Advanced Characterization, Modelling and Data Science for Coatings and Thin Films
Room Town & Country C - Session CM3-3-ThA

Data-Driven Thin Film Design: High-Throughput Experimentation, Simulation, and Machine Learning III

Moderators: Kevin Kaufmann, Oerlikon, USA, **Sebastian Siol**, Empa, Switzerland

1:20pm **CM3-3-ThA-1 Transforming Thin-Film Research Through Autonomous Experimentation: From Synthesis to Long-Term Device Performance***, **Davi Febba [davimarcelo.febba@nrel.gov]**, Brooks Tellekamp, William Callahan, Andriy Zakutayev,* National Renewable Energy Laboratory, USA **INVITED**

The synthesis and characterization of thin-film materials traditionally require lengthy experimental campaigns, where processing conditions are iteratively adjusted to achieve desired compositions and properties. This challenge intensifies when studying complete devices, as interfacial phenomena play a decisive role in performance.

Autonomous experimentation is transforming this paradigm. By automating repetitive tasks and deploying AI-driven experiment planners, researchers can dramatically accelerate the materials discovery and optimization pipeline while reducing manual intervention.

In this presentation, I will summarize recent advances at NREL in (i) autonomous sputtering and molecular beam epitaxy (MBE) growth of thin films[1,2], and (ii) autonomous, long-term degradation studies of electronic devices under extreme environmental conditions[3]. These platforms integrate genetic algorithms, computer-vision feedback, and multidimensional Bayesian optimization to identify the most informative experiments in real time—maximizing information gain per unit time. I will also discuss the design and deployment of AI agents for direct instrument control, demonstrating how they enhance safety, reliability, and throughput.

Finally, I will highlight the practical aspects of retrofitting existing laboratories for autonomy, including instrument-level automation, workflow orchestration, data-management infrastructure, and networking strategies that link remote servers with local command-and-control systems. Together, these elements enable seamless, closed-loop operation across diverse scientific instruments, advancing the vision of fully autonomous research laboratories at NREL.

1. Fébba, D. M. *et al.* Autonomous sputter synthesis of thin film nitrides with composition controlled by Bayesian optimization of optical plasma emission. *APL Materials* **11**, 071119 (2023).
2. Schaefer, S. *et al.* Rapid screening of molecular beam epitaxy conditions for monoclinic $(In_xGa_{1-x})_2O_3$ alloys. *J. Mater. Chem. A* (2024) doi:10.1039/D3TA07220G.
3. Fébba, D., Egbo, K., Callahan, W. A. & Zakutayev, A. From text to test: AI-generated control software for materials science instruments. *Digital Discovery* **4**, 35–45 (2025).

2:00pm **CM3-3-ThA-3 HiPIMS Process-Optimization in an Autonomous Sputter Chamber via Active Learning***, Alexander Wieczorek, Nathan Rodkey, **Sebastian Siol [sebastian.siol@empa.ch]**,* Empa - Swiss Federal Laboratories for Materials Science and Technology, Switzerland

The growing demand for data-driven discovery in materials science has spurred rapid advances in autonomous experimentation. However, these developments have so far rarely extended to physical vapor deposition (PVD) methods, largely due to the technical challenges of automating such complex systems. Yet, the PVD community faces an urgent need for systematic data acquisition, as its processes continue to gain complexity. For instance, high-power impulse magnetron sputtering (HiPIMS) has become increasingly prevalent but introduces several additional process dimensions compared to conventional DC sputtering, such as frequency, pulse width, and peak current density. This expanding parameter space complicates experimental optimization and impedes a deeper understanding of the physical mechanisms governing HiPIMS.

To address this challenge, we developed an autonomous sputter deposition platform interfaced with LabView and controlled via a Python-based code utilizing Bayesian optimization. The system efficiently explores a defined parameter space through iterative, data-informed sampling. Large datasets of HiPIMS process parameters are collected autonomously and subsequently analyzed using Shapley Additive Explanations (SHAP), a machine learning approach capable of disentangling complex, high-dimensional relationships. This combination of automation, Bayesian statistics, and interpretive modeling enables data-driven insights into the underlying physics of advanced PVD processes. In this presentation we will show the experimental setup and workflow as well as first studies of HiPIMS parameter optimisation using in-situ plasma diagnostics.

2:20pm **CM3-3-ThA-4 Accelerating Experiments with AI and Automation: Powder Materials and their Compositional Characterization***, **Andrea Giunto [agiunto@lbl.gov]**, Yuxing Fei, Bernardus Rendy,* Lawrence Berkeley Lab, University of California, Berkeley, USA*; Pragnay Nevatia,* University of California at Berkeley, USA*; Nathan Szymanski, Gerbrand Ceder,* Lawrence Berkeley Lab, University of California, Berkeley, USA **INVITED**

Computational materials science, accelerated by AI, has enabled the prediction of thousands of new inorganic compounds. However, their experimental realization remains a key bottleneck. To close this gap, automated and AI-driven laboratories are emerging. In our group, we have developed the **A-lab**, a platform for automated solid-state synthesis and characterization of powder materials [1,2]. This talk will present the A-lab's capabilities, focusing on the challenges of reaction product characterization and our automated, AI-based solutions. We combine X-ray Diffraction (XRD) for structural analysis with automated compositional analysis by Energy-Dispersive X-Ray Spectroscopy (EDS) in a desktop SEM, using a framework developed in-house, and implemented in the Python package **AutoEMXSp** [3]. I will discuss strategies to obtain accurate compositional analysis of powders and how these methods can be extended to thin-film materials.

References:

[1] Szymanski, N.J., *et al.* An autonomous laboratory for the accelerated synthesis of novel materials. *Nature* **624**, 86–91 (2023)

[2] Szymanski, N.J., *et al.* Autonomous and dynamic precursor selection for solid-state materials synthesis. *Nat Commun* **14**, 6956 (2023)

[3] Giunto, A., *et al.* Harnessing Automated SEM-EDS and Machine Learning to Unlock High-Throughput Compositional Characterization of Powder Materials, 14 October 2025, PREPRINT [https://doi.org/10.21203/rs.3.rs-7837297/v1]

3:00pm **CM3-3-ThA-6 Autonomous Experimentation with Quality Control and Cross-Facility Coordination***, **Yongtao Liu [liuy3@ornl.gov]**,* Oak Ridge National Laboratory, USA **INVITED**

Recent advancements in AI-driven autonomous experimentation (AE) are transforming the landscape of materials research These systems hold great promise for accelerating discovery, yet fully autonomous frameworks often struggle with the complexity, variability, and evolving nature of real-world experimental environments, sometimes misleading the AE process. In this talk, I will discuss our approach for overcoming these challenges by embedding quality control and expert guidance into active learning–based AE systems. Rather than relying solely on ML optimization, our framework allows experts to guide and refine the system's exploration in real time, leading to more meaningful experimentation. We have implemented this approach in autonomous thin-film synthesis and microscopy characterization, but its principles can be extended to many other AE platforms.In addition, as materials development increasingly relies on multimodal characterization to reveal the intricate chemical-structure-property relationships, most autonomous materials research platforms are limited to a narrow set of diagnostic tools due to constraints in lab space, available expertise, instrumentation capacity, etc. This hinders their ability to make informed decisions and generalize across diverse material systems. To address this gap, we further extend our approach to connect distributed AE platforms and supports hybrid integration of automated and manual tools, which broadens the diagnostic capabilities available to the autonomous research process. This framework enables real-time data exchange and coordinated decision-making across multiple systems, allowing independent platforms to collaborate seamlessly without requiring physical integration. This points toward an interconnect model of autonomous research by linking distributed facilities for more collaborative and adaptive autonomous materials discovery. Acknowledgments: This research 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. This research and workflow development was sponsored by the INTERSECT Initiative as part of the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle, LLC for the US Department of Energy under contract DE-AC05-00OR22725.

4:00pm **CM3-3-ThA-9 Self-Navigating Thin Film Laboratory: Real-Time AI-Driven Optimization of Functional Thin Films**, ***Ichiro Takeuchi [takeuchi@umd.edu]***, University of Maryland, USA **INVITED**

Autonomous experimentation can be used to reduce the number of required experimental cycles for materials optimization by an order of magnitude or more by enlisting Bayesian optimization using Gaussian Processes. We have demonstrated autonomous control of unit cell-level growth of functional thin films implemented in pulsed laser deposition. Dynamic analysis of reflection high-energy electron diffraction images is used to autonomously navigate multi-dimensional deposition parameter space in order to rapidly identify the optimum set of growth parameters for fabricating the targeted materials phase. As an example, we have set up the autonomous system to synthesize the meta-stable hexagonal phase of $TbFeO_3$ and other rare-earth ferrites where substrate temperature, oxygen pressure, and the laser repetition rate are varied simultaneously. The self-navigating algorithm is able to consistently find the optimum conditions within 10-15 iterations resulting in thin films of the phase pure hexagonal phase. Our scheme can be applied to any type of thin film/semiconductor manufacturing setting where an effective in-situ characterization tool can be used to provide real-time autonomous feedback. This work is carried out in collaboration with Haotong Liang, Mikk Lippmaa, and A. Gilad Kusne, and is supported by the center for 3D Ferroelectric Microelectronics Manufacturing (3DFeM2), an Energy Frontier Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences under Award Number DE-SC0021118.

4:40pm **CM3-3-ThA-11 Advances in the Rapid Characterization of Sputter-Deposited, Binary Metal Thin Films Made by Combinatorial Techniques**, ***David Adams [dpadams@sandia.gov]***, *Finley Haines, Sadhvikas Addamane, Kyle Dorman, Remi Dingreville, Saaketh Desai, Brad Boyce, Mark Rodriguez,* Sandia National Laboratories, USA

Combinatorial sputter deposition techniques provide access to a rich variety of thin films that can be exploited for rapid design optimization and process refinement. Indeed, several combinatorial, magnetron sputtering approaches have been reported over the past decade demonstrating an ability to produce 10s or 100s of unique films in a single deposition experiment. In order to capitalize on the increased throughput provided by combinatorial methods, we seek to develop complementary, high-throughput film characterization techniques that accurately determine important film properties.

With this presentation, we describe two, new techniques that have accelerated the development of binary PtAu and CuAg films for use as metal contacts. First, we describe a high-throughput, X-ray reflectivity (XRR) analysis method that rapidly determines the density of >100 unique, combinatorial films produced in a given deposition. Traditionally, complex fitting procedures are applied to XRR to estimate the critical angle (angle at or below which total reflection occurs), which can then be used to calculate the film density. This study demonstrates an alternative method – using an indirect surrogate angle θ_s (instead of θ_c) that is numerically calculated (without any curve-fitting) as the minima in the first derivative of the acquired XRR profiles. It was found that density values estimated using θ_s and adjusted with a systematic offset were in agreement with the traditional curve-fitting method, with typical average error peaking at < 2% and reduced hands-on analysis time by ~95%. Second, we describe progress toward automated, ex-situ measurement of combinatorial film stress using a wafer curvature mapping approach. Employing a k-Space Co. ThermalScan instrument, we rapidly interrogate the curvature of >100 individual substrate pieces coated uniquely in a single deposition. We determine the residual stress of the various films using Stoney's equation and demonstrate extensions to automated measurements of thermal expansion coefficient. Altogether, the gathered information augments an extensive combinatorial library providing opportunities to pinpoint relevant process-structure-property relationships for improved, reliable thin film performance.

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Author Index

Bold page numbers indicate presenter

— A —
Adams, David: CM3-3-ThA-11, **2**
Addamane, Sadhvikas: CM3-3-ThA-11, 2
— B —
Boyce, Brad: CM3-3-ThA-11, 2
— C —
Callahan, William: CM3-3-ThA-1, 1
Ceder, Gerbrand: CM3-3-ThA-4, 1
— D —
Desai, Saaketh: CM3-3-ThA-11, 2
Dingreville, Remi: CM3-3-ThA-11, 2
Dorman, Kyle: CM3-3-ThA-11, 2
— F —
Febba, Davi: CM3-3-ThA-1, **1**
Fei, Yuxing: CM3-3-ThA-4, 1
— G —
Giunto, Andrea: CM3-3-ThA-4, **1**
— H —
Haines, Finley: CM3-3-ThA-11, 2
— L —
Liu, Yongtao: CM3-3-ThA-6, **1**
— N —
Nevatia, Pragnay: CM3-3-ThA-4, 1
— R —
Rendy, Bernardus: CM3-3-ThA-4, 1
Rodkey, Nathan: CM3-3-ThA-3, 1
Rodriguez, Mark: CM3-3-ThA-11, 2
— S —
Siol, Sebastian: CM3-3-ThA-3, **1**
Szymanski, Nathan: CM3-3-ThA-4, 1
— T —
Takeuchi, Ichiro: CM3-3-ThA-9, **2**
Tellekamp, Brooks: CM3-3-ThA-1, 1
— W —
Wieczorek, Alexander: CM3-3-ThA-3, 1
— Z —
Zakutayev, Andriy: CM3-3-ThA-1, 1