2022 POSTDOCTORAL RESEARCH AND CAREER SYMPOSIUM

Leadership Institute at Argonne National Laboratory

Argonne National Laboratory Postdoctoral Research and Career Symposium

October 26, 2022

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NETWORKING LUNCH

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Welcome Remarks

Dr. Paul K. Kearns
Laboratory Director
Argonne National Laboratory

Paul K. Kearns has served as Director of the U.S. Department of Energy (DOE) Argonne National Laboratory since 2017. Argonne is a growing multidisciplinary science and engineering research center with a $1.2 billion diversified research portfolio and more than 3,300 employees, 8,000 facility users, and 800 visiting researchers. Kearns has set the laboratory's strategic vision to deliver pivotal discoveries, pioneering leadership, and powerful scientific tools and facilities. He has strengthened sponsor relationships and fostered a welcoming and inclusive culture valuing diversity, innovation and collaboration, and laboratory impact.

A biologist and accomplished steward of diverse scientific resources, Kearns has managed complex research and development enterprises for over 30 years, enabling them to achieve ambitious goals in energy, environment, and national security. As Argonne laboratory director, Kearns oversees multiple projects critical to Argonne's mission of accelerating science and technology to drive U.S. prosperity and security. Upgrading the Advanced Photon Source and launching the first exascale computer in the U.S. at the Argonne Leadership Computing Facility are critical to maintain U.S. leadership in science and technology. Argonne's leadership of the Joint Center for Energy Storage Research reflects the laboratory's long history of battery science expertise and collaboration.

As Argonne chief operations officer from 2010 to 2017, Kearns directed over 900 staff providing mission support services in financial management, human resources, safety performance, business systems, technology commercialization, and facilities management. Kearns also guided the establishment of an independent energy storage start-up and directed construction of cutting-edge research laboratories.

Keynote Address

Dr. Asmeret Asefaw Berhe
Director of the Office of Science
U.S. Department of Energy

Dr. Asmeret Asefaw Berhe is the Director of the Office of Science for the U.S. Department of Energy. Dr. Berhe was most recently a Professor of Soil Biogeochemistry; the Ted and Jan Falasco Chair in Earth Sciences and Geology; and Interim Associate Dean for Graduate Education at the University of California, Merced. Her research was at the intersection of soil science, global change science, and political ecology with an emphasis on how the soil system regulates the earth's climate and the dynamic two-way relationship between the natural environment and human communities.

She previously served as the Chair of the US National Committee on Soil Science at the National Academies; was a Leadership board member for the Earth Science Women's Network; and is currently a co-principal investigator in the ADVANCEGeo Partnership – a National Science Foundation funded effort to empower (geo)scientists to respond to and prevent harassment, discrimination, bullying and other exclusionary behaviors in research environments. Her scholarship on how physical processes such as erosion, fire, and changes in climate affect the biogeochemical cycling of essential elements in the earth system and her efforts to ensure equity and inclusion of people from all walks of life in the scientific enterprise have received numerous awards and honors. She is a Fellow of the American Geophysical Union and the Geological Society of America, and a member of the inaugural class of the US National Academies New Voices in Science, Engineering, and Medicine.

Berhe was born and raised in Asmara, Eritrea. She received a B.Sc. in Soil and Water Conservation from the University of Asmara, an M.Sc. in Political Ecology from Michigan State University, and a Ph.D. in Biogeochemistry from the University of California, Berkeley. In 2020 she was named a Great Immigrant, Great American by the Carnegie Corporation of New York.

Career Panel

Navigating Career Paths in Industry, Academia, and Government

Dr. Nigel Becknell
Principle Technology Development Scientist
Nanograf

Dr. Nigel Becknell has over ten years experience in developing materials and electrochemical interfaces that solve challenges in a wide range of battery and fuel cell technologies. He recently completed a postdoctoral fellowship at Argonne National Laboratory where he synthesized novel materials for electrochemical systems and applied advanced characterization techniques. Nigel received a Ph.D. in Chemistry from the University of California, Berkeley and a B.S. in Chemical Engineering from the University of Wisconsin-Madison.

Prof. Hrant Hratchian
Associate Professor and Vice Provost and Dean for Graduate Education
University of California, Merced

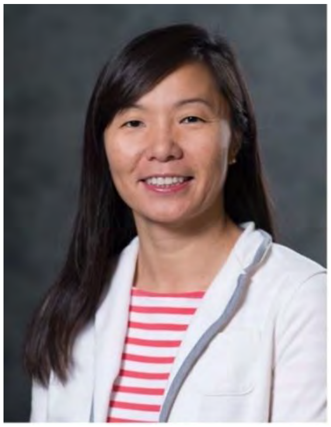
Hrant P. Hratchian is an Associate Professor in the Department of Chemistry and Biochemistry and Vice Provost and Dean for Graduate Education at the University of California Merced. A Michigan native, he obtained his B.S. degree in chemistry from Eastern Michigan University (Ypsilanti, MI) and completed doctoral studies under the tutelage of Professor H. Bernhard Schlegel at Wayne State University (Detroit, MI) where he was an NSF-IGERT Graduate Fellow. From 2005-2008 he was the Ernest R. Davidson Postdoctoral Fellow at Indiana University (Bloomington, IN), where he worked with Professor Krishnan Raghavachari. From 2008-2013, he was a Research Scientist at Gaussian, Inc. (Wallingford, CT). In 2013, he joined the faculty at the University of California Merced in the Department of Chemistry & Biochemistry.

Prof. Caroline Chick Jarrold
Herman B Wells Endowed Professor of Chemistry
Indiana University

Caroline Chick Jarrold received her B.S. in Chemistry at the University of Michigan, Ann Arbor, in 1989, and her Ph.D. in Physical Chemistry from the University of California, Berkeley, in 1994. After being a University of California President's Postdoctoral Fellow at UCLA, she joined the Chemistry faculty at the University of Illinois, Chicago in 1997, and was there until her move to the Indiana University Chemistry Department in 2002. Professor Jarrold's research involves applying a combination of gas-phase reactivity, mass spectrometry, anion photodetachment spectroscopies, and computational chemistry toward issues of importance in energy and environment.

Dr. Shalaka Shinde
Senior Project Manager of Research Program
Pivot Bio

Dr. Shalaka Shinde is an experienced product manager who has developed and launched products into global market. She is a skilled researcher in molecular biology, next-generation sequencing, transcriptomics, proteomics, and plant-microbe interactions with over 15 years of experience. She is proficient in communicating scientific concepts to professionals at all levels, including scientists, business executives, technicians, legal staff, and field workers; with experience in working with regulatory bodies, including the USDA, EPA, and DOE.



Prof. Lucy Zhang
Professor of Mechanical Engineering
Rensselaer Polytechnic Institute and National Science Foundation

Prof. Lucy T. Zhang is a professor of Mechanical Engineering at Rensselaer Polytechnic Institute and currently serving as a program director at NSF for Mechanics of Materials and Structures (MoMS) and Biomechanics and Mechanobiology (BMMB) programs. Her research focuses on computational mechanics with special emphasis on fluid-structure interactions, computational biomechanics, and multiscale modeling of interfacial interactions. Prof. Zhang received her B.S from Binghamton University, and obtained her M.S.and Ph.D. from Northwestern University, respectively. Upon the completion of her Ph.D., she joined the Mechanical Engineering department at Tulane University in New Orleans as an assistant professor. Due to Hurricane Katrina, she then moved and became a faculty in the Department of Mechanical, Aerospace, and Nuclear Engineering at RPI.

Over her illustrious career, she has been recognized for her exemplary research, international collaborations, and outreach endeavors. In 2013, she received the Japan Society for the Promotion Science (JSPS) Faculty Fellowship followed by the Young Investigator Award at the International Conference on Computational Mechanics in 2016. She was also named a fellow of the American Society of Mechanical Engineers in 2020. Very recently, she has started hosting a podcast series, This Academic Life, aimed at everyone who aspires to be a successful STEM educator and researcher. In each episode, they share stories and look into the incredible journey of people from the varied branches of academia.

Abstracts

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APPLIED MATERIALS

25 Investigating Various Failure Models on Commercial SiC CSP Receivers

Pawan Chaugule[1], Mark C. Messner[1], Bipul Barua[1], and Dileep Singh[1]

[1]Argonne National Laboratory, 60439.

Structures and components made from ceramic materials are often brittle and can fail by the unstable growth of existing flaws such as voids and cracks. There have been several failure criteria developed for ceramics, in the past and they are broadly categorized based on their dependency on crack geometry. The present work implements eight failure criteria using an open-source software package – *srlife*, which predicts the lifetime or failure probability of concentrated solar power (CSP) structural components. The present work also checks the viability of building a ceramic CSP receiver, by analyzing the reliability predictions from *srlife* for a SiC ceramic. The reliability predictions for a transverse loading problem indicated the Shetty Mixed-Mode criterion gives the most conservative predictions. Whereas, in case of the CSP receiver, the predictions show that the co-planar strain energy criterion gives the most conservative predictions as it is agnostic towards the type of stress, and therefore, is not recommended to be used designing ceramic receivers.

33 Binder Jetting Additive Manufacturing and Its Application in Solar Energy

Mark Du[1] and Dileep Singh[1]

[1]Applied Materials Division, Argonne National Laboratory, Lemont, IL 60439

Binder jetting additive manufacturing has many advantages when compared with not only traditional manufacturing methods but also other additive manufacturing ones. One application is a ceramic heat exchanger with excellent corrosion and oxidation resistance that will be used in the concentrated solar plant (CSP). Here techniques were developed for additively manufacturing ceramic materials for applications involving high temperature, high pressure, and high corrosion resistance as needed for the CSP application. Based on a previous ceramic heat exchanger design, a prototype with integrated headers and incorporated flow channel dimensional compensations was fabricated by using the binder jetting process. The printed prototype heat exchangers were successfully densified through the processes of liquid polymer infiltration and pyrolysis. In addition, the thermophysical properties of the densified silicon carbide parts were measured to provide necessary design information. Experimental heat transfer testing of the lab-scale prototype heat exchanger was conducted, and the experimental data agreed reasonably well with the simulated results.

36 Density Functional Theory Computed Descriptors for Heterogeneous Catalysis of CO_2 Sorbent Amines

Joshua J. Gabriel[1], Reginaldo J Gomes Neto[2], Tony J. Mathew[3], Jiayi Xu[4], Ritesh Kumar[2], Cong Liu[4], Noah H. Paulson[1,5], Chukwunwike Iloeje[3], and Chibueze V. Amanchukwu[2]

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[3]Energy Systems and Infrastructure Analysis Division, Argonne National Laboratory, Lemont, IL 60439
[4]Chemical Sciences and Engineering Division, Argonne National Laboratory, Lemont, IL 60439
[5]Data Science and Learning Division, Argonne National Laboratory, Lemont, IL 60439

Integrating carbon capture and conversion is a promising direction towards a decarbonized future for industries that are heavily dependent on carbon-based raw materials. Progress has been made in achieving the electrocatalytic reduction of carbon dioxide to carbon monoxide from CO_2 captured in a monoethanolamine solution on silver catalysts. In this work, we use density functional theory calculations to test a hypothesis that the coverage of alkali cation controls the relative binding strength of proposed species of reactants, byproducts, and products, and hence the reaction product selectivity. Our results support the hypothesis that the alkali cation affects the relative binding strength of species for CO_2 reduction and hydrogen evolution, with increased coverage favoring CO_2 reduction. This discovery provides a useful computational descriptor to better understand the process conditions that can control product selectivity in heterogeneous catalysis.

39 A Framework for Modeling Coupled Electro-Chemo-Thermo-Hydro-Mechano-Fracture Phenomena in Dissipative Solids

Tianchen Hu[1], Mark C. Messner[1], Pallab Barai[1], Bipul Barua[1], Brandon Talamini[2], Andrew J. Stershic[2], Michael R. Tupek[2], Wen Jiang[3], Benjamin W. Spencer[3], and John E. Dolbow[4]

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[2]Sandia National Laboratories
[3]Idaho National Laboratory, Idaho Falls, ID 83415
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Many practical engineering applications exhibit multiphysics phenomena. From both theoretical and implementational standpoints, modeling fully coupled multiphysics systems remains to be one of the most challenging tasks in the engineering community. We developed a variational framework which describes the fully coupled multiphysics system as an optimization problem. The formulation automatically guarantees thermodynamic consistency; a unique solution exists if the potentials are strictly convex; fully coupled constitutive models follow from variational principles. We also developed two applications, RACCOON and EEL, using this theoretical framework. The apps use a modular design so that engineers with different domain knowledge can work together; the apps are massively parallel with perfect scaling efficiency up to hundreds of thousands of processors; the apps provide a clean and flexible interface for end users. The framework has been successfully applied to model practical electro-chemo-thermo-hydro-mechano-fracture coupled systems with high fidelity, including charging and discharging of Lithium-ion batteries and solid-state batteries, pressurized fracture in high burn-up microstructures, cohesive fracture and soil desiccation, ductile fracture, creep fracture, and oxide spallation on high-temperature heat exchangers.

55 Additive manufacturing of mullite ceramic by digital light processing

Jung-Ting Tsai[1], and Dileep Singh[1]

[1]Applied Materials Division, Argonne National Laboratory, Lemont, IL 60439

The combination of polymer-derived ceramic (PDC) and digital light processing (DLP) provides a new pathway for ceramic additive manufacturing. Large-scale prototypes using mullite UV ceramic precursors have been successfully printed and sintered without compromising structural integrity while concurrently maintaining fine intricate (50 um) designs on the parts. The structure-property relationship of the post-sintered mullite is investigated and characterized for determining the material performances. The proposed methodology assists in decreasing manufacturing time and increases designed flexibility.

The work has been supported by the ANL’s LDRD program, project number 22022-0078

CHEMICAL SCIENCES AND ENGINEERING

2 Impact of Anchoring Groups on Photoinduced Charge Injection from Cu(I) Complexes Into Electrode Substrates

Hala Atallah and Karen L. Mulfort

Copper(I) coordination complexes have attracted significant attention from a desire to synthesize stable photosensitizers using environmentally sustainable materials. Copper is a viable element for constructing photosensitizers due to their abundance and low cost, but unfortunately, the metal-to-ligand charge transfer excited state of Cu(I)diimine complexes undergo a Jahn-Teller flattening distortion (D2d to flattened D2). This flattened state is susceptible to exciplex formation and rapid deactivation via non- radiative decay, leading to very short excited state lifetimes which is problematic for diffusional reactions. This Jahn-Teller distortion has previously been attenuated by extensive synthetic efforts to add targeted steric bulk on the diimine ligands. In this work we take a complementary approach by investigating how electrode-immobilization of molecular Cu(I)diimine complexes within confined environments allows us to systematically investigate the effects of confinement on photo-induced excited state dynamics, and eventually electron transfer between molecular electron donors and acceptors, as well as electron transfer across tailored interfacial chemistry. Our group showed, that decreasing the pore size of the anodic aluminium oxide (AAO) framework from 40 to 10 nm results in more than twice increase in the ^{3}MLCT lifetime than in solution which is consistent with inhibited structural flattening as pore size decreases. Building on our previous work, my current work is focused on desigining and synthesizing new heteropletic Cu(I)diimine complexes with different anchoring groups and fully characterize their behavior in a variety of environments, including attached to electro-active surfaces and in nanometer-sized pores to understand the impacts of molecular confinement.

6 Temperature Measurements in Heavily-Sooting Ethylene/Air Flames Using Synchrotron X-ray Fluorescence of Krypton

Colin Banyon[1], Matthew J. Montgomery[2], Hyunguk Kwon[3], Alan L. Kastengren[1], Lisa D. Pfefferle[2], Travis Sikes[1], Yuan Xuan[2], Charles S. McEnally[3], and Robert S. Tranter[1]

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High-fidelity temperature field measurements have been made for several heavily-sooting ethylene/air flames that have historically been challenging environments for conventional optical diagnostics. These challenges have largely been overcome here by conducting x-ray fluorescence (XRF) measurements of a Kr fluorescent agent in the hard x-ray regime (15 keV). The current methodology presents a more economical diagnostic than a previously reported implementation of the Kr-XRF method, by limiting seeding of the expensive fluorescent agent to only the fuel stream flows. Detailed reacting flow simulations have been used to interpret experimental signals by tracking the mole fraction of the fluorescent agent in the flow field. Simulated Kr densities are in excellent agreement with measurements throughout the flow field. Temperature measurements of the flow field also agree well with simulations and recent literature studies. However, uncertainties in the measurements become increasingly large downstream of the burner surface as the krypton fraction drops due to mixing of the fuel and co-flow streams. Additionally, we demonstrate that soot particles in the heavily sooting flames studied do not impede the Kr-XRF measurements.

7 Utilization of Proton-Responsive Ligands to Facilitate Electrochemical CO_2 Capture and Conversion

Jeffrey M. Barlow[1], David M. Kaphan[1], and David M. Tiede[1]

[1]Chemical Sciences and Engineering Division, Argonne National Laboratory, Lemont, IL 60439

Alkoxides are a promising platform for CO_2 capture systems, capable of atmospheric capture applications. These systems remain highly inefficient however, due to the requirement of thermal swings to cycle between the capture and release steps. Electrochemical reduction of the alkyl carbonate species formed upon CO_2 binding avoids the need for thermal alkoxide regeneration. Rhodium bis-phosphine complexes featuring hydroxyl functionality were synthesized and their ability to capture and reduce CO_2 investigated. Upon reduction of the complexes, deprotonation of a hydroxyl group occurs to form a Rh–H bond. Once deprotonated, the resulting alkoxide can bind to CO_2 to form an alkyl carbonate species. The activity of each complex is compared to the corresponding bis-phosphine complexes lacking hydroxyl functionality.

12 Mechanistic Study of Formamide Photo-dissociation at 193 nm via Chirped-Pulse Fourier Transform Millimeter Wave Spectroscopy

Kacee L. Caster[1], Nathan A. Seifert[1,2], and Kirill Prozument[1]

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Formamide (H_2NCHO) is the simplest molecule containing a peptide linkage [-NH-C(=O)-] and plays an essential role in the study of prebiotic chemistry, in part to its many desirable chemical properties and ubiquitous presence throughout the universe. Its exposure to UV irradiation, allows formamide to decompose and act as a prebiotic feedstock in the formation of nucleobases and other necessary starting materials. The photo-dissociation mechanism of gaseous formamide at 193 nm is studied using chirped-pulse Fourier transform millimeter wave (CP-FTmmW) spectroscopy in the 260‒290 GHz spectral region in a room-temperature flow-tube reactor. Photoproducts of deuterated formamide precursors (H_2NCDO and D_2NCHO) are also examined to gain additional insight into the photo-dissociation mechanism. The photoproducts identified in this region include HCN, HNC, HNCO, HCO, DCN, DNCO, NH_2D, and NHD_2. Experimental branching ratios are reported, and the time evolution profiles analyzed to elucidate the reaction dynamics. The formamide decomposition pathways discussed are dehydration (HCN/HNC +H_2O), dehydrogenation (HNCO + H_2, HNCO + 2H), decarbonylation (NH_3 + CO), and simple bond fission (NH_2 + HCO).

13 Charge Separation in Natural Photosynthesis at the Single Protein Level Using Diamond NV Center Detection

Claire W. Chang[1], Mouzhe Xie[2], Ignacio Xionkon Chi Duran [2], Xiaofei Yu, Stella Wang[2], Peter Maurer[2], Jens Niklas [1], Lisa M. Utschig[1], and Oleg G. Poluektov[1]

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Our research is focused on understanding fundamental mechanisms that control solar energy conversion in photosynthetic proteins. Light-induced electron transfer initiates energy conversion reactions in integral membrane proteins called reaction centers (RCs). This process involves rapid, sequential electron transfers that result in efficient charge separation across the membrane, establishing an electrochemical potential. As a first step of charge separation, two unpaired electron spins, one located on the primary donor and the other on a quinone acceptor, are created. These spins form a so-called Spin Correlated Radical Pair (SCRP) where two electron spins are entangled with each other. Currently, SCRPs are extensively investigated by advanced EPR techniques. Most of this research is done on an ensemble of proteins in solution. Recently, a quantum sensing technique based on the nitrogen vacancy (NV) centers in diamond was developed that allows single electron spin detection. We propose to use NV center in diamond as a sensor to study charge separation processes at the single protein level. Spin dynamics of charge separation processes in a single protein will improve our understanding of correlated spin process in natural photosynthetic proteins. The first step in the project involves the immobilization and characterization of individual photosynthetic proteins on the surface of a diamond sensor. As a model protein for this project, we used bacterial reaction center protein (bRC) from Rb. sphaeroides. The R26 bRC is a carotenoidless mutant that has long-lived charge separated state. To characterize RC location on the surface of diamond sensor we used a fluorescent-maleimide label cy3, which we selectively covalently bound to bRC. Three immobilization methods were established and are currently being tested in the NV-center based setup which will be used for ensemble and single molecule detection. This work will provide the basis for future studies for single molecule imaging using diamond NV-center and will provide new insight.

30 Understanding The Ultrafast Electron Injection From CuHETPHEN Into TiO_2 For Efficient Solar Energy Conversion Applications

Kaustav Dey,[1] Brian T. Phelan,[1] Lin X. Chen,[1,2] Karen L. Mulfort[1]

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Photoinduced electron transfer (ET) from photosensitizers to semiconductor metal oxide surface has significant impact in solar fuel generation, photocatalysis and dye sensitized solar cells (DSSCs). Therefore, it is very important to obtain detailed insight into the kinetics of molecular complexes bound to semiconductor surfaces for the development of efficient and cost-effective photovoltaics. In this work three heteroleptic copper(I) bisphenanthroline (CuHETPHEN) complexes with surface anchoring carboxylate groups have been immobilized on TiO_2 nanoparticulate thin films and their ground state (UV-Vis, cyclic voltammetry) and excited state (ns and fs-TA) properties have been studied to understand the chemical stability and photophysical properties of the CuHETPHEN-TiO_2 hybrid system. We observed a notable difference in the Cu (I/II) oxidation potential for the complexes after immobilization which presumably due to the change in the lowest unoccupied molecular orbital (LUMO) levels of CuHETPHEN-TiO_2 hybrids. The excited-state dynamics of the CuHETPHEN-TiO_2 hybrid showed a highly efficient and ultrafast (< 300 fs) interfacial photoinduced electron injection from the ^{1}MLCT state to the conduction band of the TiO_2 and prolonged charge recombination (< 10 μs). In summary, we have designed the CuHETPHEN complexes in such a way that by fine-tuning the structural constraint in the ligand geometry, the directionality of initial MLCT and charge transfer kinetics from photosensitizer to metal oxide surface was changed in a significant way which will have potential impact in solar energy conversion.

37 Ultrafast Spectroscopy of Strongly Cooperative Spin-Crossover Nanoparticles

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We investigated the light-induced switching events in nanoparticles of the strongly cooperative $[Fe(Htrz)_2(trz)](BF_4)$ spin-crossover material. Spin-crossover materials serve as a prototypical bistable materials to study photo-induced phase transformations towards material control by reversible switching.

After excitation into an MLCT band and subsequent ultrafast high spin (HS) formation, we observe dynamics across several decades in time. In the first few picoseconds, we see size-dependent oscillations which were identified to be acoustic breathing modes. Over the next 30 ps, a strong spectral shifting occurs in the transient signal. We determined that this spectral evolution is due to the build-up of HS states at distorted geometries within the nanoparticles. The mechanically relaxed lattice ultimately undergoes biexponential decay due to the different HS stability between bulk and distorted geometries.

When the laser-induced excitation fraction exceeds 2%, we see another feature: an additional series of LS→HS switching during the lattice expansion. Until now, this effect had not been observed in nanoparticles. By measuring nanoparticles of different sizes, we showed that the elastic amplification time was consistent with the longitudinal expansion timescale. For our smallest particles, the elastic step was 5 ps, which is orders of magnitude faster than previously observed.

38 Development of Operando Grazing-Incidence Pair Distribution Function for Analysis of Cobalt Oxide Water-Splitting Catalysts Under Electrochemical Bias

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Operando grazing-incidence pair distribution function (GIPDF) of molecular-domain water-splitting catalysts under electrochemical bias can give key insights on the mechanism of oxygen production. Of particular interest is amorphous cobalt phosphate formed through electrolytic deposition ("CoPi") which has respectable water-splitting efficiency at catalytic oxidative potentials. The structure of CoPi has previously been deduced using PDF of a scraped-off film; however, resolving the structure of native film under electrochemical bias remains a challenge. This requires analysis of native thin (<500 nm) films which can be fully biased without significant impedance present in thick films. GIPDF offers excellent sensitivity and fast acquisition times needed for operando experiments. Here is demonstrated a custom electrochemical cell design which allows for GIPDF on ultrathin CoPi films. GIPDF has been performed on CoPi films as thin as 30 nm, the thinnest first-row transition metal oxide film studied by PDF to date. Orientational analysis is also possible from a single GIPDF scan. Additionally, depth-dependence of CoPi films was accomplished by varying the incident angle of the incoming X-rays to distinguish the structure of the surface versus bulk. Finally, a CoPi film under a layer of electrolyte solution was resolved, allowing for future operando experiments under water-splitting electrocatalytic biases.

41 Doping Rare Earth Nitrate Hybrid Double Perovskites from Aqueous Solution

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Hybrid double perovskites are a family of materials with the general formula $A_2BB'X_6$ (A = organic cation, B and B' = metal cations, X = inorganic anion). These materials, especially those containing trivalent f-elements, have recently become of interest due to their potential applications in dielectric switches, piezoelectrics, and photoluminescent materials. We have isolated a new family of crystalline, cubic ($Fm3-3$-m) hybrid double perovskites containing tetramethylammonium (TMA) as the organic cation which is isostructural across the entire rare earth series, as well as for americium and curium, with the general formula $(TMA)_2MK(NO_3)_6$ (M = Y, La-Nd, Sm-Lu, Am, Cm).

Preliminary attempts to dope Eu^{3+} into the $(TMA)_2LaK(NO_3)_6$ phase have shown promise. Synthetic preparations with varying La/Eu mole ratios have all produced single crystals which are isostructural with the undoped phase. Additionally, there is good agreement between experimentally added dopant amounts, crystallographic refined occupancies, and metals composition determined by ICP-MS for lower Eu concentrations (<10 mol %). However, at higher dopant concentrations (15-50 mol %) the resulting product is heterogeneous, indicating that the solubility difference between La and Eu plays a significant role in the composition of the product. A low-temperature phase change has also been observed by VT-Raman spectroscopy.

This work was conducted at ANL, operated by UChicago Argonne LLC for the United States Department of Energy (U.S. DOE), and supported by the U.S. DOE Office of Science, Office of Basic Energy Sciences, Chemical Sciences Geological and Biosciences Division, Heavy Elements Chemistry program under Contract DE-AC02-06CH11357.

42 Upcycling of polyolefins using supported catalysts

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Polyolefins constitute over half of the industrial plastic market, with 80% utilized in single-use products. Current plastic waste management options are typically limited and consist of incineration, pelletizing- and molding-based recycling, and disposal into overflowing landfills. Furthermore, these methods often lead to hazardous emissions, downcycled products with restricted applications, and widespread environmental concerns. Therefore, there is a substantial need to seek alternative processes to upcycle used plastics into value-added products.

This work targets the upcycling of polyethylenes using supported platinum nanoparticle catalysts selective for deconstruction via hydrogenolysis. This process yields monodisperse liquid or wax products with significantly reduced molecular weights and increased branch density. Initial results suggest that the size and structure of the starting polymer only marginally affects the size and properties of the resulting product, allowing the potential to upcycle multiple streams of waste polyethylenes into a single product.

45 Sorption of Y(III) on orthoclase (001) studied by X-ray Reflectivity

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The mobility of heavy metals in the environment is controlled by their interactions with charged mineral phases. In this study, we investigate the adsorption of Y(III), as a representative for rare earth elements and an analogue of trivalent actinides, onto the orthoclase (001) basal plane, a naturally abundant K-feldspar mineral. *In-situ* high-resolution X-ray reflectivity is used to determine the sorption capacity and molecular sorption structure of interfacial Y species as a function of Y^{3+} concentration and pH. An inner-sphere (IS) sorption complex in a vertical distance of 1.5 Å from the orthoclase surface is observed, which is suggested to have a bidentate, binuclear binding mode. Additionally, an outer-sphere (OS) sorption complex at a distance of 3–4 Å is found. The total Y coverages of IS and OS species are max. 1.3 Y^{3+}/A_{UC} for all Y^{3+} concentrations, which is in the expected range for the amount needed for surface charge compensation of orthoclase (001).

50 PET and Long-Lived Charge-Separated States Promoted by Intermolecular Interactions Between Copper(I) diimines and Methyl Viologen

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Efficient solar energy conversion into chemical fuels requires careful control over the flow of electrons after photoexcitation. Bi- and multi-molecular systems of chromophores, catalysts and/or electron relays that rely on diffusion to bring discrete components together offer little control over the directionality of electron transfer. Additionally, the chromophores that are suitable for such a system must have excited-state lifetimes longer than the diffusion time scale. Although covalently tethered donors and acceptors can facilitate rapid photoinduced electron transfer (PET), this strategy risks increasing the charge recombination rate. Weaker, self-repairing intermolecular interactions offer an intriguing middle ground between diffusion-based and covalently connected systems and can bring donors and acceptors close for efficient PET while allowing them to diffuse apart afterwards to discourage rapid charge recombination. This work investigated PET from two copper(I) phenanthroline chromophores bearing negatively charged sulfonate groups to the dicationic electron acceptor, methyl viologen, to explore the influence of electrostatic interactions on charge separation and charge recombination in 1:1 acetonitrile:water and in neat water. The results suggest that intermolecular interactions can enhance the efficiency of PET and produce long-lived charge-separated states, and underscore the importance of the solvent environment in determining the magnitude of enhancement and nature of the interactions.

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56 Designing organic Redoxomers for Nonaqueous Redox Flow Batteries

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Redox flow batteries (RFBs) are a promising technology for large-scale electrical energy storage to realize efficient utilization of intermittent renewable energy. Organic redox-active molecules are the active components of nonaqueous-RFBs. Herein, we describe a series of nonaqueous redoxomers which are less sensitive to supporting ions, exhibit enhanced solubility and conductivity, affording improved energy density and cycling performance. These redoxomers show good solubility in acetonitrile (CH_3CN) and demonstrated a surprisingly low reversible redox potential and stable cycling. This study provides guidelines to design organic redoxomers for nonaqueous-RFBs in practical energy storage applications.

DATA SCIENCE AND LEARNING

53 Machine Learning-Based Prediction with Metabolic Models of Bacterial Growth Requirements on Various Substrates

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Despite recent advances, the use of growth phenotype data from diverse biological systems to discover and validate new protein functions continues to be a significant challenge. Current efforts to understand and validate new gene functions suffer from significant barriers: (i) existing experimental data is disorganized, poorly described, and difficult to link to the explicit genome sequences of the species with which the experiments were performed; and (ii) methods for obtaining new data are laborious and scale poorly compared with the amount of sequence data available today. These ‘bottlenecks’ could be broken by adopting new computational methods such as machine learning (ML) combined with mechanistic insights from metabolic modeling. Hence, our goal is to establish steppingstones in gene function discovery and validation by predicting the functions of genes that mechanistically explain observed and predicted growth phenotypes for microbial genomes.

Much microbial growth phenotype data is derived from some variant of a phenotyping array, which permits researchers to rapidly measure the capacity of an isolate to metabolize hundreds of distinct metabolites. These arrays are 96 well plates, where each well has a different fixed media formulation with a distinct carbon, nitrogen, sulfur, or phosphate source. By monitoring OD or respiration-based dye in each well over time, growth rates are measured and reported. To build up a training set of data to use in our efforts to understand, predict, and model microbial growth phenotypes, we have curated data from the literature and performed our own experimental studies to link 178 diverse microbial genome sequences with observed growth data for 64 carbon sources measured using the Biolog phenotype array system.

We applied our gathered training set data to develop machine learning (ML) models to predict binary growth versus no-growth phenotypes for all 64 carbon sources with data; we constructed these ML models using RAST annotated protein functions as features. To compare ML model performance against performance from mechanistic approaches, we similarly constructed metabolic models based on RAST annotations in KBase, and we applied those models to predict the same phenotypes for the same genomes with flux balance analysis. ML models showed higher overall correction predictions (CP), with erroneous predictiosn being evenly balanced between false positives (FP) and false negatives (FN). In contrast, metabolic models had a lower overall accuracy, and displayed fewer FP and far more FN. This result demonstrates the vulnerability of mechanistic models to missing gene annotations (e.g. one missing annotation results in a negative prediction in a metabolic model).

Overall, ML models outperformed metabolic models substantially but lacked the power of metabolic models to mechanistically explain phenotypes and translating phenotype predictions into new protein annotations. However, by combining both approaches, we can use ML to improve models while simultaneously using models to improve protein annotations across all microbial genomes.

ENERGY SYSTEMS AND INFRASTRUCTRE ANALYSIS

51 Investigation of Precooling Unit in Hydrogen Refueling Station for Heavy Duty Vehicles

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Hydrogen fuel has several advantages over the conventional fossil-based fuel including zero emission, zero or trivial carbon footprint, and higher fuel efficiency. It has a potential of being primary source of energy in transportation sector. Furthermore, as compared to battery based electric vehicles, Hydrogen Fuel Cell Electric Vehicles (HFCEVs) have longer driving range, and shorter charging time which encourages the deployment of heavy duty HFCEVs. However, keeping the hydrogen fuel cost at pump within a certain limit is key for early market of HFCEVs. Studies shows the contribution of hydrogen refueling station (HRS) cost to be more than 50% of the cost of hydrogen at pump. Thus, the investigation of HRS cost component relating the heavy-duty HFCEVs is crucial to minimize the cost of hydrogen. In the present work, we investigate the precooling unit (PCU) in a gaseous HRS which has a scope of design optimization and potential reduction in levelized cost of HRS. Due to the large volume of hydrogen fuel dispensed during each fill of heavy-duty vehicle (50 - 100 kg) precooling and refrigeration load is substantial. Thus, we first investigate the precooling temperature required for hydrogen refueling in heavy duty vehicle employing Argonne's H2SCOPE model which is further utilized to design and optimize the PCU. Techno-economic analysis of the PCU is also performed which shows the potential reduction in levelized cost of HRS for heavy duty refueling by > $100,000 by employing the optimum design of PCU.

ENVIRONMENTAL SCIENCE

47 Phasor Networks for Neuromorphic Hardware

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Artificial neural networks (ANNs) are powerful but require many orders of magnitude more energy than biological systems capable of solving similar tasks. One critical difference is that ANN units communicate using continuous signals, as opposed to the binary spike events employed by biological networks. In this work, we extend standard ANN design by building upon an assumption that neuronal activations correspond to the angle of a complex number lying on the unit circle, or 'phasor.' Each layer in such a network produces new activations by taking a weighted superposition of the previous layer's phases and calculating this sum's phase value. This generalized architecture allows models to reach high accuracy and carries the singular advantage that mathematically equivalent versions of the network can be executed with or without regard to a temporal variable. We demonstrate the atemporal training of a phasor network on standard deep learning tasks and show that these networks can then be executed in either the traditional atemporal domain or spiking temporal domain with no conversion step needed. This provides a novel basis for constructing deep networks which operate via temporal, spike-based calculations suitable for low-energy neuromorphic computing hardware.

49 Modeling Deep Eutectic Solvents: Linking Macroscopic Behavior and Molecular Level Features

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Deep Eutectic Solvents (DESs) are a relatively young but extremely useful class of materials. Long considered to be a subset of ionic liquids, they possess a host of useful properties such as low toxicity, cheap materials, high solvent strength, and a complex and diverse design space. Type III DESs, by far the most studied subset, consist of a hydrogen bond donor (HBD) and hydrogen bond acceptor (HBA) mixture, and relatively little is known about how the fundamental structure and interactions of the HBA-HBD pair effect the useful properties of the DES. To this end, molecular dynamics simulations have been utilized in collaboration with a variety of experimental and *ab initio* methods to achieve an understanding of the link between DES structure and dynamics and bulk properties of interest. First, studies on a prototypical DES called Glyceline, a mixture of glycerol and choline chloride, explores how structural and dynamic heterogeneities emerge as a function of choline chloride concentration in glycerol. Second, an examination of structure-property relationships was performed on phenol and *ortho*-phenol derivatives mixed with choline chloride. Despite lacking a hydrogen bonding network, a high degree of structural heterogeneity and unique ion dynamics indicated potential niche uses for these specialized DESs.

MATERIALS SCIENCE

1 Substitutional Vanadium Sulfide Nanodispersed in MoS_2 Film for Pt-scalable Catalyst

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Among transition metal dichalcogenides (TMdCs) as alternatives for Pt-based catalysts, metallic-TMdCs catalysts have highly reactive basal-plane but are unstable. Meanwhile, chemically stable semiconducting-TMdCs show limiting catalytic activity due to their inactive basal-plane. Here, we propose metallic vanadium sulfide (VS_n) nanodispersed in a semiconducting MoS_2 film (V-MoS_2) as an efficient catalyst. During synthesis, vanadium atoms are substituted into hexagonal monolayer MoS_2 to form randomly distributed VS_n units. The V-MoS_2 film on a Cu electrode exhibits Pt-scalable catalytic performance; current density of 1000 mA cm^{-2} at 0.6 V, overpotential of -0.06 V at a current density of 10 mA cm^{-2} and exchange current density of 0.65 mA cm^{-2} at 0 V with excellent cycle stability for hydrogen-evolution-reaction (HER). The high intrinsic HER performance of V-MoS_2 is explained by the efficient electron transfer from the Cu electrode to chalcogen vacancies near vanadium sites with optimal Gibbs free energy (-0.02 eV). This study adds insight into ways to engineer TMdCs at the atomic-level to boost intrinsic catalytic activity for hydrogen evolution.

11 Unveiling the Cerium(III)/(IV) Structures and Charge Transfer Mechanism in Sulfuric Acid

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Redox flow batteries (RFBs) are a promising technology for grid energy storage, but the state-of-the-art RFB (all-vanadium, VRFB) is too expensive, motivating exploration of other chemistries. The Ce^{3+}/Ce^{4+} chemistry is promising because of its higher voltage than the VRFB, but the kinetics are slow, and the charge transfer (CT) mechanism is not understood. Herein, we identify the Ce^{3+} and Ce^{4+} structures and CT mechanism in sulfuric acid via extended X-ray absorption fine structure spectroscopy (EXAFS), kinetic measurements, and density functional theory (DFT) calculations. We show EXAFS evidence that confirms the Ce^{3+} is coordinated by nine waters and suggests Ce^{4+} is complexed by water and three bisulfates in sulfuric acid. Despite the change in complexation within the first coordination shell between Ce^{3+} and Ce^{4+}, we show the kinetics are independent of the electrode, suggesting outer-sphere electron transfer behavior. We identify a two-step mechanism where Ce^{4+} exchanges the bisulfate anions with water in a chemical step followed by a rate-determining electron transfer step that follows Marcus theory. This mechanism is consistent with all experimentally observed structural and kinetic data. We use this CT mechanism to provide guidelines for improving the Ce^{3+}/Ce^{4+} kinetics for RFB applications.

14 Design of naturally occurring organic redox-active materials for nonaqueous redox flow batteries from active learning

Akash Jain

Nonaqueous redox flow batteries (NRFBs) are promising systems for grid-scale energy storage applications. However, the limited energy densities and the lack of suitable redox-active molecules: catholytes, and anolytes dissolved in the electrolyte solution, limit the adoption of NRFBs at a large scale. To achieve higher energy densities in NRFBs, catholyte, and anolyte must display a large redox potential window and high solubility in the electrolyte solution. In this work, we investigate the suitability of inexpensive naturally organic flavonoid molecules for NRFB catholyte and anolyte applications. Specifically, we study flavonoid molecules as scaffolds and decorate them with functional groups to modify their properties and generate about one million new flavonoid molecules. To accelerate the design and discovery of new catholyte and anolyte molecules that display both high redox potential window and high solubility, we employ the active learning (multi-objective Bayesian optimization) algorithm to identify suitable molecules from a library of about one million molecules while using a small number of computationally expensive density functional theory (DFT) calculations. Overall, our work shows that naturally occurring organic flavonoid molecules are promising redox-active materials for NRFBs and with a multi-objective Bayesian optimization method we can design new molecules that exhibit multiple desired properties.

16 Site-Selective Atomic Layer Deposition On Rutile TiO_2:Selective Hydration as a Route to Target Point Defects

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In the field of microfabrication, atomic layer deposition (ALD) is a commonly utilized tool for its precise growth of monolayers on through self-limiting surface reactions. Though ALD became popular several decades ago, the technique has not been significantly extended for more selective growth. Our group previously developed a technique for step selective ALD through a technique called selective hydration where the desorption on temperature on the step was leveraged to control the location of ALD reactants. However, this investigation was limited to only comparing step edges to pristine terrace sites. Here we evaluate the feasibility of this strategy again but with respect to oxygen vacancy and titanium interstitial point defects four rutile TiO_2 facets. First principles simulations were used to evaluate the adsorption free energies for molecular and dissociative adsorption of H_2O on the various substates. We predict that some form of point defect selectivity is possible on all of the four considered facets and was further corroborated by experimental ALD growth profiles.

19 Reactivity of Al and Ga Doped Lithium Garnet (LLZO) at the Li Metal Interface

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All-solid-state lithium-ion batteries (ASSB) are a critical technology for the future development and expansion of lightweight, energy dense, and safer battery technologies. Of the potential solid electrolyte materials, the lithium garnet, $Li_7La_3Zr_2O_{12}$ (LLZO) has been touted as a leading candidate due to its high ionic conductivity (10^{-3} S/cm), toughness (shear moduli ~60GPa) and purportedly strong electrical and chemical stability (0.02-4.0 V vs Li/Li^+). However, progress in advancing the deployment of ASSB has been slow due to the difficulty of engineering a stable solid-solid interface between the solid electrolyte and lithium metal. One possible explanation for this is the formation of Li-Ga eutectics that enhance the interfacial contact between the Li metal and solid electrolyte. In this study we look to investigate the stability of aluminum and gallium dopants at the $LLZO|Li^o$ interface through a combined effort of theoretical modeling and experimental techniques including, XPS, impedance spectroscopy, neutron diffraction.

22 Vibrational and Optical Spectral Characterization of Sequential Infiltration Synthesis Derived Indium Oxyhydroxide Clusters for CO_2 Capture

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Sequential infiltration synthesis (SIS) is a versatile route to hybrid organic-inorganic materials as well as purely inorganic materials templated by polymeric structures. While SIS is inspired by atomic layer deposition (ALD) and often utilizes the same precursors and tools, SIS depends upon infiltration of these vapor phase precursors into a polymer film or bulk with reactive functional groups acting as nucleation sites. Careful tuning of SIS conditions including temperature, precursor exposure, and the number of SIS cycles can potentially enable a range of inorganic species ranging from single metal atom sites to few atom clusters, to a contiguous inorganic network. In this work we probe the evolution of a select few $InO_x(OH)_y$ cluster within a PMMA matrix prepared via SIS at 80 °C via in-situ FTIR and UV/vis spectroscopy. We demonstrated that these clusters have unique molecular structure resembling small units of the crystal structure of cubic In_2O_3. Lastly, the $InO_x(OH)_y$ modified PMMA membranes were also characterized for applications towards CO_2 capture and conversion. We demonstrate that the $InO_x(OH)_y$ clusters enhance CO_2 uptake especially at low pressures compared to pristine PMMA. The mode of CO_2 adsorption was also probed via in-situ DRIFTS which reveals the complexity of the CO_2 adsorption modes in the studied materials.

24 Magnetic Breakdown and Topology in the Kagome Superconductor CsV_3Sb_5 under High Magnetic Field

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The recently discovered layered Kagome metals of composition AV_3Sb_5 (A = K, Rb, Cs) exhibit a complex interplay among superconductivity, charge density wave order, topologically non-trivial electronic band structure and geometrical frustration. Here, we probe the electronic band structure underlying these exotic correlated electronic states in CsV_3Sb_5 with quantum oscillation measurements in pulsed fields up to 86 T. The high-field data reveal a sequence of magnetic breakdown orbits that allows the construction of a model for the folded Fermi surface of CsV_3Sb_5. The dominant features are large triangular Fermi surface sheets that cover almost half of the folded Brillouin zone that have not yet been detected in angle resolved photoemission spectroscopy (ARPES). These sheets display pronounced nesting at the charge density wave (CDW) vectors, which may stabilize the CDW state. The Berry phases of the electron orbits have been deduced from Landau level fan diagrams near the quantum limit without the need for extrapolations, thereby unambiguously establishing the non-trivial topological character of several electron bands in this Kagome lattice superconductor.

27 What Affects the Lithium Electrode-Electrolyte Interface in Composite Polymer Electrolytes?

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Composite polymer electrolytes (CPEs) are attractive solid-state electrolytes for lithium metal batteries. However, they frequently have high interfacial resistances against lithium metal, which limits their fast-charging abilities and can lead to dendrite formation. Despite this challenge, it is still unclear how the addition of the ceramic component to the polymer phase in CPEs affects the lithium-electrolyte interface. Here, we detail the formation of the solid electrolyte interphase (SEI) with and without Li^+-conducting $Li_7La_3Zr_2O_{12}$ (LLZO) nanofibers in poly(ethylene oxide)-lithium bis(trifluoromethanesulfonyl)imide (PEO-LiTFSI) electrolytes. X-ray photoelectron spectroscopy, cyclic voltammetry, and electrochemical impedance analysis show that SEI formation is totally dependent on PEO-LiTFSI when formed chemically or electrochemically. LLZO plays no part in SEI formation and is not present at the electrode interface, despite electrolytes with LLZO exhibiting higher critical current densities. Mesoscale modelling demonstrates that the microstructure of the LLZO nanofibers alone slows dendrite growth velocity, highlighting a new mechanism for dendrite prevention in CPEs. Our work offers several design rules for optimizing CPE interfaces for practical lithium metal batteries.

28 Quantum Critical Behavior And Collective Modes Near The Superfluid-Mott Glass Transition

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Quantum phase transitions (QPTs) between ground states of interacting many-body systems have been a defining topic of modern condensed matter physics. While much is known about the physics of QPTs in pure systems, the effects of disorder (impurities, defects, etc.) on QPTs is still not completely understood. Moreover, the nature of the collective excitations near the phase boundaries of disordered QPTs requires further study. In this study, we investigate the effects of disorder on both the quantum critical behavior and fundamental collective excitations near the superfluid-Mott glass QPT. We consider a Bose-Hubbard model of disordered interacting bosons in d-dimensions which we map onto a thermodynamically equivalent (d+1)-dimensional classical XY model and simulate via large-scale Monte Carlo techniques. We determine the phase diagram of the system for a range of dilution strengths, calculate the disordered critical exponents, and show that the thermodynamics of the disordered system are of conventional power-law type. Interestingly, upon calculation of the spectral densities of the order parameter fluctuations for the disordered system, we see that the Higgs mode may become localized, and the Goldstone mode undergoes a striking localization-delocalization transition, showing that disordered systems may exhibit unconventional dynamical behavior despite having conventional underlying thermodynamic behavior.

32 Sequence-Controlled Secondary Structures and Stimuli-Responsiveness of Bioinspired Polyampholytes

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A comprehensive study focusing on the influence of the sequence charge pattern on the secondary structure preferences of annealed polyampholytes and their responsiveness to external stimuli is presented. Two sequences are designed composed entirely of ionizable amino acids (charge fraction, $f=1$), and an equal number of positive and negative charges ($f_+=f_-=0.5$) with distinct charge patterns consisting of lysine and glutamic acid monomers. The study reveals that the sequence charge pattern has a significant influence on the secondary structure preferences of polyampholytes at physiological pH. Furthermore, it shows that external stimuli such as pH, ionic strength and solvent dielectric constant can be used to modulate the secondary structure of the two studied sequences. The observed secondary structure transformations for the two sequences are also substantially different from those determined for uniformly charged homo-polypeptides under matching conditions.

MATHEMATICS & COMPUTER SCIENCE

3 Spiking Neural Network-based Continual Learning for Energy-Efficient Neuromorphic Hardware at the Edge

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Neuromorphic computing (NmC) based on spiking neural networks (SNNs), a computing paradigm inspired by the biological concepts of the mammalian brain, provides a number of advantages for data-, energy-, and resource-efficient machine learning at the edge. Hardware implementations of NmC, executing SNNs, enable large-scale data analysis beyond what is feasible with the emerging high-performance-computing-to-edge computing paradigm. This key advantage can be attributed to the low-power design of the underlying computing circuits, the distributed implementation of its compute and storage, and novel technology integration in the form of non-volatile memory-based neuro-synaptic cores. In order to deploy the NmC capabilities at the APS, however, SNNs need to continually adapt to variations in the data and the NmC hardware. Existing state-of-the-art SNN-based continual learning (SCL) algorithms are not tolerant to process, operational, and reliability variations in NmC hardware. To overcome these challenges, we propose an SCL framework to train data-, energy-efficient, and hardware-fault-tolerant SNN-based models on NmC hardware. The proposed SCL algorithm will allow SNN-based models to learn new tasks and quickly adapt to changing data and hardware variations. The proposed research will provide unique capabilities to deploy continual-learning-capable SNNs at the edge, which can potentially transform large-scale data analysis at the APS.

10 Automating Performance Measurements on Supercomputers

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Supercomputers allow us to undertake big computational and data-intensive workloads such as training artificial intelligence (AI) models to predict cancer diagnosis. These systems and workloads must be carefully configured to minimize system bottlenecks and ensure good performance. However, the software tools used to evaluate configurations provide limited views of the workload or system under investigation.

To improve the efficiency of performance analysis for AI workloads, we need to understand the 'blind spots' of current tools and the ability to prevent these ‘blind spots’ by easily combing different tools.

We work towards developing a framework that automates performance measurement using multiple software tools on Argonne supercomputers. We build on ReFrame, a framework for automating tests on large systems, by adding support for the performance tools used to study complex AI workloads. We catalog some of the major limitations of popular tools and demonstrate how our framework use them to enable more comprehensive performance analysis.

15 Workflow Anomaly Detection with Graph Neural Networks

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Reliable execution of scientific workflows is a fundamental concern in computational campaigns. Therefore, detecting and diagnosing anomalies are both important and challenging for workflow executions that span complex, distributed computing infrastructures. In this paper we model the scientific workflow as a directed acyclic graph and apply graph neural networks (GNNs) to identify the anomalies at both the workflow and individual job levels. In addition, we generalize our GNN model to consider a set of workflows together for the anomaly detection task rather than a specific workflow. By taking advantage of learning the hidden representation, not only from the job features, but also from the topological information of the workflow, our GNN models demonstrate higher accuracy and better runtime efficiency when compared with conventional machine learning models and other convolutional neural network approaches.

20 Thin Film Particle Detectors for High Energy Physics Applications

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Current models for spatial extremes are concerned with the joint upper (or lower) tail of the distribution at two or more locations. Such models cannot account for teleconnection patterns of two-meter surface air temperature (T2m) in North America, where very low temperatures in the contiguous Unites States (CONUS) may coincide with very high temperatures in Alaska in the wintertime. This dependence between warm and cold extremes motivates the need for a model with opposite-tail dependence in spatial extremes. This work develops a statistical modeling framework which has flexible behavior in all four pairings of high and low extremes at pairs of locations. In particular, we use a mixture of rotations of common Archimedean copulas to capture various combinations of four-corner tail dependence. We study teleconnected T2m extremes using ERA5 reanalysis of daily average two-meter temperature during the boreal winter. The estimated mixture model quantifies the strength of opposite-tail dependence between warm temperatures in Alaska and cold temperatures in the midlatitudes of North America, as well as the reverse pattern. These dependence patterns are shown to correspond to blocked and zonal patterns of mid-tropospheric flow. This analysis extends the classical notion of correlation-based teleconnections to considering dependence in higher quantiles.

21 Robust Real-Time Digital Twin Models from Experimental Data

Nikita Kuklev

Various machine learning tools have been proposed for accelerator optimization, anomaly detection, and other purposes. Due to limited beam time, majority of their testing and debugging is performed on simulated data. In some machines, simulations have poor agreement with experimental results. We explore development of digital twin environments and associated surrogate models purely from or augmented by experimental data, which can then be used to improve experimental ML tool performance. Our main contribution are methods to compensate for deficiencies in experimental data — limited parameter space, inefficient and sparse sampling, noise, and imbalance in terms of parameter and objective distributions. We propose and benchmark three such methods - label distribution smoothing, Bayesian optimal sampling strategies, and physics-informed ensemble modelling. We also describe a distributed hyperparameter tuning strategy to pick best architectures for specific systems in real-time. Our results show significant improvement in accuracy as compared to standard surrogate models, with little to no performance degradation.

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31 Deploying Multiphysics Computational Fluid Dynamics Simulations to the Cloud

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Open source multiphysics scientific software instruments like Flash-X provide robust infrastructure to parallelize and scale complex Computational Fluid Dynamics (CFD) problems on heterogeneous architecture of modern computing systems. In comparison to commercially available software products like Ansys and COMSOL, which restrict user interaction with underlying source code, an open-source design allows for user customization and creativity when designing complex simulations for practical engineering problems. The appeal of open source for commercial use has been limited due to the complexity introduced by dependency on external libraries and the corresponding software environment required to build and run simulations. Recent developments in containerization and cloud computing technology, however, present an avenue for effectively using open-source tools for commercial problems. We will present how one can leverage Maple, a wrapper over existing containerization services like Singularity and Docker to seamlessley deploy high-fidelity multiphysics CFD simulations using Flash-X within a containerized environment. We will discuss performance metrics when running this type of containerized service, and its applicability to more advanced problems of integrating machine learning with simulations.

34 Stochastic Trust-Region Algorithm In Random Subspaces

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This work proposes a framework for large-scale stochastic derivative-free optimization (DFO) by introducing STARS, a trust-region method which achieves scalability using random models in random low-dimensional affine subspaces. STARS significantly reduces per-iteration costs in terms of function evaluations, thus yielding strong performance on large-scale stochastic DFO problems. The user-determined dimension of these subspaces can be chosen via so-called Johnson--Lindenstrauss transforms, and independently of the dimension of the problem. For convergence purposes, both a particular quality of the subspace and the accuracies of random function estimates and models are required to hold with sufficiently high, but fixed, probabilities. Convergence and expected complexity results of STARS are obtained using martingale theory.

57 LibPressio: A Unifying Data Compression Interface for Users and Developers

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Scientists have lots of data that they need to store, transport, and use. Lossy compression could be the solution, but there are 32+ compressors, each with its own interface and the interfaces of the most recent compressors often evolve. Moreover, compressors are missing key features: provenance and configuration parameter optimization. LibPressio addresses all these issues by providing a unifying interface with advanced engines for provenance and configuration optimization. This poster presents a variety of research accomplishments using this tool and future research directions including: ROIBIN-SZ a state of the art lossy compressor able to reduce massive serial crystallography datasets by 196x at high rates. OptZConfig a tool to configure error bounded lossy compressors to preserve user specified error bounds which out performs prior blackbox approaches, specialized compressors, and compressors that preserve bounded linear functionals while supporting many new classes of metrics including those relevant to the climate community. Manifest is a tool which preserves the provenance of compression operations. Lastly we future work in in-line transparent compression for stencils and numeric solvers.

ARGONNE LEADERSHIP COMPUTING FACILITY

4 In Situ Machine Learning for Exascale CFD

Riccardo Balin[1], Filippo Simini[1], Ramesh Balakrishnan[2] and Venkat Vishwanath[1]

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Large eddy simulation (LES), which requires modeling of the sub-grid stress (SGS) tensor, can offer a compromise between accuracy and efficiency of numerical computations of turbulence flows. Data-driven approaches, such as neural networks (NN), have recently emerged and present encouraging results for improved predictive capacity over traditional models. However, since NN for LES closures must be trained on instantaneous high-fidelity turbulent data, learning from high Reynolds number and complex flows requires multi-terabyte databases to store the training data. This limitation is resolved by performing online (in situ) learning, wherein the NN model is trained concurrently with the flow simulation producing the data and thus eliminating the need to store large training datasets on disk. This talk will cover the software infrastructure developed to perform online learning and inference on current and future supercomputers and its performance at increasingly larger scale.

8 Data-Driven Modeling of Compressible Reacting Flow Using Hardware-Oriented Algorithms

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High-fidelity numerical simulations of combustion processes in next-generation hypersonic propulsion devices (including, but not limited to, rotating detonation engines and scramjets) play a crucial role in enabling robust design strategies for real-world deployment. These simulations, however, require full-geometry numerical solutions of the compressible reacting Navier-Stokes equations. Spatiotemporal resolution requirements stemming from multi-scale interactions between turbulence, shockwaves, and chemical reactions contained in these governing equations induce computationally prohibitive bottlenecks that render the required long-time resolved simulations of these propulsion devices infeasible. A particularly elusive bottleneck comes from the treatment of detailed chemical kinetics required to accurately describe the time evolution of species concentrations and flow-chemistry interactions. The goal of this work is to provide a physics-guided data-driven modeling strategy based on a physics-guided clustering algorithm for accelerating high-fidelity compressible reacting flow solvers via elimination of the chemistry bottleneck. Emphasis is placed on (a) ensuring the modeling framework can be extended to in-situ (or online) integration with flow solvers, such that the method is not tied down to single geometric configurations, and (b) ensuring that the algorithms used are compatible with modern high-performance computing trends dominated by GPU-centric node architectures.

NANOSCIENCE AND TECHNOLOGY

23 Exploring Kinetic Pathways for Materials Synthesis using Evolutionary Reinforcement Learning

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Synthesis of materials with tailored properties is an overarching goal of material scientists. Equilibrium phase diagrams are often a reasonable starting point for designing synthesis protocols, but in typical experimental settings, kinetic effects are often more prominent and therefore determine the feasibility of such protocols. Selective synthesis of polymorphs of a material can also be limited by such kinetic barriers. Here, using molecular dynamics simulations we investigate how neural networks trained by evolutionary reinforcement learning can generate feasible synthesis pathways for materials. These previously unknown pathways can enable us to optimally synthesize a particular material and simultaneously provide physical insight into the kinetics of phase transitions. Transformation pathways in materials, such as Carbon, are discussed here. The presented evolutionary scheme can be applied to investigate a broad range of problems (such as self-assembly, experimental design, and additive manufacturing) wherein transition pathways from one state to another are unknown.

Schwenker, E. *et al.* “Ingrained: An Automated Framework for Fusing Atomic-Scale Image Simulations into Experiments”, *Small* 18, 19 (2022)

40 Optical Refrigeration on CdSe/CdS (Core/Shell) Quantum Dots

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Optical refrigeration (OR) in solids was proposed for the first time by the German physicist Peter Pringsheim in 1929 [1], where vibrational energy can be removed from a substance by spontaneous energy up-conversion and emission of absorbed light. In 2012, Zhang and his coworkers successfully cooled a CdS nanobelt by 40 K [2]. Their work revealed the possibility of realizing OR in semiconducting nanomaterials, triggering a frenzy of research in this field. In our previous research, we have showed that net energy up-conversion in photoluminescence on surface passivated CdSe/CdS QDs is achievable with laser excitation energy much lower (>80 meV) than the band gap [3]. Based on this observation, optical refrigeration on CdSe/CdS QDs was carried out and a cooling effect of 0.68±0.07 K was suggested by the experimental data. This development paves the way to use QDs' cooling in new industrial and fundamental research approaches.

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[2] J. Zhang, D. Li, R. Chen and Q. Xiong, "Laser cooling of a semiconductor by 40 kelvin," *Nature Letter,* vol. 493, pp. 504-508, 2012.
[3] M. Hua and R. S. Decca, "Net energy up-conversion processes in CdSe/CdS (core/shell) quantum dots: A possible pathway towards optical cooling," *Phys. Rev. B,* vol. 106, no. 8, p. 085421, 2022.

43 Operando XAS Studies of High-Entropy Oxides in Li-ion Batteries

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Today’s energy storage demands require a combination of high energy and power density for an increasing variety of applications. As a result, conversion type materials, specifically transition metal oxides (TMOs) with the capacity for multi-electron redox processes, are of interest as a solution for the relatively limited Li uptake imposed by intercalation anodes. Previous studies on high-entropy oxides (HEO) as electrodes in LIBs reported high capacities and good cycling stability. This promising electrochemical performance was first demonstrated for the (MgCoNiCuZn)O compound having the rocksalt structure. Preliminary electrochemical data demonstrates the ability to change the electrode's storage capacity, cycling stability, and working potential, depending on the elements present in the HEO. Given the heterogeneous and transient nature of the conversion, in situ EXAFS was performed to capture the operando short-lived reactions and non-equilibrium environment, which significantly contribute to the reversible charging states. The results show different degrees of activity for the metals depending on the system studied, and their role in the overall storage capacity. Herein, we discuss the electrochemical reaction mechanism of these compounds based on the Co, Ni, Cu, and Zn K edges of the HEO-family compounds and relate the electrode’s performance with the operando EXAFS and XANES.

48 Ingrained: Automated Tool for Merging Atomic-Scale Theory and Experiment

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Though scanning tunneling microscopy (STM) can provide a great deal of information about the properties of a surface, the exact structure cannot be determined strictly through imaging. As STM measures the charge density at a specific energy level relative to the Fermi level of a surface, the exact atomic structure is not directly measured. To address this limitation, density functional theory (DFT) calculations are combined with the software package *Ingrained*[1] to fit a theoretical STM simulation to an experimental image. A variety of tunable parameters, including the distance of the tip from the surface, smearing, strain, and energy range, are optimized to match an experimental STM image to the theoretical image generated using DFT. We use two systems, Cu2O (110) and Sn-bilayer on a Si wafer, as examples of successful applications of this tool, providing a foundation for investigating CO_2 reduction and temperature-driven phase change, respectively.

58 Fully Automated Nanoscale to Atomistic Structure from Theory and Experiments

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Prior to computational investigation into the structural and electronic properties of materials, precise knowledge of their atomistic structure is required. Various experimental spectroscopic techniques are commonly used to probe and characterize the structure of a material, but unless the material has been previously studied and reference data can be matched against, inverting spectra to fully determine the atomistic structure is a significant challenge. Theoretical insight requires searching a vast structural space where it is critical to not only match the experimental spectroscopic data but also minimize other quantities such as the energy to ensure that the structures are both physically plausible and realizable. In response, we have developed the FANTASTX (Fully Automated Nanoscale to Atomistic Structure from Theory and eXperiment) code, a multi-objective evolutionary algorithm which performs machine-learning informed data-driven structure search using genetic algorithm and basin hopping methods. It includes full support for transmission electron microscopy (TEM), X-ray diffraction (XRD), pair distribution function (PDF) and x-ray absorption spectroscopy (XAS) analysis. The FANTASTX code has demonstrated significant speed-ups in the inversion of spectra which previously required manual inversion and we have also produced novel insights into the structures of transition metal catalysts, blue-layer amorphous IrO_2 and gold nanoclusters.

59 Machine Learning of Borophene-Borides Interaction

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Borophene, two-dimensional (2D) boron, has novel properties and its metallicity is unusual within the 2D materials family and enriches the diversity of available materials properties. However, a big challenge in this field is the achievement of freestanding borophene. It is difficult to lift borophene from the substrates due to the strong interaction between them. Thus it is important to search for optimal substrates for borophene synthesis and separation. Metal borides have the potential to be superior substrates, compared to metals, for borophene synthesis and separation. The understanding of the interaction in borophene-boride hetero-structures would assist the search of optimal substrates as well as reveal the synthesis mechanism. What borides surfaces are optimal substrates for borophene synthesis? What intrinsic properties of borides dominantly affect the interaction in borophene-boride hetero-structures? How does the charge transfer between borophene and borides affect the interaction? Here we explore these questions through machine learning (ML) and density functional theory (DFT) calculations of their structures, binding energies, and charge transfer. This work allows us to explore alternative routes of borophene synthesis.

PHYSICS & HIGH ENERGY PHYSICS

17 Design of a Barrel Imaging Electromagnetic Calorimeter for the Electron-Ion Collider

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The Electron-Ion Collider (EIC) will be an experimental facility to explore the gluons in nucleons and nuclei, shedding light on their structure and the interactions within. Among the detector concepts driven by the EIC community, the ANL group proposes an Imaging calorimeter in the central barrel region. It is a hybrid design utilizing Imaging calorimetry based on monolithic silicon sensors and scintillating fibers embedded in lead. The barrel calorimeter aims to fulfill all the requirements posed by the rich EIC science program, providing precise measurements of energy and position, and an excellent PID between pion and electrons. In particular, the barrel electromagnetic calorimeter must also detect energy and position of neutral particles such as photons and identify single photons originating from Deeply Virtual Compton Scattering (DVCS) process and photon pairs from π_0 decays. In this work, I will present the expected performance of photon and neutral pion reconstruction based on simulations of the electromagnetic calorimeter in the central region for the ATHENA proto-collaboration.

18 Thin Film Particle Detectors for High Energy Physics Applications

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Silicon tracking detectors have grown to cover larger surface areas up to hundreds of square meters, and are even taking over other sub-detectors, such as calorimeters. However, further improvements in tracking detector performance are more likely to arise from the ability to make a low mass detector comprised of a high ratio of active sensor to inactive materials, where dead materials include electrical services, cooling, mechanical supports, etc. In addition, the cost and time to build these detectors is currently large. Therefore, advancements in the fundamental technology of tracking detectors may need to look at a more transformative approach that enables extremely large area coverage with minimal dead material and is easier and faster to build. The advancement of thin film fabrication techniques has the potential to revolutionize the next-to-next generation of particle detector experiments. Some thin film deposition techniques have already been developed and widely used in the industry to make LED screens for TV's and monitors. If large area thin film detectors on the order of several square meters can be fabricated with similar performance as current silicon technologies, they could be used in future particle physics experiments. This paper aims to review the key fundamental performance criteria of existing silicon detectors and past research to use thin films and other semi-conductor materials as particle detectors in order to explore the important considerations and challenges to pursue thin film detectors.

29 Search for new phenomena in multi-body invariant masses in events with at least one isolated lepton and two jets using $\sqrt{s}$ = 13 TeV proton–proton collision data collected by the ATLAS detector

Smita Darmora

A search for resonances in events with at least one isolated lepton (e or μ) and two jets is performed using 139 $fb-1$ of $\sqrt{s}$ = 13 TeV proton–proton collision data recorded by the ATLAS detector at the LHC. Deviations from a smoothly falling background hypothesis are tested in three- and four- body invariant mass distributions constructed from leptons and jets, including jets identified as originating from bottom quarks. Model-independent limits on generic resonances characterised by cascade decays of particles leading to multiple jets and leptons in the final state are presented. The limits are calculated using Gaussian shapes with different widths for the invariant masses. The multi-body invariant masses are then used to set 95% confidence level upper limits on the cross- section times branching ratios for the production and subsequent decay of resonances as predicted by several new physics scenarios.

46 Measurement of (α) Reactions and Development of Analysis Tools with the MAJORANA DEMONSTRATOR Experiment

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Neutrinoless double beta decay ($0\nu\beta\beta$) is a hypothesized nuclear transition which, if observed, would unambiguously demonstrate the violation of an observed symmetry of the Standard Model (SM) and establish the Majorana nature of neutrinos. The MAJORANA DEMONSTRATOR experiment searches such decay in ^{76}Ge using P-type point contact high purity germanium detectors. The energy resolution of the detectors, understanding of various possible backgrounds, and their suppression techniques are essential factors in increasing the discovery sensitivity. The DEMONSTRATOR has achieved the best energy resolution among current generation $0\nu\beta\beta$ experiments and one of the lowest background indexes. In this talk, I will explain my efforts in energy calibration, understanding radiogenic neutron background, and developing a machine learning approach for background suppression for the MAJORANA DEMONSTRATOR experiment.

POWER SYSTEMS

9 Data-driven Dynamic Surrogate Modeling of Thermo-physical Systems

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Thermo-physical systems are dynamical processes that have complex multi-scale physics. Being able to model them is an important first step in being able to study the system response at a variety of operational points, and thus design more efficient systems. Reduced-order models are fast but do away with the complex physics, while full-scale numerical simulations or experiments are more accurate but expensive. Data-driven approaches enable fast and accurate dynamical predictions in applications where physics-based models are either absent or are too complex to be used efficiently for analysis and control. This poster presents a deep state-space modeling (SSM) framework that combines the feature extraction capabilities of convolutional neural networks (CNNs) with the efficient sequence prediction properties of gated recurrent units (GRUs); the CNN-GRU SSM. This approach is validated on several public datasets and demonstrated on predicting multi-input, multi-output, coupled, and nonlinear heat-exchanger dynamics observed in vapor compression cycles [1]. This can then be augmented with Gaussian process (GP) based multi-fidelity modeling to learn from all available data from sources of different fidelities. Leveraging a GPyTorch based GP model lends immense flexibility since it can be seamlessly melded with PyTorch based deep learning modules, thus easily making a general-purpose dynamical surrogate learner.

[1] C. Bhattacharya, A. Chakrabarty, C. Laughman, H. Qiao, "Modeling nonlinear heat exchanger dynamics with convolutional recurrent networks", MECC 2022, Jersey City, USA, 2022

26 CFD Simulations of Combustion with High Hydrogen Blends for a Microturbine Combustor

Joshua Christopher[1], Chandrachur Bhattacharya[1], Munidhar Biruduganti[1], Debolina Dasgupta[1], and Sibendu Som[1]

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Combined heat and power (CHP) gas turbines provide flexible energy generation that can be used to stabilize a variable renewable power grid, and microturbines are well suited for use in CHP with a variety of commercial and industrial applications. However, gas turbines have traditionally been powered by carbon-based fuels such as natural gas. Hydrogen provides a path towards decarbonization for the power generation sector and has seen increasing use in microturbines. Hydrogen fuel blends in microturbines face several challenges, including flame flashback and auto-ignition upstream of the combustion chamber and higher NO_x emissions compared to hydrocarbons due to the hotter burning flame. To facilitate the integration of CHP microturbines into the power grid, the complex multi-scale multi-physics flow in the combustor with high hydrogen fuel blends is studied across the operability map. The finite volume computational fluid dynamics (CFD) code CONVERGE is used to simulate a 65kW microturbine combustor across a range of input parameters, including fuel blend and power loading. This work presents the model development and resulting flow paths, flame shapes, NO_x emissions, unburnt hydrocarbons, and combustion efficiencies found across the operability map.

54 Facilitating a move towards sustainable aviation using Aeronomie: a dynamic simulation tool.

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In the aerospace industry, electric propulsion is triggering substantial shifts in aspects of aircraft design, production, and operations, leading to large-scale innovation not seen since the introduction of jet engines. A strong political and societal push to dramatically cut gaseous emissions deriving from air transportation is currently undergoing to avoid making aviation the black sheep in the race to zero-emission modes of transport.

Apart from the push towards *sustainability,* there are several advantages and opportunities brought in by electric-powered aviation, such as increased safety, reduced noise, gaseous emissions, and increased take-off and landing performance due to aero-propulsive interaction.

This work aims at contributing to the research effort towards more sustainable aviation, within the Vehicle & Mobility Systems Group of ANL. Our first goal would be to model a small commuter aircraft, making use of Aeronomie, the aircraft version of the well-established Autonomie vehicle system simulation tool. A 9-seater aircraft will be modeled and adopted as a theoretical testbed for the introduction of a variety of innovative powertrain architecture like electric, hybrid-electric, and hydrogen-powered. These models will then be employed to assess energy consumption, gaseous emissions, performance, and costs, eventually expanding the study to fleet level impacts of such aircraft.

60 Numerical Modeling of Fuel-Air Mixing in a Direct Injection Hydrogen Engine

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Hydrogen as an energy carrier has recently become a promising option for advanced energy and transportation systems with the goal to achieve effective decarbonization. Within the framework of internal combustion engines (ICE), direct injection (DI) of gaseous hydrogen during the compression stroke offers great potential to avoid abnormal combustion behaviors, as opposed to port fuel injection. To advance physical understanding, this study focuses on the CFD modeling of hydrogen DI process in a light-duty hydrogen optical engine designed by Sandia National Laboratories. Gaseous hydrogen was injected into the cylinder via a centrally located single-hole injector with different injection pressures. Two configurations, namely low- and high-tumble, are investigated to understand the impact of in-cylinder flow patterns on fuel-air mixture preparation. Numerical simulations are carried out with CONVERGE v3.0. The in-cylinder turbulence is modeled with an unsteady Reynolds-averaged Navier-Stokes (URANS) formulation closed by the renormalization group (RNG) k-ε model. Several sub-models and model constants are evaluated over a wide range of injection pressures and timing. The numerical results are systematically compared against experimental measurements of velocity and mixing fields to assess and understand the performance of the CFD model and to establish best practices for accelerating numerical design of novel hydrogen engines.

61 LES of Turbulent Flow in Gas Turbine Combustor using Nek5000

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Understanding the behaviors of the fuel and combustion within gas turbine combustors is essential to the stable operation of engines. The flame stability, structure and shape during engine operation may be significantly affected by the fuel properties. The combustors that operate at very lean conditions tend to have undesired events, such as the lean blow out (LBO) which occurs due to the flame instability and may lead to the degradation of engine performance. In this study, wall-resolved large-eddy simulations (WRLES) of the turbulent flow in the Army Research Laboratory's midsize combustor (ARC-M1) were performed using Nek5000. The statistical behaviors within the M1 combustor are first validated against available experimental data while the turbulent structures are further explored using proper orthogonal decomposition (POD) to advance flow modeling for engineering simulations.

X-RAY SCIENCE

5 Imaging of Three-Dimensional Magnetic Structures using Hard X-rays

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Imaging of three-dimensional magnetic structures of materials at nanoscale is critical for several applications such as identifying properties of functional and quantum materials. In order to unravel the heterogeneity of magnetization of materials in the nano-world, polarization dependent resonant X-ray spectroscopy related methods are crucial. The Velociprobe instrument in beamline 2-ID-D at the Advanced Photon Source is used to identify the internal 3D magnetization. Circularly polarized hard X-rays at photon energies corresponding to the L_2 absorption edge of Nd is incident on a cylindrical shaped $Nd_2Fe_{14}B$ single crystal for varying projection angles in a dichroic nano ptycho-tomographic setup. Jitter and misalignment of the rotation axis caused by vibration or drift in the experimental setup leads to misalignment between reconstructed ptychographic images for different tomographic projections or between the two circular polarization states and needs to be corrected. In order to address this issue, image registration and alignment is first done on these projection images. Subsequently, tomographic reconstruction is carried out using the difference of the images for different polarizations which then provides magnetization components in 2D. With projection data at 2 different sample tilts, the full 3D magnetization vector field is reconstructed without any prior knowledge or assumptions.

35 Investigating Structural Transitions in Aromatic Polymers up to 32 GPa with a Double Stage Compression Technique and Pair Distribution Functions

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Aromatic polymers include materials such as polyurea, an amorphous elastomer capable of dissipating large amounts of energy under dynamic loading. Although computational research exists that investigates the atomic-level response of polyurea and other amorphous aromatic polymers to extreme conditions, there is little experimental work to validate these models 1) at the atomic-scale and 2) under high pressures characteristic of extreme dynamic loading. As many such polymers undergo pressure and temperature-induced structural transformations, understanding their structure-property relationships at the atomic-level is key to formulate accurate predictive models.

This work investigates the high-pressure structural response of aromatic polymers at the atomic-level with pair distribution functions. Structural data was obtained *in situ* via multiangle energy dispersive X-ray diffraction (EDXD) experiments at the Advanced Photon Source for polyurea as well as polysulfone, an additional aromatic polymer. As high pressures ($\lesssim\lesssim$ 32 GPa) were applied using a double stage compression technique, indications of structural transformations were observed that appear similar to the sp^2-sp^3 hybridization in compressed carbon. The changes are largely reversible, consistent with pressure-driven, reversible graphite-diamond transformations in the absence of applied temperature. These results constitute some of the first *in situ* observations of the mechanisms that drive pressure-induced structural transformations in aromatic polymers.

44 Coherent Surface Scattering Imaging

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Coherent Surface Scattering Imaging (CSSI) brings together conventional imaging techniques, such as lensless X-ray Coherent Diffraction Imaging (CDI), transmission-geometry ptychography and laminography, and a surface sensitive technique of Grazing Incidence Small Angle X-ray Scattering (GISAXS). CSSI will be the feature technique of the new 9-ID beamline of the Advanced Photon Source Upgrade. For image reconstruction purposes, we need a physical model that can reproduce complex GISAXS scattering patterns, such as dynamical scattering fringes that are observed near sample horizon. Distorted Wave Born Approximation (DWBA) cannot reproduce such dynamical fringes. On the other hand, multislice simulations compute wave propagations through objects one slice at a time along the beam direction and are already widely used in transmission geometry electron microscopy and X-ray transmission experiments. The multislice formalism could also be applied to CSSI reflection-geometry setups and can successfully reproduce dynamical scattering phenomena near critical angles. This multilice formalism has been implemented in fast performing GPU codes which can do forward calculations in a few seconds. Here, it is discussed how backward propagation of the model and Pytorch auto-differentiation tool enable us to do image reconstructions in the form of CSSI-CDI on experimental data and simulations.

52 Non-Invasive Temperature Probing In Multi-Species Liquid Sprays

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Multiphase flows dominate many different areas of life, including the medical, agricultural, propulsion, and chemical industries. Gaining insight into the dynamic processes that drive these multiphase flows can therefore have far-reaching impact in many sectors of scientific research. Of key interest is the non-invasive tracking of important state properties such as the mass and temperature distributions in high optical depth multiphase flows. To accomplish this, X-ray diagnostic approaches are utilized due to their ability to probe complex phenomena without being hampered by multiple scattering that arise from complex interactions at the flow surface boundaries. Efforts in ascertaining the temperature distributions in liquid/gas flows is done through an application of wide angle X-ray scattering, a technique that is commonly used in the materials, chemistry, and biology sciences but has yet to be widely used in the propulsion community. These newly developed X-ray scattering measurements are accomplished through the use of a focused monochromatic beam available at the Advanced Photon Source synchrotron facility, and is applied first in calibration jets and later towards more complex dynamic sprays and multi-species liquid solutions.

62 Imaging Optical Wavelength-Dependent Nanostructure Creation in PbTiO3/SrTiO3 Ferroelectric Superlattices

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$(PbTiO_3)_n/(SrTiO_3)_n$ (PTO/STO) superlattices on a $DyScO_3$ (DSO) $(001)_{pc}$ substrate (where pc refers to the pseudocubic notation, and n represents the number of unit cell) host novel polar vortices among conventional a1/a2 ferroelectric domains [1]. When these superlattices are driven by a 400 nm, above PTO-bandgap, optical excitation with sufficient fluence, a metastable supercrystal phase with a mesoscale periodicity of 30 nm in all three directions can be created [2]. We used the recently enabled 343 nm wavelength laser-pump, x-ray nanodiffraction probe (nano-XRD) at Sector 26 to image in-situ the nucleation and growth of the supercrystal phase at the nanoscale as a function of optical energy dose. A new optically induced phase was discovered, as well as an intermediate, less ordered, optically induced phase before converting to a final supercrystal phase after sufficient exposure time and fluence. It is hypothesized that the different supercrystal phases come about due to critical photo-carrier densities forming in either one or both sets of the superlattice layers. These new phases have novel electrical and optical properties, and the insights gained from this study may lead to new optically driven routes towards optically driven control of phase transformations.

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