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Catalysis Club of Philadelphia

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Thursday, January 25, 2007

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

"Syngas and Chemicals by Catalytic Autothermal Reforming of Renewable Fuels"

Prof. L. D. Schmidt

Department of chemical Engineering and Materials Science
University of Minnesota, Minneapolis MN
schmidt@cems.umn.edu

&

"New Insights into the Solid Acidity of Supported WO₃/ZrO₂ Catalysts"

Elizabeth I. Ross, Lehigh University

(Student Presentation, 15 min)

Social Hour	5:30 p.m.
Dinner	6:30 p.m.
Meeting	7:30 p.m.

Menu:

Chicken Forrestiere - Wild Mushroom Cognac Sauce
London Broil - Served with Sauce Roben
Rigatoni a la Carmelo

Members	\$30.00
Walk Ins & Non-members	\$35.00
Students & Retired Members	\$15.00

For meal reservations, please notify your **company representative** or **Carl Menning** (phone: 302-893-9398, fax: 302-831-1048, e-mail: menning23@gmail.com) by **Thursday, January 18.**

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

Membership – dues for the 2006-2007 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.

Next Meeting will be held on Thursday, February 22, 2007. The speaker will be Prof. Galen Stucky from University of California at Santa Barbara.

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Thursday, January 25, 2007

“Syngas and Chemicals by Catalytic Autothermal Reforming of Renewable Fuels”

Prof. L. D. Schmidt

Department of chemical Engineering and Materials Science
University of Minnesota
Minneapolis MN
schmidt@cems.umn.edu

Abstract

We compare the reforming of different types of biofuels by autothermal reforming at millisecond contact times to produce synthesis gas, hydrogen, and chemicals. Fuels examined are alcohols, esters, carbohydrates, biodiesel, and vegetable oils.

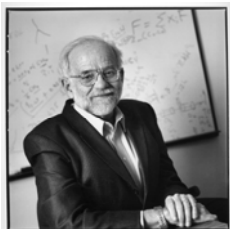
Biofuels generally have higher conversions than fossil fuels because the hydroxyl and ester linkages in these fuels produce higher sticking coefficients than saturated alkanes. Consequently, conversions of all biofuels in these processes are nearly 100%. Highly oxygenated feedstocks tend to produce mostly syngas with little olefins or oxygenated products because surface reactions dominate, and these larger products are formed predominantly by homogeneous reaction processes after all oxygen is consumed.

Esters have been examined to determine their reaction channels. We compared ethyl propionate and ethyl lactate, esters that differ only by the addition of an OH group in the lactate. Both esters can be made to produce mostly synthesis gas at low C/O, but at higher C/O considerable ethylene is formed. More ethylene is formed from ethyl propionate than the lactate, suggesting that in the lactate bonding is through the OH group which leads to complete decomposition to synthesis gas.

The effects of catalyst materials, loading, and support geometries have been examined extensively for these systems. Rhodium is a stable catalyst for all of these reactions, but the variation of metal loading from 0.5 to 10% by weight in α -alumina foams has a very small effect on performance. Addition of a γ -alumina wash coat increases syngas and decreases larger products, but wash coat thickness has a small effect. Addition of Ce to Rh increases syngas formation, and this appears to result from increased WGS and reforming activity compared to Rh alone.

Production of syngas by reactive flash volatilization of nonvolatile liquids will also be described. We show that, by impinging cold liquid drops onto the hot catalyst surface, the process can be operated in steady state with no carbon formation for many hours. This occurs because, while pyrolysis of vegetable oils and carbohydrates at low temperatures produces carbon, above $\sim 600^\circ\text{C}$ the equilibrium shifts to produce syngas rather than solid carbon. Recent results examining other liquids and the presence of species such as K, P, and S in nonvolatile liquids.

Speaker's Biography



Lanny D. Schmidt is a Regents Professor in the Department of Chemical Engineering and Materials Science at the University of Minnesota.

Professor Schmidt's research focuses on various aspects of the chemistry and engineering of chemical reactions in situations with technological applications. Reaction systems of recent interest are catalytic oxidation processes to produce products such as hydrogen, syngas, olefins, oxygenates, and HCN by partial oxidation and NO_x removal and incineration by total oxidation. Applications include direct conversion of alkanes and renewable fuels into chemicals, the production of hydrogen, and fuel reforming for fuel cells.

Professor Schmidt has published over 320 papers in refereed journals. He has supervised approximately 75 Ph.D. theses and 15 M.S. theses at Minnesota. He is a member of the National Academy of Engineering.

Student Presentation (15 min)

“New Insights into the Solid Acidity of Supported WO_3/ZrO_2 Catalysts”

E.I. Ross^{*}, T. Kim^{*}, W.V. Knowles⁺, M.S. Wong⁺ and I.E. Wachs^{*}

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⁺Departments of Biological & Chemical Engineering and Chemistry
Rice University, Houston, TX, USA

Abstract

The acidic characteristics of supported WO_3/ZrO_2 catalysts have been known for some time now, but many fundamental questions still remain about the origin of its interesting solid acidity characteristics. A combined Raman, IR, UV-Vis and temperature programmed surface reaction (TPSR) *operando* spectroscopy study was undertaken in order to better understand the solid acidity of supported WO_3/ZrO_2 catalysts. Both well defined model supported WO_3/ZrO_2 catalysts and calcined $WO_3/ZrO(OH)_2$ catalysts were investigated. Monolayer surface coverage was determined to occur at 4.5 W/nm^2 and the same tungsten oxide species were found to be present in both catalytic systems at the same surface W density: isolated surface WO_4 species, polymeric surface WO_5 species, WO_3 nanoparticles (NPs) and bulk-like WO_3 particles. The WO_x domain size for both catalytic systems, reflected in the UV-Vis E_g value, was essentially identical with surface WO_x coverage on ZrO_2 . Methanol dehydration to CH_3OCH_3 was selected as the chemical probe reaction and found to be very sensitive to the different tungsten oxide structures. The methanol dehydration TOF was found to increase with increasing tungsten oxide domain size: isolated surface WO_4 species < polymeric WO_5 species < WO_3 NPs < bulk-like WO_3 . However, the number of catalytic active sites varied inversely: bulk-like WO_3 < WO_3 NPs < surface WO_x species. Consequently, the turnover rate for methanol dehydration exhibits a maximum with surface W density at $\sim 8\text{-}10 \text{ W/nm}^2$. The bridging W-O-Zr bond is the catalytic acid site and its acidic character was found to strongly depend on the ZrO_2 support domain size.

Speaker's Biography

Elizabeth I. Ross is a graduate student finishing a PhD in Chemical Engineering at Lehigh University under the advisement of Israel E. Wachs. She received her B.S. in 2002 from Lafayette College. Elizabeth's research focuses on metal oxide catalysis, primarily forming metal oxide support nanoligands and determining the effect on the catalytic activity of surface species and active sites. She is the recipient of a Distinguished Air Products Graduate Fellowship and her work has received awards from both the Philadelphia and New York Catalysis Societies.