The Catalysis Club of Philadelphia

(In Person) Thursday, January 18th, 2024

Brandywine Plaza Hotel
630 Naamans Road, Claymont, DE 19703

Designing solvent microenvironments in zeolite catalysts using atomistic simulations for sustainable fuels and chemicals

Speaker: Prof. Brandon C. Bukowski

Johns Hopkins University

Observation of Dual Catalytic Sites for Selective Oxidation of Ethanol over Zeolite-Supported Gold Catalysts

Student Speaker: Yiteng Zheng

Princeton University

Meeting Agenda:

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

Meeting Registration:

Members: $45.00
Non-Members: $55.00
Stud. & Retired Members: $35.00

Please register online for this In-person meeting by ***Thursday, January 11st*** at CCP website.

Meal Selection (Included):

Please make one selection for your dinner (included in registration) when you sign-up for the meeting from the following options:

1. Herb roasted chicken. Dessert: strawberry cheesecake.
2. Sliced roast sirloin. Dessert: strawberry cheesecake.
3. Eggplant parmesan (vegetarian). Dessert: strawberry cheesecake.
4. Cavatappi pasta (vegan). Dessert: fruit cup.

Salad comes with any option: Caesar Salad

Membership Registration:

Membership dues for CCP 2023-24 meeting season will be $25 ($5 for the local chapter and $20 for the national club). Dues for students, post-docs and retirees will be $10 ($5 for the local club and $5 for the national club). Please sign-up membership (Link) for more benefits on meeting registrations and net-working events!

Please contact our Treasurer Steve Hardwick (sjh.wilm.de@gmail.com) or Chair Angela Zheng (angela.zheng@matthey.com) if you need any assistance.

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Prof. Brandon C. Bukowski

Designing solvent microenvironments in zeolite catalysts using atomistic simulations for sustainable fuels and chemicals

Department of Chemical and Biomolecular Engineering, Johns Hopkins University

Abstract: There is a critical societal need to develop new selective heterogeneous catalysts that enable large-scale decarbonization of the transportation and chemical industries. Catalytic valorization processes that utilize biomass derived from agricultural waste are typically selectivity-limited which limits their commercial viability. One exemplar catalyst family includes crystalline nanoporous solids such as zeolites which are employed in a wide range of chemical processes due in part to their tunable shape selectivity and framework-substituted cations. Reaction kinetics at internal zeolite acid sites can be modified by changing pore size, pore architecture, polarity, or acid site identity. These active site microenvironments impart shape-selectivity that preferentially stabilizes transition states and unique water matrices that in turn influence the stability of reactive intermediates. The large design space of zeolite catalysts including their pore architecture, polarity, and active site identity complicates comprehensive kinetic studies to predict new selective catalysts. Computational modeling of kinetic mechanisms in heterogeneous catalysis has emerged over the last few decades as a tool to predict both the structure and function of active sites to accelerate catalyst design. Here we will discuss how synergistic combinations of density functional theory (DFT), molecular dynamics, and machine learning techniques can provide design principles to design new selective zeolite catalysts for fuels and fine chemicals.

Speaker Bio: Brandon Bukowski is an Assistant Professor in the department of Chemical and Biomolecular Engineering and associate researcher in the Ralph O'Connor Sustainable Energy Institute at Johns Hopkins University. He holds BS and PhD degrees in chemical engineering from Worcester Polytechnic Institute and Purdue University, respectively. At Purdue he was advised by Jeffrey Greeley, received the Faculty Lectureship Award in 2019, and was supported in part by a Dick Reitz fellowship from the Center for the Innovative and Strategic Transformation of Alkane Resources NSF ERC. He performed post-doctoral research at Northwestern University under the supervision of Randall Snurr. Bukowski started at Johns Hopkins University in July of 2021. He has received a Ralph E. Powe award from Oak Ridge Associated Universities and a Doctoral New Investigator grant from the ACS Petroleum Research Fund. He is a member of the ACS Catalysis division board as Twitter Chair.

Observation of Dual Catalytic Sites for Selective Oxidation of Ethanol over Zeolite-Supported Gold Catalysts

Yiteng Zheng

Advisor: Prof. Bruce E. Koel, Princeton University

Abstract: Selective oxidation of alcohols to aldehydes, acids and esters is required in the production of numerous products in the chemical industry. Some current commercial technologies use non-catalytic stoichiometric reactions with oxidants, such as chromate or permanganate, which are expensive and toxic. It is, therefore, highly desirable for green chemistry and improved efficiency to replace these current technologies with catalytic processes using gas-phase oxygen as the oxidant. In this work, Au/ZSM-5 catalysts were synthesized, characterized and tested for ethanol selective oxidation. Effects of the Au loading, zeolite acidity as well as proximity of Au and zeolite acid sites were studied with X-ray diffraction (XRD), transmission electron microscopy (TEM), diffuse reflectance ultraviolet–visible spectroscopy (UV-vis DRS), in situ diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) and density functional theory (DFT) calculations.

The ethanol reaction rate over 1 wt % Au/ZSM-5 (Si/Al=15) was 26 times higher than that over 1 wt % Au/SiO_2. 1 wt% Au/S-1 was found to be more active than Au/SiO_2 by a factor of 3. Furthermore, Au/ZSM-5 exhibited better stability than the SiO_2-supported catalyst. The ethanol reaction rate increased with increasing Al concentration in the zeolite. At the highest tested Al concentration (Si/Al = 15), the ethanol reaction rate over Au/ZSM-5 was 9 times higher than that over Au/S-1 at the same Au loading. Physical mixtures of 1 wt% Au/S-1 and blank ZSM-5 (Si/Al = 15) with different ratios were tested to evaluate the influence of proximity of Au and Al atoms. The physical mixtures had reaction rates similar to that of pure 1 wt% Au/S-1, and the rates were much lower than Au/ZSM-5. Therefore, Al zeolite sites do not serve as independent catalytic sites, but they enhance the catalytic activity of Au sites when Au and Al atoms are in proximity.

Speaker Bio: Yiteng Zheng holds a M.S. in Chemical Engineering from Stevens Institute of Technology and a Ph.D. in Chemical Engineering also from Stevens Institute of Technology under the guidance of Professor Simon G. Podkolzin. He is currently a postdoctoral research associate at Princeton University under the guidance of Professor Bruce E. Koel. Yiteng has several years of experience on combing density functional theory (DFT) calculations and in situ/operando characterizations to understand the structure-activity relationship for heterogeneous catalysts. Other than thermal catalysis, he is currently working on designing in situ characterization reactors (Raman and X-ray absorption spectroscopy) for plasma-assisted catalysis.