

Purdue University

SCHOOL OF CHEMICAL ENGINEERING GRADUATE SEMINAR SERIES

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“Designing Reaction Pathways to Novel Chemicals and Materials Using Kinetic Modeling”

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3:30 - 4:30 p.m.

FRNY G140

ABSTRACT:

Reaction pathway analysis is a powerful tool to design routes to chemicals and materials that are novel and lead to materials with unique and tailored properties. We have developed methods for the assembly of kinetic models of substantive detail to be built that enable the atomic scale to be linked with the process scale. We have applied our methodology to a wide range of different problems, including production of silicon nanoparticles, biochemical transformations, polymerization and depolymerization, and tropospheric ozone formation. While the chemistries we have studied are seemingly very disparate, applying a common methodology to study them reveals that there are many features of complex reaction networks that are ubiquitous. This presentation will focus on two of the systems we have examined: biochemical production of fuels and chemicals and design of gradient copolymers. It will be shown how reaction pathway analysis can be used to discover novel routes to small molecules and also guide the transformation of small molecules to macromolecules with tailored backbone sequences.

The first portion of the talk will focus on designing novel pathways for the sustainable microbial production of high-value organic compounds as an attractive alternative to organic syntheses that utilize petrochemical feedstocks. For example, the high cost of and the numerous applications for 3-hydroxypropanoate (3HP) make it a valuable target for biosynthesis. We applied the Biochemical Network Integrated Computational Explorer (BNICE) framework for the automated construction and evaluation of metabolic pathways to explore novel biosynthetic routes for the production of 3HP from pyruvate. Among the pathways to 3HP generated by the BNICE framework were all of the known pathways for the production of 3HP and numerous promising novel pathways. The pathways generated for the biosynthesis of 3HP were ranked based on four criteria: pathway length, thermodynamic feasibility, maximum achievable yield to 3HP from glucose during anaerobic growth, and maximum achievable intracellular activity at which 3HP can be produced. Thermodynamic feasibility was assessed using a group contribution method in combination with Thermodynamics-based Metabolic Flux Analysis (TMFA). TMFA was also utilized in all yield and maximum achievable activity calculations. Four pathways emerged from this ranking as the most promising pathways for the biosynthesis of 3HP, and three of these pathways, including the two shortest pathways discovered, were novel. We also discovered novel routes for the biosynthesis of 28 commercially available compounds that are currently produced exclusively through organic synthesis.

The second part of the presentation will focus on the synthesis of gradient copolymers. A gradient copolymer has a gradient in the repeat units comprising the backbone of the polymer arranged from predominantly

monomer A to predominantly monomer B along the copolymer chain. Because of the gradual change of the composition along the copolymer chains, gradient copolymers exhibit distinct physical properties compared to those of random or block copolymers of the same composition. Furthermore, the different sequence distribution along gradient copolymer chains may lead to quite different physical properties. Kinetic Monte Carlo (KMC) models, which track molecules instead of concentration, were developed in order to track the explicit sequence distribution for each copolymer chain. Nitroxide-mediated controlled radical polymerization (NM-CRP) was used in synthesizing S/AS and MMA/S gradient copolymers because of its' pseudo-living property. The effects of different synthesis factors on the formation of the compositional gradient along copolymer chains will be described, and the ability to tailor the monomer-by-monomer sequence will be demonstrated.

BIO:

Linda Broadbelt is Sarah Rebecca Roland Professor in and Chair of the Department of Chemical and Biological Engineering University at Northwestern University. She received her B.S. in chemical engineering from The Ohio State University and graduated *summa cum laude*. She completed her Ph.D. in chemical engineering at the University of Delaware in 1994 where she was a Du Pont Teaching Fellow in Engineering, a National Science Foundation Graduate Fellow, and a DuPont PhD in Engineering Fellow. At Northwestern, she was appointed the Donald and June Brewer Junior Professor from 1994-1996. She recently completed the short course Business for Scientists and Engineers through the Kellogg Graduate School of Management.

Her research and teaching interests are in the areas of multiscale modeling, complex kinetics modeling, environmental catalysis, novel biochemical pathways, and polymerization/depolymerization kinetics. She was Associate Editor for Energy and Fuels (2002-2010). She currently serves on the Executive Board of the National Program Committee of AIChE and recently completed a five-year term as the chair of programming for the Division of Catalysis and Reaction Engineering of AIChE. She was also appointed to the Scientific Organizing Committee for the 21st and 19th International Symposium on Chemical Reaction Engineering and served on the Science Advisory Committee of the Gulf Coast Hazardous Substance Research Center from 1998-2005. She is currently on the editorial boards of *Industrial Engineering & Chemistry Research*, *International Journal of Chemical Kinetics* and *Chemical Engineering Journal*.

Her honors include selection as the AIChE Women's Initiative Committee Mentorship Excellence Award winner, a Fellow of the American Association for the Advancement of Science, a Fulbright Distinguished Scholar Award, a CAREER Award from the National Science Foundation, a McCormick Excellence Award, appointment to the Defense Science Study Group of the Institute for Defense Analyses, and selection as the Ernest W. Thiele Lecturer at the University of Notre Dame and the Allan P. Colburn Lecturer at the University of Delaware.