Advanced Characterization, Modelling and Data Science for Coatings and Thin Films
Room Palm 1-2 - Session CM3-1-ThA

Accelerated Thin Film Development: High-throughput Synthesis, Automated Characterization, and Data Analysis I

Moderators: Davi Marcelo Febba, NREL, USA, **Sebastian Siol**, Empa, Switzerland, **Andriy Zakutayev**, NREL, USA

1:40pm **CM3-1-ThA-2 Collaborative Intelligence in Thin Film Research for Clean Energy Technologies***, **Shijing Sun (shijing@uw.edu)**,* University of Washington, USA **INVITED**

Addressing global environmental challenges, particularly in the realm of energy storage and conversion, necessitates innovative approaches. In this context, artificial intelligence (AI) has emerged as a transformative tool, catalyzing the discovery of new materials. However, the practical application of computational models in laboratory settings presents distinct challenges. This talk will explore the evolving role of AI in scientific research, focusing on its capacity to enhance rather than replace human expertise. This synergy paves the way for advanced collaborative efforts in the development and analysis of thin films.

Drawing from my experience as both an experimentalist and a materials data scientist in academic and industrial settings, I will showcase data-driven approaches that accelerate the formulation of precursors for solution-processed thin films. Additionally, I will delve into how AI-assisted image characterisation can effectively detect imperfections and establish crucial structure-property correlations. These advancements are particularly significant in the pursuit of clean energy solutions, demonstrating the integral role of AI in accelerating scientific innovation in thin film technology.

2:20pm **CM3-1-ThA-4 Discovery and Design of a New Functional Amorphous Nitride: Y-W-N***, **Oleksandr Pshyk (oleksandr.pshyk@empa.ch)**, S. Zhuk, J. Patidar, A. Wieczorek,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland*; A. Sharma, J. Michler,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Thun, Switzerland*; C. Cancellieri,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland*; V. Stevanovic,* Colorado School of Mines, USA*; S. Siol,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland

Amorphous thin films are employed in many applications and offer unique characteristics, which are not observed in their crystalline counterparts.

We present the discovery and design of amorphous Y-W-N ceramic thin films. We performed an exploratory synthesis and high-throughput characterization of $Y_{1-x}W_xN$ thin films. The compositions are performed using combinatorial, reactive radio-frequency magnetron co-sputtering of Y and W targets in Ar/N_2 atmosphere, resulting in materials libraries with orthogonal composition and deposition temperature gradients. This allows for a rapid screening of the synthesis phase space. A composition window within $0.1 \leq x \leq 0.85$ is covered, whereas the substrate temperature (T_s) is varied from 80 ℃ up to 600 ℃. High-throughput screening of the composition and structure of the libraries by means of XRF and XRD reveals a wide composition range of $0.2 \leq x \leq 0.6$ where thin films grow with an amorphous structure without precipitation. Moreover, the amorphous structure shows remarkable temperature stability of up to 600 ℃. Optical properties mapping using an automated high-throughput UV−vis photo-spectroscopy system suggests a band gap of >2.5 eV and confirms the phase purity showing only negligible sub band gap absorption. The band gap can be tuned by varying the cation composition, whereas the highest absorption onset for $Y_{1-x}W_xN$ thin films is found for Y-rich samples with x=0.3 implying an optical band gap of ~3.3 eV. Mechanical properties mapping using an automated nano-indentation system shows that the hardness of $Y_{1-x}W_xN$ thin films with $0.3 \leq x \leq 0.5$ doesn't change significantly as a function of T_s and the highest hardness of 9.45±0.05 GPa is found for samples with x=0.5 while the increase of Y concentration deteriorates hardness down to 8.45±0.06 GPa for films with x=0.3. A representative pair of amorphous $Y_{1-x}W_xN$ thin films with x=0.3 and 0.5 are selected for a detailed study. A thorough structural and compositional analysis of the latter films by means of high-resolution TEM reveals a homogeneous amorphous structure of the films with no signs of elemental segregations or crystallization. To comprehend the application potential of these materials, a comprehensive study of a wide range of functional and physical properties is performed including a set of optical and dielectric constants, diffusion barrier performance, oxidation resistance, and thermal stability. Experimental findings are corroborated by theoretical calculations for a better understanding of a complex relationship between the elemental composition of amorphous $Y_{1-x}W_xN$ thin films and their physical and functional properties.

2:40pm **CM3-1-ThA-5 Deposition of Highly Crystalline AlScN Films Using Synchronized HiPIMS – From Combinatorial Screening to Piezoelectric Devices***, **Jyotish Patidar (jyotish.patidar@empa.ch)**,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland*; S. Bette,* aixACCT Systems GmbH, Germany*; O. Pshyk, K. Thorwarth,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland*; R. Kessels,* aixACCT Systems GmbH, Germany*; S. Siol,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland

Piezoelectric thin films are crucial for many technologies, in particular for RF components for telecommunication. Wurtzite Aluminum Nitride (w-AlN) is one of the most widely used material for these types of applications. Recently, Aluminium Scandium Nitride (AlScN), is becoming more popular due to its increased piezoelectric coefficient. Highly crystalline and textured thin films are essential for high-performing piezoelectric devices. In our prior work, we demonstrated that metal-ion synchronized HiPIMS (MIS-HiPIMS) with moderate substrate bias potentials can offer key advantages in the deposition of these materials.[1] Here we explore how these concepts can be applied to AlScN films.

In AlScN films, high Sc concentrations enhance the piezoelectric response by softening of the phonon modes. However, high Sc content can also lead to structural frustration and precipitation of cubic ScN (c-ScN). Investigating the Sc non-equilibrium solubility and structural evolution upon scandium alloying is experimentally involved and thus rarely discussed. In our work, we employ a combinatorial approach for an accelerated estimation of the solubility limits and optimization of film's properties, for varying synthesis environments. We investigate different synthesis routes by hybrid co-sputtering of Al/Sc in a reactive environment through a combination of direct current magnetron sputtering (DCMS) and HiPIMS processes, along with different biasing strategies.

The combinatorial screening reveals a striking correlation between the ion kinetic energy and non-equilibrium Sc solubility. In addition, certain deposition modes prove to be more resilient against structural frustration than others. Particularly, a reduction of misaligned grains is observed with the application of a negative substrate bias potential. Based on the results from the screening, uniform $Al_{0.8}Sc_{0.2}N$ thin films were deposited on Ti/Pt contacts, for each synthesis strategy. Detailed characterization of these films show that based on the chosen synthesis modes, the stress state can be tailored from -1.5 to 1.5 GPa. On the other hand, measurements of the piezoelectric coefficient $d_{33,f}$ show a performance comparable to the current state-of-the-art.

The results of this study showcase how high-throughput experiments can facilitate the development of complex sputter processes but also highlight the potential of synchronized HiPIMS processes for the deposition of piezoelectric thin films and other defect-sensitive materials.

[1] Patidar, Jyotish, et al., Surface and Coatings Technology (2023), 129719.

3:00pm **CM3-1-ThA-6 Advancing Metallic Glasses for Biomedical Applications: A Comprehensive Study on CuAgZr Alloys Using Combinatorial Synthesis, High-Throughput Characterization, and Machine Learning***, **Krzysztof Wieczerzak (krzysztof.wieczerzak@gmail.com)**,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Laboratory of Mechanics of Materials and Nanostructures, Switzerland

This work presents a study on CuAgZr metallic glasses (MGs), highlighting their potential in biomedical applications due to their exceptional strength, corrosion resistance, and antibacterial properties. Our research employs a novel approach combining combinatorial synthesis, high-throughput characterization, and machine learning to explore the mechanical properties of these alloys. We introduce the material library of CuAgZr alloys produced using direct current magnetron sputtering (DCMS) and employ advanced characterization methods to assess their composition, structure, and mechanical properties. A key finding of our study is the significant influence of high oxygen content in Cu-rich regions, a result of post-deposition oxidation, on the mechanical behavior of these alloys. This insight is pivotal in understanding the role of oxygen in synthesized alloys and its correlation with the growth mechanism and chemical composition. The introduction of the "Scanning Indenter" device in our study marks a significant technological advancement, enabling the automatic mapping of full wafers and integration of various characterization techniques like X-ray fluorescence (XRF) X-ray diffraction (XRD), and nanoindentation. This facilitates a multimodal dataset creation, enhancing our understanding of material properties. Our findings also reveal the critical impact of nanoscale structural features on plastic yielding and flow in these alloys, with a notable correlation between atomic size mismatch, oxygen content, and hardness. Furthermore, we demonstrate the efficacy of machine learning, particularly the multi-layer perceptron (MLP) algorithm, in predicting the hardness of untested alloys, offering valuable insights for future research. This study not only sheds light on the mechanical properties of CuAgZr MGs but also underscores the potential of integrating combinatorial synthesis, high-throughput characterization, and machine learning for the development of new metallic glasses with enhanced strength and economic feasibility.

4:00pm **CM3-1-ThA-9 Accelerating Materials Discovery by Combining Combinatorial Synthesis of Thin-Film Libraries, High-Throughput Characterization and Data Science***, Alfred Ludwig (alfred.ludwig@rub.de),* Ruhr University Bochum, Germany **INVITED**

Discovery of new materials is a key challenge in materials science: e.g., new materials for sustainable production/storage/conversion of energy carriers are necessary to improve existing and to enable future energy systems. Efficient methods for discovery and optimization of new materials are necessary: Thin-film combinatorial materials science (1) is presented as an effective means to produce large datasets on new materials. This approach is useful for validation of theoretical predictions (e.g., from high-throughput computations), and production of large, consistent and complete experimental datasets which can be used for materials informatics. The approach comprises fabrication and processing of thin-film materials libraries by combinatorial sputter deposition processes and optional post-deposition treatments, followed by the high-throughput characterization of the different thin-film samples contained in these libraries, and finally the organization of the acquired multi-dimensional data in adequate databases as well their effective computational analysis and visualization. The importance of defining adequate screening parameters and according designs of materials libraries is addressed. High-throughput material characterization methods are automated, fast, and mostly non-destructive: examples are EDX, XPS and RBS for composition, XRD for crystal structure, high-throughput test stands for temperature-dependent resistance (phase transformation), magnetic, optical and mechanical properties as well as scanning droplet cells for (photo)electrochemical properties screening. Results for up to quinary systems are visualized in the form of composition-processing-structure-function diagrams, interlinking compositional data with structural and functional properties. The talk will cover and discuss examples of combinatorial discoveries and development of new materials in different materials classes and forms (films, nanoparticles) with a focus on compositionally complex materials. Furthermore, a new approach to accelerate atomic-scale measurements for complex alloys is presented as well as applications of materials informatics to accelerate and improve the materials discovery process.

4:40pm **CM3-1-ThA-11 Autonomous Sputter Synthesis and Data Management for Nitride Thin Films***, **Davi Febba (davimarcelo.febba@nrel.gov)**, K. Talley, K. Johnson, S. Schaefer, S. Bauers, J. Mangum, R. Smaha, A. Zakutayev,* National Renewable Energy Laboratory, USA

Autonomous experimentation has emerged as an efficient approach to accelerate the pace of materials discovery. Although instruments for autonomous synthesis have become popular in molecule and polymer science, solution processing of hybrid materials, and nanoparticles, examples of autonomous tools for physical vapor deposition are scarce yet important for the semiconductor industry.

Moreover, sputtering reactors usually available in the market can be challenging to incorporate into an autonomous workflow, mainly due to the lack of comprehensive programming support. This makes it difficult to interface the instruments with optimization and active learning routines, common to an autonomous setup. To overcome these limitations, we recently designed and built a highly automated co-sputtering reactor featuring extensive programming capabilities and support for server-client interactions with other programming languages [1].

Furthermore, this high vacuum instrument is equipped with four cathodes, each with a dedicated channel for plasma monitoring via optical emission spectroscopy. It allows the exploration of a wide substrate temperature range, from cryogenic temperatures up to 1000 °C, in addition to RF and DC substrate biasing. Additional capabilities include real-time deposition data logging of sputtering parameters (such as power, voltage, pressure, gas flow), control of gas distribution to individual targets, time-sequenced shutters, and turbo gate position. All of these enable the user to execute complex programmable synthesis recipes.

In this presentation, we will discuss the details of this unique sputtering instrument and its integration with Python routines, resulting in an autonomous workflow for the synthesis of nitride thin films with controlled composition [1]. Moreover, we will also outline the integration of the time-series data automatically generated by this sputtering instrument with the research data infrastructure (RDI) [2]. NREL's RDI catalogs experimental data from inorganic thin films experiments at NREL and enables the High-Throughput Experimental Materials Database (HTEM-DB) (https://htem.nrel.gov/) [3], which stores information about synthesis conditions, chemical composition, crystal structure, and optoelectronic properties of materials.

[1] APL Mater 11, 071119, 2023

[2] Patterns, 2, 100373, 2021

[3] Scientific Data 5, 180053, 2018

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