

## The Catalysis Club of Philadelphia

Webinar: 7 PM, Thursday, April 21<sup>st</sup>, 2022

Zoom link shared after registration

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**Speaker: Prof. Hongliang Xin**

*Virginia Tech*

**Student Speaker: Christopher Rzepa**

### Meeting Schedule:

6:30 PM EST: Welcome  
7:00: Talk by Christopher Rzepa  
7:25: Talk by Prof. Xin  
8:30: Meeting adjourns

### Meeting Fees:

Free to all who register

### Meeting Etiquette:

Please remember to mute your microphone and arrive early to solve any technical issues. Camera sharing prior to the talks is encouraged.

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Online Registration – Please register online by Wednesday, April 20<sup>th</sup> at <http://catalysisclubphilly.org/webinar-registration/> or Arrangements Chair, [hrenjing@seas.upenn.edu](mailto:hrenjing@seas.upenn.edu).

A Zoom meeting invite will be provided through the confirmation email. If you do not receive a confirmation email immediately after registration, please contact Renjing Huang, [hrenjing@seas.upenn.edu](mailto:hrenjing@seas.upenn.edu).

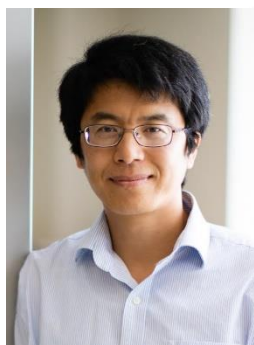
Membership – Dues for the 2021-22 season will be \$25 (\$5 for the local chapter and \$20 for the national club). Dues for students, post-docs and retirees will be \$10 (\$5 for the local club and \$5 for the national club).

## Advancing Catalytic Materials for Ammonia Redox Cycling by Machine Learning

Hongliang Xin, Virginia Tech

**Abstract** Ammonia ( $\text{NH}_3$ ), an essential molecule of the global nitrogen cycle, has been widely used in industrial domains, including medicine, food, agriculture, and the manufacture of household chemicals. Particularly attractive is its application as a working medium for energy storage and generation devices because of easy liquefaction and high volumetric energy density. Artificial nitrogen fixation in  $\text{NH}_3$ , also known as the Haber-Bosch process, is imperative to providing food for the booming world population. However, this century-old technology is quite energy intensive (accounting for 1-2% of global energy consumption) and results in substantial carbon dioxide ( $\text{CO}_2$ ) emissions. Electrochemically reducing nitrate ( $\text{NO}_3^-$ ) to  $\text{NH}_3$  as a renewable fuel and green fertilizer has the potential to enable a sustainable  $\text{NH}_3$  economy. On the oxidation side of the electrical-chemical energy interconversions, the electrocatalytic  $\text{NH}_3$  oxidation has been widely investigated as a crucial component in the context of direct  $\text{NH}_3$  fuel cells and wastewater treatment technologies. Pt-based electrocatalysts have shown promising activity for oxidizing  $\text{NH}_3$  to  $\text{N}_2$ . However, the low energy efficiency, the scarcity of precious metals, and surface poisoning issues are tremendous roadblocks for its widespread adoption.

In this talk, I am going to use the  $\text{NH}_3$  redox cycling as a specific case, i.e.,  $\text{NO}_3^-$  reduction to  $\text{NH}_3$  and electrooxidation of  $\text{NH}_3$  to  $\text{N}_2$ , and discuss the opportunities and challenges of machine learning (ML) techniques for accelerating catalytic materials discovery. Herein, we develop an explainable AI (XAI) framework that seamlessly integrates ML algorithms with catalysis theories for interpretable prediction of transition-metal catalysts. With adsorption energies of key intermediates, e.g.,  $^*\text{N}$  species, as reactivity descriptors, we demonstrate that the XAI approach outperforms the purely data-driven ML methods in prediction performance and interpretability, especially for intermetallics and ternary alloys with unseen structural and electronic features. Incorporation of scientific knowledge of physical interactions into learning from data sheds further light on the nature of chemical bonding and opens up new avenues for ML discovery of novel motifs with desired electronic structure and catalytic properties.



**Bio** Hongliang Xin is an Associate Professor of Chemical Engineering at Virginia Polytechnic Institute and State University. He received his Ph.D. in Chemical Engineering from the University of Michigan in 2011. Dr. Xin joined the ChE faculty in August 2014 after postdoc at Stanford/SLAC. At Virginia Tech, his research focus is the development of a machine learning framework for tackling problems in fundamental surface science and catalysis. He received the recognition from the Journal of Materials Chemistry A as one of the 2017 Emerging Investigators. He received the Dean's award for Outstanding New Assistant Professor in 2018 and Engineering Faculty Fellow in 2019. He is one of the 2019 Class Influential Researchers from ACS Industrial & Engineering Chemistry Research. He is the recipient of the prestigious NSF CAREER Award (2019). He is currently serving as the President of the Southeastern Catalysis Society.

Inferring structure-entropy correlations of zeolite-adsorbate interactions using Monte-Carlo simulations & machine learning

Christopher Rzepa, Lehigh University

Advancements in ab-initio simulations have made possible the quantification of a reaction system's energy landscape, and subsequently the development of microkinetic models. Of the contributions to the free energy, entropic losses within zeolite catalysis have shown to be crucial toward rate inhibition and therefore product selectivity. However, calculating entropic losses induced by pore topology can be challenging or computationally expensive. Traditionally, the entropy has been rationalized in terms of confinement, or how well the adsorbate fits within the framework's pore. From this viewpoint, the adsorption entropy has been shown to follow straightforward correlations with considerable predictive capabilities using physical descriptors.

Experimental adsorbate entropies on two-dimensional catalytic surfaces compiled by Campbell & Sellers have shown that the ratio of the adsorbed-phase entropy to the gas-phase entropy was approximately two-thirds across all molecules/surfaces. This suggests that the adsorption of a molecule from an unhindered gas phase onto a two-dimensional surface would eliminate a dimension of freedom, i.e. the adsorbate behaves as a 2D gas. This idea was expanded to multiple dimensions by Dauenhauer & Abdelrahman, who showed that the entropic loss upon adsorption within zeolites correlates linearly with the molecule's gas-phase translational and rotational entropies; and that the occupiable volume of a framework is a useful descriptor in predicting such losses. Recently, we have implemented Monte-Carlo simulations to quantify the adsorption entropies for one-hundred-eighty-five molecule-zeolite combinations. Our results show that these linear correlations persist for our larger and more chemically diverse dataset and are governed by descriptors of the zeolite's topology. The elegance of such linear correlations not only offer predictive capabilities, but suggest an underlying physical origin.

Herein we expand our dataset to over seven-thousand molecule-zeolite combinations and implement a data-driven approach to reveal the key descriptors which govern these entropic correlations. We have compiled over fifty molecule-zeolite features from cheminformatics databases, including but not limited to: molecular eccentricity, radius of gyration, largest/limiting pore diameter, and total framework pore volume. Our ultimate goal is to use these models to better understand the structure-property relationships in the adsorption entropies.

## **Bio**

Christopher Rzepa is a 5th year Graduate student at Lehigh university within the department of Chemical & Biomolecular Engineering. He is advised by Professor Srinivas Rangarajan and co-advised by Professor Jeetain Mittal. He earned his B.S. in Chemical Engineering (Minor: Math) at Penn State University-University Park. Chris uses computational techniques to study two categories of problems: (i) elucidating the mechanism of catalytic reactions, and

(ii) shape selectivity from the confinement effects of organic and inorganic materials. In particular, he uses a combination of density functional theory, thermodynamic Monte-Carlo, and microkinetic modeling to rationally design confinement in porous catalysts.

