## Advanced Characterization, Modelling and Data Science for Coatings and Thin Films
## Room Town & Country D - Session CM3-1-MoA

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

**Moderators: Kevin Kaufmann**, Oerlikon, USA, **Po-Liang Liu**, National Chung Hsing University, Taiwan , **Sebastian Siol**, Empa, Switzerland

1:40pm **CM3-1-MoA-1 Predicting Outcomes of Thin-Film Synthesis from First Principles**, ***Vladan Stevanovic [vstevano@mines.edu]***, Colorado School of Mines, USA **INVITED**

The laws of thermodynamics are often used to predict outcomes of materials synthesis. However, thermodynamics alone cannot explain why some materials are easier and some much harder to grow, or why some systems exhibit strong tendencies toward forming stable amorphous phases and others do not. The situation is much more complicated for non-equilibrium synthesis methods (e.g. vacuum deposition), which often produce states that are not the thermodynamic ground states. In this talk I will describe the computational, first-principles technique we have developed to predict the likelihood for experimental realization of different states. We have found that many of the observed synthesis outcomes across material systems and growth methods can be explained using probabilistic arguments. The critical quantities are the sizes ("widths") of various local minima on the potential energy surface representing the states available to the system. By measuring the "widths" of local minima using the first-principles random structure sampling we have found that wider the local minimum (more probable in the random sampling) higher are its chances for experimental realization.[1] [#_edn1] This hypothesis has been validated against experiments and used to predict and explain synthesis outcomes more broadly. Examples include our recent work explaining why many layered ternary nitrides preferentially adopt a disordered rocksalt structure in as-grown thin-films despite the existence of ordered ground states much lower in energy.[2] [#_edn2] Or why $Y_2WN_4$ exhibits a strong tendency toward forming amorphous films, which are shown experimentally to be particularly resistant to crystallization.[3] [#_edn3] I will also discuss our approach to modeling kinetics of structural transformations and its utility in predicting the results of post-growth annealing.

[1] [#_ednref1] V. Stevanovic, Phys. Rev. Lett. 116, 075503 (2016)

[2] [#_ednref2] A. Zakutayev, M. Jankousky, L. Wolf, Y. Feng, C. L. Rom, S. R. Bauers, O. Borkiewicz, D. A. LaVan, R. W. Smaha, and V. Stevanovic, Nat. Synth. 3, 1471 (2024)

[3] [#_ednref3] O. V. Pshyk, S. Zhuk, J. Patidar, A. Wieczorek, A. Sharma, J. Michler, C. Cancellieri, V. Stevanovic, S. Siol, Adv. Mater. 2501074 (2025)

2:20pm **CM3-1-MoA-3 A Refined Toolbox for Predicting Phase Formation in PVD Thin Films**, ***Christian Gutschka [christian.gutschka@tuwien.ac.at]***, TU Wien, Austria*; David Holec,* Montanuniversität Leoben, Austria*; Jochen Schneider,* RWTH Aachen University, Germany*; Helmut Riedl-Tragenreif,* TU Wien, Austria

In recent decades, there has been a growing trend in the use of experimental and simulation-based screening methods across various fields of Materials Science. The main goal of these methods is to reduce the time and costs associated with laboratory experimentation. In the area of thin film technologies, especially regarding Physical Vapor Deposition (PVD) methods, combinatorial sputter deposition has emerged as a well-established experimental technique. However, ab initio methods, such as Density Functional Theory (DFT), often face limitations in accurately predicting essential properties like mechanical properties and the solubility of alloy components. The latter is of particular importance when novel thin film materials, such as carbides, nitrides, or borides, are engineered, as the extreme cooling rates during PVD promote the formation of metastable, often uncharted, solid solutions. Here trustful predictions in unexplored phase spaces, would be very helpful to optimize experimental work. However, one reasons why ab initio techniques frequently fail to yield satisfactory results for PVD thin films is that the resulting phase diagrams are markedly different compared to their equilibrium states – typically obtained from CALPHAD.

Nevertheless, over a decade ago, a model was proposed that connected combinatorial sputter deposition experiments with data obtained from DFT and CALPHAD. This model was capable of incorporating the effects of substrate temperature, target power and residual stress in the films. According to literature, the model has proven to be of adequate predictive accuracy, in the case of the metallic $W_{1-x}Cu_x$ and $V_{1-x}Cu_x$ [1] and the ceramic $Ti_{1-x}Al_xN$ and $V_{1-x}Al_xN$ [2,3] systems.

The present study aims to extend the methodology by virtue of two principal factors. Firstly, it negates the necessity for existing CALPHAD databases. Secondly, it incorporates the effects of interfacial energies stemming from microstructural causes, such as preferred crystal orientation and grain geometry. The present study focuses on ceramic thin films, and the initial stage involves a concise analysis of model dependencies regarding data from experiment and DFT. Subsequently, a reproduction and extension for the $Ti_{1-x}Al_xN$ and $V_{1-x}Al_xN$ systems is presented, with an outlook and presentation of ongoing work to test the method for established carbide and boride systems.

[1] Chang K., et al. Sci. Technol. Adv. Mater. 2016;17:210.

[2] Liu S., et al. Acta Mater. 2019;165:615.

[3] Liu S., et al. Acta Mater. 2020;196:313.

2:40pm **CM3-1-MoA-4 Pathways for the Preparation of Functional Coatings by Multiscale Modelling**, ***Jiri Houska [jhouska@kfy.zcu.cz]***[1], University of West Bohemia, Czechia **INVITED**

The lecture will cover different ways how to support the experimental research in the field of functional coatings by computer simulations. Various levels of theory (ranging from solid-state physics through atomic-scale ab-initio simulations to atomic-scale simulations based on empirical interaction potentials) and various simulation algorithms (ranging from static calculations of properties through searching for a local energy minimum to reproducing the time evolution of growing films) will be considered. Special attention will be paid to recent developments of the methodology. In all cases, the results will be compared to the experiment.

The first part will deal with a design of multilayered $VO_2$-based thermochromic coatings for smart energy-saving windows. Integral luminous transmittance and modulation of integral solar energy transmittance will be optimized in parallel, instead of a tradeoff between them. Prediction of coating color will be mentioned as well.

The second part will deal with an identification of maximum N content in amorphous Si–B–C–N networks of various compositions, assuming that it is limited by the formation, presence and loss of $N_2$ molecules. For example, the difficulties with the preparation of $C_3N_4$ will be explained by a maximum achievable stable N content in $CN_x$ of 42%.

The third part will deal with reproducing the atom-by-atom growth of functional coatings in a wide range of conditions, using examples such as $ZrO_2$, Cu-Zr or Ti-Al-N. Effects of energy distribution function (not only average energy delivered into the films), mass of arriving particles (momentum delivered into the films) and growth temperature will be explained.

3:20pm **CM3-1-MoA-6 HADB Database: From Data Generation to AI-Supported Predictions of Properties of Hard-Coating Alloys**, ***Igor Abrikosov [igor.abrikosov@liu.se]***, *Sheuly Ghosh, Lalith Kumar Gurram, Jonatan Wästlund, Davide Sangiovanni, Ferenc Tasnádi,* Linköping University, IFM, Sweden **INVITED**

We introduce the HADB, a Hard-coating Alloys DataBase, available at https://hadb.funmat-ii.se/. HADB addresses a critical gap in existing materials databases by focusing on random alloys, important for industrial hard coatings applications such as metal cutting tools, with key data on thermodynamic, elastic, and mechanical properties. Focusing on ternary nitride alloys in two prototype crystal structures, rocksalt and zincblende, we present a reproducible workflow for high-throughput data generation in the framework of the Density Functional Theory (DFT). The framework integrates structure preprocessing, strategic composition selection to maximize successful data generation while minimizing usage of computational resources, automated DFT job submission, robust error handling, and post-processing analysis into a scalable pipeline. Further, data generated in DFT calculations at zero temperature is complemented by data at finite temperatures generated through ab initio Molecular Dynamics simulations. We discuss the technical implementations of the database infrastructure including support for browse, query, retrieval, and API access through the OPTIMADE API to make this data findable, accessible, interoperable, and reusable (FAIR). Finally, we demonstrate the potential of HADB to facilitate efficient alloy design in combination with a machine-learning (ML) Predictor. Our ML Predictor utilizes Crystal Graph Convolutional Neural Network (CGCNN) that has been trained on elastic properties for around 8000 compounds from Materials Project database as

[1] **Bill Sproul Awardee Honorary ICMCTF Lecture**

well as data for about 100 nitride alloys containing elements Ti, Hf, Zr, Al from HADB. Given an alloy composition beyond the data generated with DFT and its crystal structure, the model predicts with high accuracy single crystal elastic constants, bulk modulus, shear modulus, Young's modulus and Poisson ratios for the alloys.

4:00pm **CM3-1-MoA-8 The Intersection of Energy, Entropy, and Exploration: Data-Driven Discovery of High-Entropy Materials**, ***Corey Oses [corey@jhu.edu]***, Johns Hopkins University, USA **INVITED**

High-entropy materials, including oxides, metal alloys, and halides, are opening transformative possibilities for hydrogen generation, fuel cells, catalysis, energy storage, waste-heat recovery, radioactive waste immobilization, and radiation tolerance. However, the immense combinatorial complexity of these systems presents significant challenges for discovery and optimization. We employ data-driven approaches rooted in thermodynamics and chemistry to accelerate materials exploration, integrating high-throughput simulation, machine learning, and experimental feedback in a closed-loop workflow. This strategy efficiently guides exploration toward stable, high-performance compositions. Case studies demonstrate robust agreement with experimental results in mapping phase stability and uncovering functional materials. By advancing closed-loop discovery, we highlight scalable pathways to next-generation materials for critical energy applications.

4:40pm **CM3-1-MoA-10 Optimal Catalysts for Methane Pyrolysis by Atomistic Modelling of Molecule-Surface Interactions**, ***David Holec [david.holec@unileoben.ac.at]***, *Martin Matas,* Montanuniversität Leoben, Austria **INVITED**

Methane pyrolysis, heat decomposition into solid C and gas $H_2$, offers a promising technology for converting natural gas into hydrogen without causing $CO_2$ emissions. However, the necessary operating temperatures are too high for large-scale hydrogen production by catalyst-free methane pyrolysis. Therefore, catalytic methane pyrolysis, using liquid-metal bubble column reactors, has gained widespread interest. In this context, finding suitable catalysts that lower the operating temperatures and thus make methane pyrolysis economically viable and environmentally bearable has become an important scientific goal.We approach this topic by modelling interactions between $CH_n$ ($n$=4, 3, ..., 1) molecules with different metallic surfaces using first-principles simulations. In the first part, we will present the results of the Sabatier analysis employing 0K adsorption energies on elemental metal surfaces. The results yield so-called volcano plots, which can be used for guiding the selection of the most efficient catalyst for selected conditions, i.e., temperature and methane partial pressure. In the second part of the talk, we will employ ab initio molecular dynamics to study the decomposition process directly at finite temperatures. We will also discuss the impact of the actual metal alloy composition and show that the catalytic efficiency is not a linear function between the two end members of the binary alloy. We will discuss quantities suitable for automatic statistical evaluation of the AIMD trajectories (such as bond-length and bond-length oscillations, or molecule-surface distance).These case studies will present state-of-the-art in gas–surface interaction modelling at the atomistic level: on the one hand, reliable qualitative predictions of trends guiding the experiments, and, on the other hand, the limitations for quantitatively capturing the complex experimental scenarios.

5:20pm **CM3-1-MoA-12 Multiscale Simulations from Precursors and Surface Chemistry to Thin Film Properties**, ***Fedor Goumans [goumans@scm.com]***, *Nestor Aguirrre, Nicolas Onofrio,* Software for Chemistry & Materials, Netherlands

Advanced device integration requires process-aware material descriptors that capture how precursor chemistry, plasma species and growth kinetics determine thin-film properties. We present a multiscale pipeline that couples DFT energetics, an active-learning M3GNet interatomic potential, automated PES exploration, and 3D kinetic Monte-Carlo growth simulations to produce spatially resolved property descriptors (growth rate, composition maps, defect/trap proxies, band-gap and dielectric indicators).

Starting from a foundation M3GNet universal machine learning interatomic potential (MLIP) and a small DFT seed set, we fine-tune and use uncertainty-guided sampling to automatically find intermediates and transition states; high-uncertainty configurations are re-computed with DFT and fed back to the MLIP, accelerating accurate exploration of the Potential Energy Surface (PES) at a little over MLIP cost with near-DFT accuracy. The verified reaction network parameterizes kMC to predict thin flim formation and etching, as functions of precursor, pulse timing, temperature and flux. We demonstrate the workflow on a Ru–H ALE case study: ML-accelerated PES exploration uncovered dissociative channels that shift band-gap and fixed-charge proxies; kMC maps reveal process windows minimizing interface trap formation while preserving selectivity. The approach provides compact, validated descriptors for process tuning and device correlation, shortening R&D cycles and guiding targeted experiments.

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