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

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

Moderators: Davi Marcelo Febba, NREL, USA, **Okeksandr Pshyk**, Empa, Switzerland, **Sebastian Siol**, Empa, Switzerland

8:20am **CM3-2-FrM-2 Combinatorial Synthesis and High-Throughput Characterization of Cu-Ag and Ni-Pt Thin Films Fabricated by Confocal Magnetron Sputter Deposition***, **Kyle Dorman (krdorma@sandia.gov)**, R. Kothari, N. Bianco, M. Kalaswad, C. Sobzcak, S. Desai, J. Custer, S. Addamane, M. Jain, F. DelRio, B. Boyce, R. Dingreville, D. Adams,* Sandia National Laboratories, USA

Nanocrystalline thin films feature the potential for enhanced or altered material properties compared to their bulk single crystal counterparts. While such possibilities are frequently limited by a lack of thermal stability, nanocrystalline thin films comprised of binary metal alloys such as $Pt_{0.9}Au_{0.1}$ have demonstrated greater resistance to annealing (P. Lu et al., Materialia, 2019) which is consistent with predicted thermodynamic preferences for a minority solute element to enrich and stabilize grain boundaries (J.R. Trelewicz et al., PRB, 2009). Recent studies on Pt-Au binary thin films have emphasized the role of grain boundary character in this solute stabilization (C. M. Barr et al., Nanoscale, 2021), and means of high-throughput combinatorial synthesis (McGinn, ACS Comb. Sci., 2019) have been developed to complement automated characterization and simulation capacity. To further develop understanding of the properties and synthesis of similar nanocrystalline binary metal systems, the suite of tools developed for Pt-Au analysis is now turned towards Cu-Ag and Ni-Pt combinations in search of optimized material properties and greater comprehension of nanocrystalline systems. Our study utilized simultaneous confocal sputter deposition of each pair of elements, with pulsed DC magnetron methods directing single element sources with a variety of approaches. The result, with the substrate fixed rather than rotated and the employment of photolithography, is a varied atomic composition across 112 samples on a single 150 mm diameter wafer. A series of such depositions, varying the gun-tilt angle and power at each cathode, allows swift examination of nearly the full range of alloy compositions. Wavelength Dispersive Spectroscopy, Atomic Force Microscopy, X-ray Diffraction, X-ray Reflectivity, sheet resistance, optical profilometry and nanoindentation were employed for high-throughput and fast-paced analysis. The binary collision Monte Carlo program SiMTra (D. Depla et al., Thin Solid Films, 2012) assisted with the deposition design to minimize the necessary quantity of sample batches, and enabled analysis of the energetic and compositional properties of the wafer at deposition with respect to the resultant hardness, modulus, film density, crystal texture, resistivity, and chemical stability for the case of tarnishing Cu-Ag combinations. The resulting correlations are examined with a goal of optimization of nanocrystalline material properties and identifying the corresponding fabrication conditions.

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8:40am **CM3-2-FrM-3 Combinatorial Screening of Al-Si-N-O Protective Coatings with Tunable Refractive Index***, **Stefanie Frick (stefanie.frick@empa.ch)**, A. Wieczorek, K. Thorwarth, O. Pshyk, J. Patidar, S. Siol,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland

Rising requirements of next-generation technologies lead to an increasing demand for multifunctional thin-film coatings. In order to accelerate the co-optimization of the functional properties of these coatings, combinatorial materials synthesis in combination with high-throughput automated characterization and data analysis is instrumental. The quarternary material system Al-Si-N-O is a promising candidate for protective optical coatings due to the hardness of its nanocomposite phase at low Si contents [1] and the tunability of the refractive index via the variation of the oxygen content [2]. In this work, thin film combinatorial libraries were deposited via reactive Hybrid Al-HiPIMS/Si-DCMS covering a significant fraction of the quarternary phase space by applying orthogonal anion- and cation gradients. Different approaches to obtain intentional oxygen gradients in combinatorial libraries, including varying localization of gas inlets as well as target configurations, will be addressed and the respective advantages and drawbacks will be discussed. Cation spreads mainly between 5 and 35 cation % Si were investigated as well as various oxygen contents depending on the respective anion gradient approach. Subsequently, the libraries were comprehensively characterized via automated mapping procedures with XPS, XRD, nanoindentation and UV-Vis spectroscopy. The latter allowed for automated refractive index determination employing the envelop method for transmission spectra according to Swanepoel [3]. The analysis of the data sets reveals the achievement of an indentation hardness of 20-24 GPa over a range of 10-35 cation % of Si corresponding to a refractive index of 2.03-2.05 in a pure nitride library. In addition, it is shown that the refractive index can be reduced down to 1.57-1.71 for a nearly pure oxide library and higher silicon contents (15-57 cation % Si) at the expense of hardness values of 7-9 GPa. The comprehensive combinatorial data sets allow for deeper insights in the composition-structure-property relationship in this complex material compared to experiments based on serial experimentation. Finally, the influence of the application of an RF bias at the insulating substrates, for an acceleration of the incident ions, on the film properties will be elaborated.

[1] A. Pélisson *et al.*, Surface and Coatings Technology, (2007), 202(4-7), 884-889.

[2] M. Fischer *et al.*, Science and Technology of Advanced Materials, (2019), 20(1), 1031-1042.

[3] R. Swanepoel *et al*, Journal of Physics E: Scientific Instruments, (1983), 16, 1214-1222.

9:00am **CM3-2-FrM-4 From Automated to Autonomous Thin Film Deposition Experiments***, **Andriy Zakutayev (andriy.zakutayev@nrel.gov)**,* NREL, USA **INVITED**

In this presentation, I will discuss recent progress towards demonstration of autonomous thin film deposition experiments in a highly automated combinatorial co-sputtering instrument at NREL. The autonomous operation will be demonstrated on the example of thin film reactively sputtered ternary nitride materials used for energy and electronic applications. A conceptual extension to other materials classes (e.g. oxides) ans other deposition methods (e.g. molecular beam epitaxy) will be discussed. A relation of autonomous deposition instruments to autonomous electrical characterization instruments, data processing pipelines, and high throughput computational reference data will be discussed.

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