Advanced Characterization, Modelling and Data Science for Coatings and Thin Films
Room Town & Country D - Session CM3-2-ThA
Accelerated Thin Film Development: High-throughput Synthesis, Automated Characterization and Data Analysis II

Moderators: Dr. Davi Marcelo Febba, NREL, USA, **Dr. Sebastian Siol**, Empa, Switzerland

1:20pm **CM3-2-ThA-1 Feature Selection and High-Throughput Synthesis: Can They Be Used to Predict Adsorption Energies on Multinary Materials?**, ***Hannah-Noa Barad [hannah-noa.barad@biu.ac.il]***, Bar-Ilan University, Israel **INVITED**

Electro-reduction of CO_2 to sustainable fuels and value-added chemicals is one of the most promising paths for closing the anthropogenic CO_2 cycle. The catalyst, the main component of the electrochemical CO_2 reduction reaction (CO_2RR), is used to reduce CO_2 dissociation activation energy. Metal and metal oxide catalysts have been studied as catalysts for CO_2RR, yet selectivity towards desired products remains elusive. To overcome this issue, discovery of new materials with more components (*e.g.*, ternary, or quaternary materials), is paramount. These multinary materials, have the potential to improve the selectivity and activity toward a desired product, due to synergistic effects between the elements. However, the exploration space is enormous and needs to be decreased. An important descriptor for realizing the reaction mechanism leading to a specific product by a given catalyst is the adsorption energy of the rection intermediates, like *CO. Yet, adsorption energies on these new and complex materials have not been studied systematically.

Here, we present the development of a machine learning model for the prediction of adsorption energies of materials. Our model is based on a simple description of the adsorption environment by choosing very basic features, and more intricate structural features, like orbital field matrix.[1] We also apply the moments theorem for the density of states (DOS)[2] to depict our materials in terms of closed paths in their lattices, from which we obtain features relating to the adsorption site. We also use high-throughput synthesis and characterization methods to try and obtain more experimental data points on new multinary materials to enhance out dataset. These methods will support prediction of adsorption energies of multinary materials to discover new highly active and selective CO_2RR catalysts.

[1] T. Lam Pham, H. Kino, K. Terakura, T. Miyake, K. Tsuda, I. Takigawa, H. Chi Dam, *Sci. Technol. Adv. Mater.* **2017**, *18*, 756.

[2] J. P. Gaspard, F. Cyrot-Lackmann, *J. Phys. C Solid State Phys.* **1973**, *6*, 3077.

2:00pm **CM3-2-ThA-3 Development of Cu, Ni-Co-Doped $Bi_2Te_2.7Se_{0.3}$ for Thermoelectric Energy Generation Using Pulsed Laser Deposition**, ***Yakubu Sani Wudil [yaqubswudil@gmail.com]***, King Fahd University of Petroleum and Minerals, Saudi Arabia

This work reports the preparation of ternary Cu/Ni/$Bi_2Te_{2.7}Se_{0.3}$ nanocomposite thin films via pulsed laser deposition. For comparison, pure $Bi_2Te_{2.7}Se_{0.3}$ (BTS) and binary Cu/$Bi_2Te_{2.7}Se_{0.3}$ and Ni/$Bi_2Te_{2.7}Se_{0.3}$ nanocomposites were also synthesized. Morphological characterizations revealed the presence of abundant grains typical of the BTS sample. Energy-dispersive spectroscopy confirmed trace amounts of Cu and Ni within the films, while X-ray photoelectron spectroscopy indicated that both metals were present as unoxidized metallic atoms, free from telluride formation. Structural analyses using X-ray diffraction and Raman spectroscopy showed peaks consistent with the pure BTS structure, suggesting that the dopants were primarily located at the grain boundaries within the BTS matrix. The ternary nanocomposites were prepared using a specialized configuration at three different Cu/Ni concentrations. The highest room temperature thermoelectric figure of merit (ZT) of 0.97 was achieved at the optimal doping concentration (BTS-2Cu/Ni), attributed to a simultaneous increase in power factor (2988 $\mu W/mK^2$) and a decrease in thermal conductivity (0.93 W/mK). The enhanced thermoelectric power factor resulted from the selective filtering of low-energy charge carriers, which improved the Seebeck coefficient. Additionally, the introduction of Cu and Ni into the nanocomposites created abundant grain boundaries that scattered phonons, reducing intrinsic lattice thermal conductivity and thereby enhancing the ZT value.

2:20pm **CM3-2-ThA-4 Autonomous Experiments for Thin Films and Solid Materials**, ***Taro Hitosugi [hitosugi@g.ecc.u-tokyo.ac.jp]***, The University of Tokyo, Japan **INVITED**

Integrating machine learning, robotics, and big data analysis into established research methodologies can significantly accelerate materials science research. Many studies have already demonstrated the potential of autonomous (self-driving) experiments in materials science [1, 2]. The rapid advancement of digital technologies is changing the way we conduct research.

Here, we discuss the status and prospects of data- and robot-driven materials research using autonomous experiments. We have developed an autonomous experimental system for thin-film materials. We constructed a system that automates sample handling, thin-film deposition, optimization of growth conditions, and data management. By using Bayesian optimization in conjunction with robots, our approach facilitates high-throughput experiments and generates comprehensive datasets that cover many aspects of materials (X-ray diffraction, Raman spectroscopy, scanning electron microscopy, optical transmittance measurement, electronic conductivity measurement). We tuned the hyperparameter for Bayesian optimization using the domain knowledge of chemistry; the number of trials to reach the global optimum is reduced.

The system demonstrated the synthesis and optimization of the electrical resistance in Nb-doped TiO_2 thin films [5]. Moreover, this autonomous approach has enabled the discovery of new ionic conductors [6]. We discuss the potential impact of this technology in accelerating materials science research, particularly in solid materials.

[1] Autonomous experimental systems in materials science, N. Ishizuki, R. Shimizu, and T. Hitosugi, STAM Methods 3, 2197519 (2023).

[2] The rise of self-driving labs in chemical and materials sciences, M. Abolhasani and E. Kumacheva, Nature Synthesis 2, 483–492 (2023).

[3] Tuning of Bayesian optimization for materials synthesis: simulation of the one-dimensional case, R. Nakayama, T. Hitosugi *et al.*, STAM Methods 2, 119-128 (2022).

[4] Tuning Bayesian optimization for materials synthesis: simulating two- and three-dimensional cases, H. Xu, R. Nakayama, T. Hitosugi *et al.*, STAM Methods 3, 2210251 (2023).

[5] Autonomous materials synthesis by machine learning and robotics. R. Shimizu, T. Hitosugi *et al.*, APL Mater. 8111110 (2020).

[6] Autonomous exploration of an unexpected electrode material for lithium batteries. S. Kobayashi, R. Shimizu, Y. Ando, T. Hitosugi, ACS Materials Lett. 5, 2711–2717 (2023).

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