AI/ML/Autonomous Experimentation for Thin Films Processing
Room 317 - Session AIML1-ThM
Linking AI/ML Tools with Diagnostics and Film Growth: PLD/CVD/ALD

Moderators: Renato Camata, University of Alabama at Birmingham, **Gregory Doerk**, Brookhaven National Laboratory

8:00am **AIML1-ThM-1 AI-Driven Synthesis by Pulsed Laser Deposition: Autonomous Experimentation and Human-AI Collaboration***, **Sumner B. Harris,*** Center for Nanophase Materials Sciences, Oak Ridge National Laboratory **INVITED**

Thin-film synthesis with pulsed laser deposition (PLD) presents a vast design space of chemistries, structures, and non-equilibrium processing pathways that is challenging to navigate when targeting specific film properties. By combining automated synthesis platforms equipped with multimodal in situ diagnostics and a range of AI methods, we can generate testable hypotheses, autonomously explore large parameter spaces, uncover non-trivial correlations between diagnostics and film properties, and control the growth trajectory in real time. In this talk, I will discuss human–AI collaborative autonomous synthesis, which employs an LLM as a co-scientist to help generate testable hypotheses for oxide remote epitaxy and assists with additional LLM-based data analysis between batches of fully autonomous PLD experiments. I will further discuss how this example sets the stage for Agentic AI-driven synthesis and characterization platforms, incorporating cross-modal knowledge to make decisions and analyze results.This work was supported by the Center for Nanophase Materials Sciences (CNMS), which is a US Department of Energy, Office of Science User Facility at Oak Ridge National Laboratory. PLD synthesis was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division

8:30am **AIML1-ThM-3 Physics-Informed Gaussian Process Active Learning for Modeling of PLD Plume Dynamics***, **Zahra Nasiri**, Dorien Carpenter, Jacob H Paiste,* University of Alabama at Birmingham*; Sumner B Harris,* Center for Nanophase Materials Sciences, Oak Ridge National Laboratory*; Renato P Camata,* University of Alabama at Birmingham

In pulsed laser deposition (PLD), accurate models of plume particle flux and kinetic energy as functions of laser fluence and spot area are essential for autonomous thin film growth optimization. It is well established that Gaussian Process (GP) active learning can construct high-fidelity representations of these functions from a small number of strategically selected experiments. Physics-inspired structured mean functions have been identified as a promising direction for improving model performance and sample efficiency. Here, we present a fully developed physics-informed GP framework that embeds this physical structure directly into the surrogate modeling process.

We replace the conventional zero-mean GP prior with physically motivated baselines derived from an isentropic expansion model. For kinetic energy, the structured mean expresses the kinetic energy of ablated species in terms of a stagnation temperature at the beginning of expansion and the terminal Mach number of the plume. The terminal Mach number is derived from the Knudsen number for each combination of fluence and spot area through a known analytical relation involving the heat capacity ratio. The stagnation temperature is modeled through a secondary GP trained on fluid dynamics simulation data. For particle flux, the structured mean captures the scaling with the initial vapor density and the terminal Mach number, where the initial thermodynamic state is determined through the laser-target interaction. The primary GP learns only residuals from these baselines, reducing model complexity and improving sample efficiency.

Performance is evaluated via root mean square error (RMSE) and compared against zero-mean GPs and random sampling. Results from 100 independent runs confirm that embedding isentropic expansion physics dramatically reduces model error, with RMSE dropping from approximately 3.5% to 0.1%. We carry out experimental validation through real-time ion probe measurements of KrF excimer laser ablation of copper in vacuum. We will discuss how to interpret the time-dependent Mach number extracted from the surrogate model and its use to predict the angular distribution of the PLD plume, which is critical for film uniformity and thickness control. Finally, we generalize our approach to complex compound materials, with the iron-based superconductor FeSe as an initial target where stoichiometry control presents additional modeling challenges.

8:45am **AIML1-ThM-4 Toward Autonomous Screening of oMLD Thin Films for Next-Generation Lithium-ion Battery Materials***, **Campbell A. Sweet**[1], Kim A. Gerhard, Blaine D. Kelly, Lucas R. Kuehnel, Matthias J. Young,* University of Missouri

Organic conductive polymers with high lithium-ion redox capacities are promising as sustainable positive electrode materials and as performance-enhancing coatings for established cathodes such as lithium iron phosphate. Oxidative Molecular Layer Deposition (oMLD) enables the delivery of conformal organic conductive polymer thin films on porous powder substrates with precise thickness control, making it a promising platform for this application domain. However, exploration of oMLD chemistries remains limited because the experimental workflows are time-intensive and require substantial manual intervention. Given the large chemical design space of organic precursors, traditional oMLD workflows are not readily scalable for rapid materials discovery. Here, we discuss progress to date on establishing a rapid and autonomous workflow for screening oMLD thin films as organic positive electrode coating materials for lithium-ion batteries. We specifically describe our efforts to accelerate deposition, automate precursor handling, and streamline electrochemical characterization and analysis. Workflow analysis revealed key bottlenecks including low precursor volatility, slow performance characterization, and inefficient chemical data mapping and search. We report progress toward autonomous workflow operation using LLM agents to manage repetitive and cumbersome tasks. These advances provide a framework for accelerating oMLD-driven materials discovery and may extend to other challenging-to-accelerate workflows.

9:00am **AIML1-ThM-5 QCMPy: Open-Source Python Codebase for in-Situ QCM Kinetics Monitoring***, **Eden Goodwin**[2], Maram Bakiro, Victoria Velez, Derek Reid, Seán Barry,* Carleton University, Canada

It is well known that not all chemical systems are equally prepared for modelling with Artificial Intelligence (AI). Well documented chemical systems with predictable structures and invariable synthesis have yielded phenomenal modelling results, as is best exemplified by the Nobel protein modelling AI software AlphaFold.[1] Small molecule systems are more difficult to model as they have inconsistent documentation, more dimensions of structural diversity, and significant variability in synthetic approach. Structural modifications can introduce substantial changes to the synthetic procedure, new decomposition pathways, intermolecular interactions, or even unpredicted functionality at the surface. These factors make iteration on and testing of models challenging. As such a different approach may yield better results: extraction of critical information from under-analyzed data.

A prime target for this data collection is an under utilized, robust, affordable, in-situ metrology: Quartz Crystal Microbalance (QCM). QCMs are used within Atomic Layer Processing (ALP) to measure in-situ the growth, etch, and modification of surfaces as they are exposed to highly reactive precursors. The relative mass gain due to each step of a sequential ALPs can provide valuable chemical insights, especially when coupled with other in-situ techniques. Careful kinetics analysis of sequential adsorptions of N-Heterocyclic Carbenes on surfaces revealed a previously unreported phenomenon: Atomic Layer Restructuring.[2] A reduction in the maximal coverage of the physiosorbed component revealed a significant reduction in the number of available surface sites and consequently surface roughness.

To translate kinetics modelling to a broader audience we have developed an open-source tool "*QCMPy*" that (1) separates a QCM trace of into cycle by cycle traces for each precursor then (2) aligns cycle traces using pulse detection algorithms and finally (3) conducts comprehensive cycle by cycle adsorption modelling. To demonstrate QCMpy we probe the growth kinetics of trimethyl aluminum using three different co-reactants: Water, Fumaric Acid and Salicylic acid. Analysis of the modeled parameters (ka, θmax) over each individual Molecular Layer Deposition cycle reveal pulsing drift and variability in our ALD-reactor, predict the transition from interfacial to bulk growth (Figure 1), predict the ideal growth per cycle from sub-saturative recipes and quantify the chemisorption/physisorption ratio of TMA and all co-reactants.

References

1- Jumper, J. et. al. *Nature* **2021**, *596* (7873), 583–589.

[1] **JVST Highlighted Talk**
[2] **TFD James Harper Award Finalist**

2- Goodwin, E. et al. *ACS Nano* **2025**, *19* (16), 15617–15626.

9:15am **AIML1-ThM-6 Accelerating Peald Process Development with Explainable Machine Learning**, ***Hamidur Rahman***, University of Michigan, Ann Arbor; *Ian Campbell,* IMEC USA; *Paula Arellano-Vasquez, Joshua Kammeraad, Paul Zimmerman, Ageeth Bol,* University of Michigan, Ann Arbor

Plasma-enhanced atomic layer deposition (PEALD) has emerged as a promising route to synthesize emerging 2D materials, yet rationalizing its highly nonlinear, multi-parameter process space remains an open challenge. Machine Learning (ML) promises to address this challenge, but requires consensus on which models, validation schemes, and feature sets are appropriate for the relatively high-noise, small data sets that are typically generated in PEALD studies. We address this issue by a systematic, end-to-end methodology study to optimize the crystallinity and surface morphology of PEALD MoS_2. Building on a critical survey of prior approaches, we systematically evaluate ML practices on our own controlled experimental data set, collected through experiments designed to optimize material properties, to clarify which ML models reliably translate to the small-sample, high-noise regime characteristic of PEALD.

A PEALD process to synthesize MoS_2 is developed using metal $Mo(N^tBu)_2(NMe_2)_2$ precursor and combinations of Ar/H_2 plasmas and a tert-butyl disulfide co-reactant. Various ALD parameters, such as process pressure, plasma power, gas composition, etc., were systematically varied to guide the film growth towards more crystalline and minimal out-of-plane growth using Atomic Force Microscopy (AFM). The data were subsequently modeled using eight algorithms encompassing linear regressors, k-nearest neighbors, random forest variants, and gradient boosting methods. A comparative evaluation of these models is conducted to explore the relationships inherent in PEALD process parameters and forecast material properties. Incorporating Raman Spectroscopy features as auxiliary inputs was also explored, highlighting the potential of multimodal data in low-data ALD studies. Model interpretability techniques were applied to examine the contributions of individual steps in the PEALD cycle to material properties, providing physically meaningful insights that complement human interpretation of the underlying chemistry. The trained models drive inverse recipe design, yielding candidate process windows for target film properties. We further explore in-situ spectroscopic ellipsometry as a real-time surrogate for ex-situ characterization, paving the way toward closed-loop control. Finally, we outline a roadmap for a transferable PEALD digital twin, where MoS_2-trained models can be adapted via transfer learning to analogous precursor systems, laying the foundation for autonomous, ML-guided PEALD.

9:30am **AIML1-ThM-7 Non-Negative Matrix Factorization for in Situ Gas Analysis During Atomic Layer Deposition**, ***Andreas Werbrouck***, *Aditya Chalishazar, Dirk Poelman, Philippe F. Smet, Jolien Dendooven, Christophe Detavernier,* Ghent University, Belgium

In situ characterization of atomic layer deposition (ALD) processes enables the study of reaction mechanisms, kinetics, and industrial process control. An established way to monitor gas species in a reactor is quadrupole mass spectrometry (QMS). In past work, we have shown that time-resolved, full-range mass spectrometry data with very good sensitivity can be collected provided that steady-state reaction conditions are maintained. Another method for gas-phase characterization is capturing the optical emission from a cold cathode discharge (optical emission spectroscopy or OES). OES could quickly detect and characterize gas-phase constituents under non-steady-state conditions. Both methods yield dense, time-resolved series of spectra, showing multiple correlated and potentially overlapping peaks. Therefore, we adopt non-negative matrix factorization (NMF), a machine learning technique, as a holistic way to analyze the obtained datasets. NMF is able to identify fingerprints of gas phase elements, and their time evolution. We evaluated the use of NMF for QMS and OES analysis by studying the trimethylaluminum (TMA)-H_2O and diethylzinc (DEZ)-H_2O ALD chemistries.

In the case of OES, we demonstrate that a naive analysis method of tracking channel intensities corresponding to specific emission lines is not sufficient to identify and separate reaction products. Using NMF, we were able to identify spectra and time signatures from precursor fragments and reaction products, confirming the established reaction mechanism between TMA and H_2O yielding CH_4. The NMF analysis also suggests that the discharge generates spurious H signals that can give rise to wrong interpretation of the H emission lines.

For QMS under steady state conditions, NMF easily and cleanly separates precursors and reaction products. Interpretation of mass spectra is easier, compared to OES, and the clean spectra and time traces NMF yields allow for faster identification of novel reaction products. This way, by using NMF on OES and QMS data, we reveal autocatalytic alkene formation during the DEZ-H_2O process, in addition to the generation of ethane.

With this work, we aim to draw attention to a relatively simple machine learning method that can be straightforwardly adopted by experimentalists. While in situ experiments are capable of yielding (very) large datasets, NMF helps to draw a complete picture of the gas phase during ALD processes.

9:45am **AIML1-ThM-8 Machine Learning Assisted Multiscale Simulation of Metal Deposition**, ***Michael Nolan***[1], Tyndall Institute, Ireland; *Karl Ronnby, Samuel Delgado,* Tyndall National Institute, University College Cork, Ireland

The deposition of metals onto a multitude of substrates and the resulting interfaces are critical elements of many technologies, e.g. semiconductor devices, catalysts, sensors, etc. Atomistic simulations provide a powerful tool to select between different metal-substrate combinations for target technologies. Semiconductor devices using 2D materials require good quality interfaces between metal contacts and the 2D channel, back end of line interconnects require smooth conducting metal films that are immune to metal diffusion and catalysts using support metals benefit from metal islands with high density of active sites. In this contribution we present atomistic multiscale simulations of metal deposition and morphology evolution that use machine learning potentials to accelerate simulations and reach time and length scales that are out of reach with density functional theory (DFT). We use DFT to explore how the modification of TaN can promote a change in morphology of deposited copper from islands to lateral growth, computing activation barriers for critical metal migration processes using neural network ML potentials, which reduces the time for barrier calculation to a few minutes. With these barriers we employ kinetic Monte Carlo simulations to predict key film properties including roughness, covered surface area and density of defects. A similar approach is employed for Ru, which is of high interest for metal interconnects. Cu-Ru alloys are of interest and we present genetic algorithm studies of global minimum energies structures with an alloy ML potential and apply this to atom migrations. Finally, we explore Ru-TMD interfaces and dynamics to understand formation of Ohmic contacts found experimentally.

[1] **JVST Highlighted Talk**

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