WINTER RESEARCH REVIEW

4TH YEAR TALKS
WEDNESDAY, JANUARY 22, 2020
Welcome to our Annual Winter Research Review.
We are pleased that you can join us. The focus of today’s program—research presentations by our fourth-year graduate students—provide one of the best opportunities to learn about the research of our senior graduate students and their faculty advisors. Throughout the day you can also visit research posters presented by our third-year students.

Our graduate program is one of the central foundations of our principal missions of scholarship and education. We hope that you will enjoy this opportunity to learn more about our department and its activities, as well as to meet the students and faculty.

Eric M. Furst
Professor and Department Chair
Department of Chemical and Biomolecular Engineering

Samantha Cassel
President of Colburn Club
The Graduate Student Organization

Colburn Club is the graduate student organization in the Chemical and Biomolecular Engineering Department, which is comprised of representatives from each year as well as a number of members filling specialized roles. The primary functions of the club are to organize research reviews and social events for the department, in addition to serving as one line of contact between the students and the faculty. We hope you enjoy this event and can join us again in the future.

The Colburn Club

www/che.udel.edu/cc
Alphabetical List of Talks

Schedule of Talks

Talk Abstracts

Jacob Anibal
Abhay Athaley
Atharv Bhosekar
Elvis Ebikade
Nathaniel Hamaker
Emily Jeng
Priyanka Ketkar
Muhuan Li
Arnav Malkani

Douglas Nmagu
Natalia Rodriguez Quiroz
Y. Summer Tein
Yifan Wang
Michiel Wessels
Gerhard Wittreich
Ou Yang
Daniel Yur

Poster Presenters List
Jacob Anibal
“The Importance of Ketyl Radical Stability in the Electroreductive Coupling of Biomass Derived Carbonyl Species”
Advisor: Bingjun Xu
Committee Members: Yushan Yan and Feng Jiao

Abhay Athaley*
“Integrated Design, Analysis and Optimization of Chemical Production from Biomass Feedstocks”
Advisor: Marianthi G. Ierapetritou
Committee Members: Shishir Chundawat, George Tsilomelekis, and Basudeb Saha

Atharv Bhosekar*
“Machine learning-based optimization frameworks for enterprise-wide optimization”
Advisor: Marianthi G. Ierapetritou
Committee Members: Ioannis Androulakis, Rohit Ramachandran, and David Coit

Elvis Ebikade
“Reductive Depolymerization of Herbaceous Biomass”
Advisor: Dionisios G. Vlachos
Committee Members: Raul F. Lobo, Bingjun Xu, and Yushan Yan

Nathaniel Hamaker
“More Efficient Site-specific Integration in CHO Cells Enabled by A CRISPR-mediated Genome Editing Reporter System”
Advisor: Kelvin H. Lee
Committee Members: Wilfred Chen and Maciek R. Antoniewicz

Emily Jeng
“Achieving High Purity CO in the Electrochemical Reduction of CO₂ to CO”
Advisor: Feng Jiao
Committee Members: Bingjun Xu and Yushan Yan

Priyanka Ketkar
“Investigation of Polymer Assembly and Lithium Salt Solvation in Tapered Block Polymer Electrolytes for Lithium-ion Batteries”
Advisor: Thomas H. Epps, III
Committee Members: Norman J. Wagner, Arhti Jayaraman, and Bingjun Xu

Muyuan Li
“Transition Metal Ion Exchanged ZK-5 Zeolite for Ethylene Separation”
Advisor: Raul F. Lobo and Craig Brown
Committee Members: Douglas J. Buttrey and Bingjun Xu

Arnav Malkani
“Impact of Forced Convection and Electrolyte Cations on Spectroscopic Observations of the Electrochemical CO Reduction Reaction”
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“Characterization and Biologic Comparability of High Producing VRC01 Reference CHO Cell Line”
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Natalia Rodriguez Quiroz
“Homogeneous Metal Salt Solution in Biomass Upgrading”
Advisor: Dionisios G. Vlachos
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Y. Summer Tein
“rheo-MAGIK Instrument Development for Investigating 2D Soft Materials via Interfacial Rheology and Structural Analysis”
Advisor: Norman J. Wagner
Committee Members: Eric M. Furst, Abraham M. Lenhoff, and Jan Vermant

Yifan Wang
“Statistical-learning-assisted First-principles Modeling of Subnanometer Catalysts Sintering”
Advisor: Dionisios G. Vlachos
Committee Members: Antony N. Beris, Bingjun Xu, and Marianthi G. Ierapetritou

Michiel Wessels
“Effect of Polymer Architecture on the Self-assembly in Amphiphilic Polymers Solutions: A Computational Study”
Advisor: Arthi Jayaraman
Committee Members: Norman J. Wagner, Eric M. Furst, and Darrin J. Pochan

Gerhard Wittreich
“Impact of Uncertainty Quantification and Density Functional Theory Error Propagation on Surface Catalyzed Kinetic Models”
Advisor: Dionisios G. Vlachos
Committee Members: Antony N. Beris and Raul F. Lobo

Ou Yang
“Flowsheet and Mathematical Modeling in Batch and Continuous Monoclonal Antibody Production”
Advisor: Marianthi G. Ierapetritou
Committee Members: Shishir Chundawat, Rohit Ramachandran, and Maen Qadan

Daniel Yur
“Targeted Delivery of Therapeutic Proteins to Inflammatory Breast Cancer Cells Using Modular Hepatitis B Virus-like Particles”
Advisor: Wilfred Chen and Millicent O. Sullivan
Committee Members: Eleftherios T. Papoutsakis and April M. Kloxin

*Non-UD Student.
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<td>8:30 – 9:00 AM</td>
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“Machine learning-based optimization frameworks for enterprise-wide optimization”
Advisor: Marianne G. Ierapetritou / Committee Members: Ioannis Androulakis, Rohit Ramachandran, and David Coit

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2:30 – 3:00 PM  SNACK BREAK

3:10 – 4:15 pm  INDUSTRY MIXER

4:15 PM  END

*Non-UD Student.
The Importance of Ketyl Radical Stability in the Electroreductive Coupling of Biomass Derived Carbonyl Species

Jacob Anibal
Advisor: Bingjun Xu
Committee Members: Yushan Yan and Feng Jiao

Biomass derived carbon-oxygenate species represent an emerging class of compounds with potential as a carbon feedstock for the renewable production of fuels and chemicals. Typically derived from lignocellulosic materials, these compounds offer a range of chemical functionalities, but their viability remains limited due to high oxygen content and low molecular weight. Electroreductive coupling offers a promising technology for upgrading these species, using an applied electrochemical potential to simultaneously reduce the oxygenates while growing molecular weight through new C-C bond formation. Applicable to many organic compounds, electroreductive coupling shows particular promise for biomass derived carbonyl species, such as derivatives of furfural and benzaldehyde, due to their wide availability and relatively facile coupling. Despite this promise, a lack of mechanistic understanding currently hinders development of electroreductive coupling catalysts. In particular, variation in carbonyl coupling ability between catalysts remains poorly understood, with some catalysts (Cu, Pb, Ti, Sn, and C) able to effectively couple while others (Pt group, Ni, Ag) cannot. To address this gap in understanding, we have investigated the electroreductive coupling of carbonyls on Au, Cu, Pd, and Pt using benzaldehyde reduction as a model reaction. Reactivity tests show a unique coupling ability for Cu among the catalysts tested. Subsequent \textit{operando} infrared spectroscopy experiments suggest this unique coupling results from an ability of Cu to better stabilize a key reaction intermediate, i.e. the benzaldehyde ketyl radical. Spectroscopic features corresponding to the ketyl radical are identified on both the Au and Cu surfaces, with a higher radical concentration likely allowing the coupling on Cu. Pt and Pd do not display these radical features, and instead show CO formation due to decarbonylation, suggesting instability of benzaldehyde adsorbates on Pt and Pd. Combined, the reactivity and spectroscopy suggest ketyl radical stability as a key descriptor in benzaldehyde coupling ability.
INTEGRATED DESIGN, ANALYSIS AND OPTIMIZATION OF CHEMICAL PRODUCTION FROM BIOMASS FEEDSTOCKS

Abhay Athaley
Advisor: Dr. Marianthi Ierapetritou
Committee Members: Dr. Shishir Chundawat, Dr. George Tsilomelekis and Dr. Basudeb Saha

Biomass processing has been identified as a promising source of energy which can replace the use of fossil fuels in the near future. It can be used to produce both high-volume and low-value fuels and high-value but low volume chemicals. The priority for the development of variate fine chemicals from biomass feedstock stepped-up over the changing time. Platform chemical derived from biomass provide notable opportunities to produce an array of derivatives to fulfill societal needs of organic chemicals and polymers. US department of energy have suggested some top-value platform chemicals derived from biomass which include levulinic acid, succinic acid, glycerol, furans etc. Bio-based products acceptance in the market depends on the competitiveness of economics and sustainability when compared to oil-based chemicals and products. The idea of bio-refinery has been proposed that uses different conversion technologies to produce multiple products. A bio-refinery, like a petroleum refinery, can benefit from the exploitation of different components in biomass to maximize the overall profit. Integration of different processes to produce chemicals and fuels by utilizing different components of biomass is difficult and is capital intensive.

Our work focuses on development and design of economical and sustainable routes for the production of various chemicals using different process system engineering tools. Promising chemicals, synthesized from the components of biomass, and having high demand in market are identified. Utilizing biomass as raw material, a new hydrolysis process was developed and integrated with the production of p-Xylene with furfural and lignin as the by-product. Furfural is a promising platform compound, which produces high quality fuel and valuable chemicals. Furfural has been selected as one of the top 30 biomass derived platform compounds by the U.S. Department of Energy on the basis of several indicators such as the raw material, estimated processing cost, technical complexity, and market potential. Promising chemicals such as Butadiene, Surfactants, Jet-fuels, Lubricants derived from furfural are investigated and integrated into the bio-refinery. The lignin formed is most commonly used to produce electricity. Lignin is linked by a robust C-C and C-O bonds and hence depolymerization can generate a complex mixture of compounds. A novel production path to produce pressure sensitive adhesive (PSA) from biomass depolymerization at high purity and yield is studied and incorporated. Alternative biomass such as food waste is also explored for producing these chemicals and fuels. Finally, multi-objective optimization is used to develop an optimal bio-refinery configuration considering economic and environmental parameters with regards to supply, demand and process uncertainties.
Machine learning-based optimization frameworks for enterprise-wide optimization

Atharv Bhosekar
Advisor: Dr. Marianthi Ierapetritou
Committee Members: Dr. Ioannis Androulakis, Dr. Rohit Ramachandran, Dr. David Coit

Optimal decision-making in the process industry is fundamental in order to improve performance and guarantee efficient operation in highly competitive and global markets. Process operations decision-making problems range across different scales and include control, operational, tactical and strategic decisions. Traditionally, these decisions are considered in a hierarchical fashion and addressed individually by different areas within an organization. However, remarkable benefits can be achieved through the proper integration of the decision-making process. The academic community has made significant progress towards this goal. Nevertheless, the available methodologies and tools do not fully meet industrial needs, and more practical and efficient methods to coordinate the different levels of decisions are needed. Effectively utilizing the available process data for decision-making presents a huge opportunity in this respect. With their ability to utilize process data coupled with recent developments in machine learning (ML) software, ML-based frameworks are continually finding new applications for various problems in process systems engineering.

In this work, efficient ML-based optimization frameworks for supply chain optimization, and modular process design optimization are proposed. For supply chain optimization, a multi-enterprise supply chain network design problem is considered where the objective is to decide optimum inventory allocation to minimize the overall cost of the supply chain. A simulation model of a supply chain network is considered where entities belong to multiple enterprises and have competing interests. The problem of deciding optimal inventory is found to display a discontinuous behavior which makes it challenging for the existing data-driven optimization algorithms. The proposed optimization framework uses data from the simulation uses it to train ML-based classification techniques to identify discontinuities. Gaussian process regression model is used to guide the search towards the global optimum. The application on three different supply chain networks demonstrate that the proposed framework provides more robust and cost-efficient solutions compared to three other optimization solvers under comparison. In the second part of this work, a novel framework is proposed to address the problem of modular process design optimization. The framework uses historical process data and ensures design feasibility using classification techniques. Application to the design optimization of an air separation unit leads to a set of optimal designs that minimize the cost and maximize the flexibility of operation under demand variability. Finally, the ongoing work on the integration of supply chain design and production planning and the role of machine learning-based optimization strategies is presented.
Reductive depolymerization of herbaceous biomass

Elvis Ebikade
Advisor: Dionisios Vlachos
Committee Members: Raul Lobo, Bingxun Xu, Yushan Yan

Cheap and abundant residues constitute a major waste stream from ethanol biorefineries and agricultural processing industries and are an alluring alternative feedstock for biorefineries. In this work we investigate the catalytic biorefining of lignin from five herbaceous biomass (HB): corn stover, miscanthus, switchgrass, sugarcane bagasse, and wheat straw. Through reductive catalytic fractionation (RCF), these feedstocks are depolymerized into phenolic monomers with high yields (~40 wt.%), next to well-preserved and de-ashed carbohydrate pulp suitable for further processing. In addition to the guaiacol and syringol monomers typically derived from virgin woody biomass, hydroxycinnamic acids and hydroxyphenols are also produced from the lignin carbohydrate complexes unique to herbaceous biomass. Contrary to trends observed for woody biomass showing a positive association between S-content and monomer yields, machine learning analysis indicates that the RCF yields from herbaceous biomass depend on the content of lignin crosslinker – ferulate, in the feedstocks. Recyclability experiments provided insights into catalyst deactivation during RCF of HB, indicating that catalyst deactivation occurs through sintering, leaching and surface fouling. A particularly unique and adaptable aspect of this work is the application of lignin oil derived directly from real waste biomass as an antimicrobial. Even at low concentrations, lignin oil from HB exhibits bacteriostatic effects against Staphylococcus aureus, highlighting a potential application of biomass derived lignin oil in functional food development and for food preservative purposes.
More efficient site-specific integration in CHO cells enabled by a CRISPR-mediated genome editing reporter system

Nathaniel K. Hamaker
Advisor: Kelvin H. Lee
Committee Members: Wilfred Chen, Maciek R. Antoniewicz

Chinese hamster ovary (CHO) cells continue to be the biomanufacturing platform of choice for the production of therapeutic glycoproteins, including monoclonal antibodies. The traditional cell line development workflow involves random integration of the transgene encoding the recombinant protein of interest. Random integration inherently leads to high levels of clonal heterogeneity and necessitates the tedious screening of multiple clones to isolate those with suitable performance characteristics. In contrast, site-specific integration (SSI) results in transgene knock-in events at user-defined genomic target sites, including prevalidated hot spots capable of supporting highly active and stable expression. SSI has therefore emerged as a powerful cell line engineering technique by enabling the accelerated development of production cell lines with more predictable performance and product quality. Despite these benefits, attempts to perform CRISPR-mediated SSI in CHO cells are typically met with remarkably low efficiencies, hindering efforts to isolate clones in which precise, rather than random, integration has occurred. Numerous methods have been proposed to remedy similar shortcomings observed in other mammalian cell types, yet none have been systematically evaluated in CHO. To overcome this obstacle, we developed the SSIGNAL (Site-specific Integration and Genome Alteration) reporter system to facilitate rapid screening of CRISPR-mediated genome editing strategies. The system uses fluorescent signals to indicate genotype states, enabling data acquisition via standard flow cytometry instrumentation without the need for single-cell cloning or deep sequencing. The system was applied in CHO cells to test several current approaches for targeted integration to determine the most effective double-strand break repair strategy with the goal of improving efficiency. Ultimately, the development of robust SSI protocols may be implemented to improve long-term stability of transgenes for better cell culture process control.
Achieving High Purity CO in the Electrochemical Reduction of CO\textsubscript{2} to CO

Emily Jeng
Advisor: Dr. Feng Jiao
Committee Members: Dr. Bingjun Xu, Dr. Yushan Yan

By utilizing electricity from renewable energy resources, the electrochemical reduction of carbon dioxide (CO\textsubscript{2}) can be an economical alternative for sustainable chemical production. One of the possible products of CO\textsubscript{2} electroreduction includes carbon monoxide (CO), which can then be further converted into multi-carbon products such as ethylene, ethanol and n-propanol, which are desirable due to their significant market potential in transportation and industrial sectors. In a flow cell electrolyzer for the conversion of CO\textsubscript{2} to carbon monoxide, one of the issues that needs to be addressed is the need for separation of the gas products after electrochemical reduction, where CO must be separated from unreacted CO\textsubscript{2}.

Studies have shown that conducting CO\textsubscript{2} electrochemical reduction in an alkaline environment results in high reaction rates while maintaining high selectivity towards CO. However, there is also the unwanted side reaction of carbon dioxide with hydroxide ions formed during the electroreduction process. We demonstrate that we can instead utilize this reaction to obtain a gas product stream with a high concentration of CO, investigating how changing the inlet flowrate and current will affect the reaction selectivity and product outlet concentrations for a CO\textsubscript{2} to CO electrolyzer.
Investigation of polymer assembly and lithium salt solvation in tapered block polymer electrolytes for lithium-ion batteries

Priyanka M. Ketkar
Advisor: Prof. Thomas H. Epps, III
Committee Members: Prof. Norman J. Wagner, Prof. Arthi Jayaraman, and Prof. Bingqing Wei

Tapered block polymers (TBPs) contain modified monomer segment distributions (e.g., gradient or random copolymer regions) at the chemical junction between two homogeneous blocks. Nanostructured polystyrene-block-poly(oligo-oxyethylene methacrylate) (PS-b-POEM) TBP electrolytes have exhibited improved ionic conductivities, shear moduli, and processabilities in comparison to their conventional block polymer analogues.1, 2 These features make TBPs attractive as alternative electrolyte materials for lithium-ion batteries, in which high ionic conductivities must be attained while simultaneously addressing the safety concerns that are present in current liquid electrolytes.3 In this work, we studied the microscopic characteristics of TBPs that impart these enhanced properties.4 Normal-, inverse-, and non-tapered PS-b-POEM TBPs were synthesized via atom transfer radical polymerization, in which the segment distributions were controlled by delivering monomers in a semi-batch manner through programmed syringe pumps. The monomer segment distributions in these lithium salt-doped TBPs were obtained via X-ray reflectivity. Through the guidance of a computational collaborator, these distributions were successfully modeled with coarse-grained molecular dynamics simulations that included strong ion solvation effects. This combined experimental-computational approach allowed the segregation strengths, chain conformations, and ion solvation energies of the salt-doped TBPs to be quantified as a function of taper sequence and salt concentration. By understanding how these polymer assembly and ion solvation behaviors affect ion transport, we can guide the rational design of higher-performance polymer electrolytes.

References:
2. Kuan, W.-F.; Reed, E. H.; Nguyen, N. A.; Mackay, M. E.; Epps, T. H., III, Using tapered interfaces to manipulate nanoscale morphologies in ion-doped block polymers. MRS Communications 2015, 5 (2), 251-256.
Effective and energy efficient ethylene separations from other small molecules can reduce significantly energy consumption, as compared to the state-of-the-art cryogenic ethylene purification. Zeolite materials offer high thermal and chemical stability, low toxicity and moderate cost comparing to other adsorbent materials. ZK-5 (KFI) zeolite can be synthesized with high Al content (Si/Al = 4) and have a large concentration of 6- and 8-rings in the framework; this combination allows high concentrations of accessible ion exchange positions for transition metal cations. Paired with metal cations with the appropriate radii (M^{2+} = Zn^{2+}, Mn^{2+}), the 6-rings in zeolite framework could allow for favorable adsorbate-adsorbent interactions with ethylene. These materials are likely more effective than the commonly used zeolite A based materials, and more economical than the recently proposed Ag-exchanged zeolites. Adsorption isotherms of ethylene and ethane show a larger difference in ethylene-ethane adsorption capacity at equilibrium for M^{2+}-ZK-5 than Na-ZK-5, demonstrating a higher ethylene-ethane selectivity at high pressures (> 0.15 atm). Isosteric heat of adsorption data show that Mn-ZK-5 obtains higher heat of adsorption than Zn-ZK-5 and also Na-ZK-5 (at lower coverages). Diffraction patterns and structural analysis are been investigated to elucidate the specific interactions between ethylene molecules and these ion exchanged zeolite materials.
Impact of Forced Convection and Electrolyte Cations on Spectroscopic Observations of the Electrochemical CO Reduction Reaction

Arnav S. Malkani
Advisor: Dr. Bingjun Xu
Committee Members: Dr. Feng Jiao, Dr. Yushan Yan, Dr. Raul Lobo & Dr. Joel Rosenthal

The electrochemical CO₂ reduction reaction is a method to convert a greenhouse gas, CO₂, into valuable chemicals and fuels using electricity generated from renewable sources. CO is a known intermediate in this reduction process which can be split into two steps. The first is the conversion of CO₂ to CO while the second is the further reduction of CO into hydrocarbons and oxygenates. This second step is known as the electrochemical CO reduction reaction (CORR) which shows selectivity towards C₂⁺ products under alkaline conditions using a Cu catalyst. The CORR has been shown to suffer from mass transport limitations owing to the low solubility of CO in aqueous electrolytes (~1 mM). Further, the nature of cations in the electrolyte has also been shown to have a significant impact on the CORR activity. However, the effect of mass transport limitations and the interaction of the cations with surface adsorbates under CORR conditions is poorly understood. In this work, we employ operando surface enhanced infrared absorption spectroscopy to study the effect of forced convection and the nature of alkali metal cations on the CO coverage and site distribution and correlate those findings with the CORR activity.

Our findings highlight the need for forced convection in spectroscopy owing to the loss of ~90% of the CO coverage when stirring was turned off in the spectro-electrochemical cell under reaction conditions. Further, when not stirring, the ratio of CO bound to the various catalyst sites in the spectra is not an accurate representation of the surface under reactivity conditions which include forced convection. Reactivity results show no change in the CORR product distribution without forced convection suggesting that the CO adsors in patches on the Cu surface. Cations on the other hand influence the distribution of CO adsorbed on the step and terrace sites, with the fraction of CO adsorbed on the step sites following the order of: Li⁺ < Na⁺ < K⁺ ~ Rb⁺ ~ Cs⁺, which coincides with the trend of Stark tuning rates of the linearly bonded CO band. The Stark tuning rate is a measure of the electric field intensity that the cations exert on surface adsorbed species and its trend could be attributed to the relative electrochemically relevant size of the hydrated cations. We show that by changing the nature of the cations by chelation we can alter both the site distribution and Stark tuning rate, thus, confirming the impact of cations on surface adsorbates.
Characterization and Biologic Comparability of High Producing VRC01 Reference CHO Cell Line

Douglas Nmagu
Advisor: Kelvin Lee
Committee Members: April Kloxin, Abraham Lenhoff

A prevalent issue facing Chinese Hamster Ovary, CHO, cell development is cell heterogeneity. As a result, the ability of researchers, especially between industry and academia, to replicate and build upon monoclonal antibody (mAb) production discoveries has been hindered. This is further compounded as academic facilities face limitations in acquiring industrially-relevant, high producing CHO cell lines due to the safeguards and sensitivity surrounding industrial intellectual property. It is important that we help bridge this gap, as we further recognize the importance of CHO produced mAbs to effectively treat a variety of human ailments, ranging from cancers to autoimmune diseases. In efforts, to make CHO produced mAbs more accessible and affordable to patients across the world, there have been advances to overall mAb production, but primarily in improvements in volumetric productivity from process scaling/optimization. However, this has lead to diminished mAb production improvements over time, as there still remains a need to improve the specific productivity, $q_P$, or rather the mAb production at the cellular level.

Previous literature studies have utilized the small molecule compounds known as histone deacetylase inhibitors, HDACis, to demonstrate the potential to increase $q_P$ 1.1 to 4 folds through global histone acetylation. The ability to fold increase $q_P$ is appealing to our interesting, however, it was concerning that many of the studies were performed on lower CHO producing cell lines. Therefore, in an effort to improve and gain insight into increasing $q_P$, we evaluated the effect of histone deacetylase inhibitors, HDACis, on various producing clones of a reference CHO cell line with modern industrially-relevant performance characteristics.
Homogeneous Metal Salt Solution in Biomass Upgrading

Natalia Rodriguez Quiroz
Advisor: Dion Vlachos
Committee Members: Raul Lobo, Yushan Yan, Bingjun Xu

The growing interest in biomass as a renewable feedstock drives research towards innovative processes and performance-advantaged bioproducts. Homogeneous metal salt solutions are promising media owing to their diverse applications as Lewis and Brønsted acids, salting-in(out) agents, and solvents. Their multifaceted roles render them attractive but also difficult to understand. Furthermore, the study of this systems is slowed down by current quantification techniques which are time consuming and result in significant liquid waste.

In this work, we couple experimental kinetics, NMR and UV-Vis spectroscopy, and thermodynamic modeling to understand the speciation of and the underlying mechanism of metal salts by correlating the intrinsic properties of ions in solution with experimental kinetics of key steps in biomass upgrading. We focus on (1) the activity of ions in solution, acidity, and speciation in the hydrolysis of cellulose in molten salt hydrates (MSH) (2) the effect of the ionic strength on the acidity in the dehydration of fructose to 5-Hydroxymethyl furfural (5-HMF) in concentrated salt solutions, and (3) metal salt speciation in dilute aqueous solutions with emphasis on the glucose to 5-HMF tandem reaction. Furthermore, we develop a method for fast in-line product quantification by utilizing machine learning for the systematic analysis of IR spectra.
Interfacial structure and rheological properties are critically important in biological (e.g. membranes) as well as many synthetic systems (e.g. emulsions), such that a better understanding how these are correlated can guide the formulation of systems with targeted interfacial properties. Mixed flow fields, typical of classical Langmuir trough experiments, convolute the interpretation of interfacial dilatational rheology for interfacial systems with both finite surface shear and dilatational moduli. Attempts have been made to resolve the anisotropic state of stress into their pure forms, either by multiple measurements in the Langmuir trough [1] or through the use of a radial trough [2]. In this presentation we describe the design and implementation of a new “Quadrotrough” to better approximate pure dilation or shear interface kinematics. The rheological capabilities of this interfacial trough are demonstrated through proof-of-concept experiments on widely studied, model systems. Importantly, this enables more clearly studying the path dependence of interfacial isotherms. Samples investigated include model steric acid insoluble monolayers as well as more complex macromolecules at the air-water interface. We critically test the hypothesis that anisotropic compression will only significantly affect the dilatational rheology for interfaces with finite surface shear moduli. This is important because convolution of mixed flow fields greatly complicates data interpretation. Combining the new Quadrotrough with both Brewster angle microscopy and neutron reflectivity provides detailed structural measurements of the interface at the microscale and nanoscale that elucidates the source of this path dependence. The potential for rheo-MAGIK, will be discussed and future investigations are reflected on for instrument development.

References
Supported metal nanoparticles on oxide supports are widely applied as environmental catalysts. The catalyst activity is strongly affected by the nanoparticle size. Recently, single atom catalysts are being explored as effective and selective catalysts for several chemistries \cite{1,2}. Yet, there is often a strong debate whether single atoms or subnanometer clusters of a few atoms carry out the chemistry. The debate arises in part due to the inability to observe the catalyst structures operando and is further complicated by adsorbates, such as CO, which facilitates catalyst reconstructing \cite{3}. Resolving the structure and composition of supported nanoparticles under reaction conditions remains a challenge. Density functional theory (DFT) is a powerful tool to predict the energetics of metal-support and adsorbate-metal interactions. However, the computational time needed to describe numerous clusters and sites at the long-time scales for sintering make direct first-principles calculations impractical. To address this multiscale problem, here we introduce statistical learning tools to rapidly predict energetics and dynamics of Pd\textsubscript{n} clusters supported on CeO\textsubscript{2}(111), which are essential in automobile three-way catalysis. We employ a three-dimensional cluster expansion (CE) to describe the energies of Pd\textsubscript{n} clusters (n = 1-21) on CeO\textsubscript{2}(111) and use LASSO regression to select the significant terms of the CE Hamiltonian. By employing the statistical learning models, the optimal cluster structures are identified by a metropolis Monte Carlo based Genetic Algorithm. Kinetic Monte Carlo simulations predict the dynamics of single atoms toward sintering under the reaction conditions. We introduce a computational framework to enable atomistic level structure determination for single atom and subnanometer cluster catalysts. We apply these methods to reveal for the first time the optimal structures and long-time stability of the catalysts under reaction conditions. The methodology is applicable to any metal/support system.

References
**Effect of polymer architecture on the self-assembly in amphiphilic polymers solutions: A computational study**

Michiel G. Wessels  
Advisor: Prof. Arthi Jayaraman  
Committee Members: Prof. Norman J. Wagner, Prof. Eric M. Furst, and Prof. Darrin J. Pochan

Nanostructured assemblies of amphiphilic block polymers are used in a wide variety of applications such as drug delivery, bio-imaging, nanoreactors, environmental cleanup, microelectronics, optics, and sensing. In the past, block polymers with linear architecture have been the focus of many computational and experimental studies investigating self-assembly into micelles both in solution and on/near surfaces. Recent advances in polymer synthesis have led to other complex non-linear polymer architectures (e.g., bottlebrush, star, comb, coil-brush) which can show previously unseen self-assembly behavior. There is, however, a lack of systematic studies aimed at relating varying non-linear polymer architectures and polymer chemistry to the self-assembly of these molecules in solution. In this talk, I will present computationally derived and experimentally validated design rules for tailoring the structure and thermodynamics of the nanostructured assemblies with varying polymer architecture along with fundamental insights into the underlying molecular driving forces leading to those design rules.

In the first part of my talk I will compare results of self-assembly obtained from my coarse-grained molecular dynamics (CGMD) simulation along with results from experiments conducted in the lab of Prof. Karen Wooley at Texas A&M, for a wide range of coil-brush polymers. The qualitative agreement in the observed morphologies for the various design parameters and quantitative agreement in sizes of spherical micelles validate our coarse-grained model. Using that validated model, we conduct CGMD simulations to compare the self-assembly of linear, bottlebrush, and star-like polymer architectures in solution and at attractive surfaces. We link the ability of different branched polymer architectures to conform and pack in the assembled state to the resulting micelle size, shape and stability in different solvent conditions. Additionally, we establish design rules that guide how one could manipulate the amphiphilic block polymer assembly structure and thermodynamics by choosing appropriate polymer architecture, block sequence, and composition. In the final part of my talk I will present our ongoing computational work focused on interpretation of the self-assembled micelle structures scattering profiles obtained from small-angle neutron scattering (SANS). Our approach called Computational Reverse-Engineering Analysis of Scattering Experiments (CREASE) overcomes one of SANS users’ major limitation of having to rely on analytical models to interpret the structure, and instead uses genetic algorithm to recreate a micelle structure whose scattering profile would match the experimental scattering profile. The success of CREASE is demonstrated by its ability to replicate, quantitatively, the results from *in silico* experiments, and by the agreement in micelle core and corona sizes obtained from microscopy of *in vitro* solutions.
Impact of Uncertainty Quantification and Density Functional Theory Error Propagation on Surface Catalyzed Kinetic Models

Gerhard R. Wittreich
Advisor: Dionisios G. Vlachos
Committee Members: Antony N. Beris, Raul F. Lobo

Parametric uncertainty currently limits the predictive ability of kinetic models and prevents them from reaching their full potential. This is particularly important for surface catalyzed microkinetic models which often contain thousands of reaction steps and intermediate surface species requiring many kinetic and thermodynamic parameters. Estimating these from first-principles density functional theory (DFT) is costly, motivating the need for DFT-based methods, such as group additivity and Brønsted–Evans–Polanyi (BEP) relationships, to quickly estimate thermochemistry and reaction barriers, respectively. Uncertainty in DFT and these methods is amplified by the Boltzmann factors in the reaction rate expressions increasing uncertainty in estimating the overall reaction rate. Furthermore, microkinetic model parameters are correlated; yet, there is lack of a framework to expose these correlations on a single catalyst with a single functional. This work develops a framework to estimate correlations in reaction networks and DFT uncertainty propagation. Uncertainty quantification (UQ) is then predicted for thermochemistry, reaction barriers, reaction path, and ultimately reaction rates. Propane combustion and ethane oxidative dehydrogenation reactions are illustrated. We will also demonstrate how to use the Python multiscale thermochemistry toolbox, pMuTT [1], to calculate uncertainty in apparent activation energy.
Flowsheet and Mathematical Modeling in Batch and Continuous Monoclonal Antibody Production.

Ou Yang
Advisor: Prof. Marianthi G. Ierapetritou
Committee Members: Shishir Chundawat, Rohit Ramachandran, Maen Qadan

Biopharmaceutical manufacturing mainly produces protein-based therapeutic products which are becoming increasingly important in recent years due to the high demand. To satisfy this demand, there is a trend of biopharmaceutical manufacturing shifting from conventional batch processing to continuous processing. Continuous processing shows a great benefit in increasing productivity, reducing the footprint, capital and operating cost over batch mode. Much research has been focused towards the development of continuous processing for biologics production including perfusion bioreactor, tangential flow filtration, and multi-column chromatography. However, to integrate upstream and downstream processing and to ensure high product quality and productivity, it is important to enhance the understanding of each unit operation involved in biological processes.

In this work, we apply multiple simulation methods to capture biopharmaceutical processes. Flowsheet modeling has been used to simulate mass balance and process scheduling to design continuous biologics production. This is compared to the conventional batch operation using techno-economic analysis. Capital cost and operating cost under different manufacturing scenarios are evaluated, including various operating throughput, upstream titers and downstream yields for both operation modes. Upstream bioreactor is one of the crucial operations in the integrated process due to its high operating cost. Additionally, its productivity and product quality affect downstream operations and the overall manufacturing performance. To further enhance the process understanding, mathematical modeling is applied to upstream bioreactor operation. This is used to capture the complex nonlinear bioprocess dynamics between process operating conditions and output variables including cell concentration, nutrient consumption and product titer. Protein glycosylation process is simulated using a detailed single cell model to predict product quality. An integrated model will be built for flowsheet modeling coupled with detailed first principle mathematical model, to capture the mass balance, process scheduling, and critical quality attributes of the overall process.
Targeted Delivery of Therapeutic Proteins to Inflammatory Breast Cancer Cells Using Modular Hepatitis B Virus-like Particles

Daniel Yur
Advisor: Wilfred Chen, Millicent O. Sullivan
Committee Members: Eleftherios T. Papoutsakis, April M. Kloxin

Protein therapeutics offer enormous potential for clinical impact in treating a variety of diseases by offering high selectivity, with fewer off-target effects in vivo than small molecule drugs. However, delivery challenges, such as short circulation half-lives, protein instability, and limited membrane translocation, severely reduce the quantity of functional proteins reaching their target, requiring frequent administration. To address these problems, protein encapsulation in nanocarriers can provide enhanced targeting and protease protection until delivery into their intended cell target. Non-viral vectors, such as polymer nanoparticles, offer high loading capacity and design flexibility by leveraging established chemical syntheses; however, large particle size and polydispersity cause rapid hepatic clearance and slow and non-uniform cargo release, which hinder effective delivery.

In nature, viruses have developed an efficient delivery system because of their regular, multifunctional architecture and their ideal size, conducive to mononuclear phagocytic system evasion. Derived from their viral analogues, virus-like particles (VLPs) are non-infectious viral capsids, which have potential for drug delivery applications because of their similar structural characteristics. The well-characterized hepatitis B virus (HBV) VLP is an ideal protein delivery vehicle because it has regular solvent exposed features for incorporating surface modifications, a large interior cavity amenable for loading various cargos, and a diameter of 36 nm for favorable pharmacokinetics. In our work, a targeted VLP delivery system was established by incorporating the SpyCatcher-SpyTag protein-peptide pair, which facilitates site- and orientation-specific nanocarrier modification in a plug-and-play manner: SpyTag was genetically inserted into the HBV capsid, while SpyCatcher was fused to potential delivery moieties, such as purification tags and targeting ligands. Simultaneously, genetic fusion at the C-terminus of the HBV monomers facilitated internal loading of two sample therapeutic proteins: prodrug-converting enzyme, yeast cytosine deaminase (yCD), and the short interfering RNA (siRNA)-binding protein, p19. Currently, we have assembled the SpyTagged HBV capsid with active p19 and yCD proteins and have demonstrated SpyTag functionality by initially ligating an elastin-like polypeptide purification tag for a temperature driven, scalable purification method. We then added an mCherry targeting module, conjugated with clustered GE11 targeting peptides, to bind epidermal growth factor receptors that are commonly overexpressed in cancer cells, and we demonstrated enhanced nanocarrier uptake in inflammatory breast cancer cells as opposed to non-malignant breast epithelial cells in our resultant targeted nanocarrier. Future work will incorporate endosomolytic peptides for targeted cytosolic delivery of siRNA and yCD in inflammatory breast cancer cells to enable analysis of targeted gene silencing and enhanced cell death.
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