in conjunction with temperature-programmed methods and catalytic reaction tests provided understanding of the nature of the catalytically active species, how they form, and how they are deactivated.

The CeO_2-supported sample containing mononuclear cationic gold was slightly active for CO oxidation at room temperature in a flow reactor, and during operation it underwent rapid activation. Similarly, MgO-supported gold was rapidly activated, and the onset of activity occurred just as methyl groups were removed from the gold while simultaneously the gold was reduced and clusters formed. The incipiently formed clusters, consisting of only a few Au atoms each, on average, are markedly more active than the gold complexes, and stable enough in excess O_2 to allow measurements of the CO oxidation kinetics in the case of the CeO_2-supported sample: $r = kP_{CO}^{0.19}P_{O2}^{0.18}P_{CO2}^{-0.44}$, where the rate r (TOF) is in molecules of CO converted (Au atom × s)$^{-1}$, the value of k at 303 K is 3.1×10^{-2} molecules (Au atom × s × mbar$^{0.07}$), and the apparent activation energy is 54 ± 8 kJ mol^{-1} (vs. 138 ± 2 kJ mol^{-1} for the mononuclear gold catalyst).

During operation, the MgO-supported catalyst underwent deactivation, accumulating species such as carbonate and formate on its surface, as indicated by IR spectra. These species accumulated only initially on the MgO support, but they accumulated continuously on the gold, blocking active sites; they were readily removed in inert gas. During CO oxidation, some of the gold on MgO (and on La_2O_3) was oxidized, as shown by an $Au^{\delta+}$–CO IR band at 2151 cm^{-1}. This observation, consistent with the presence of anionic species like carbonate on the gold, points to the involvement of cationic gold in the catalysis, even when the active species are gold clusters. The cationic gold may include species at the gold–support interface.

Speaker’s Bio

Professor Bruce C. Gates received his B.S. in chemical engineering from University of California, Berkeley in 1961 and his Ph.D. in chemical engineering from University of Washington, Seattle in 1966. After a post-doctoral fellowship at Ludwig Maximilian’s University of Munich, he joined Chevron Research Co in 1967. In 1969, he joined the Department of Chemical Engineering at University of Delaware, where he was director for the Center for Catalytic Science and Technology from 1981 to 1988. Since 1992, he has been a professor in the Department of Chemical Engineering and Materials Science at University of California, Davis.

Prof. Gates has received numerous awards, including four awards from AIChE (Alpha Chi Sigma Award for Chemical Engineering Research in 1989, William H. Walker Award for Excellence in Contributions to Chemical Engineering Literature in 1995, R. H. Wilhelm Award in Chemical Reaction Engineering in 2002, Northern California Section Award for Chemical Engineering Excellence for Professional Development, 2002/2003) and the Catalysis Club of Philadelphia Award in 1986. In 2007, he was elected to the National Academy of Engineering.