Topical Symposia
Room Pacific E - Session TS4-ThA

Big Data, Machine Learning, Artificial Intelligence and High-Throughput Methods

Moderator: Igor Abrikosov, Linköping University, IFM, Sweden

1:20pm **TS4-ThA-1 New 3D and 2D Metal Borides from Materials Synthesis Guided by High-Throughput Simulations**, ***Johanna Rosen (johanna.rosen@liu.se)***, Linkoping University, Sweden **INVITED**

Exploratory theoretical predictions in uncharted structural and compositional space are integral to materials discoveries. A more recent addition to the family of laminated metal borides, so called MAB phases, is new types of chemically ordered quaternary borides, *i*-MAB ($M'_{4/3}M''_{2/3}AlB_2$) and *o*-MAB ($M'_4M''SiB_2$), in which the M-atoms are in-plane and out-of-plane chemically ordered, respectively. Both types of phases have been identified in high-throughput simulations followed by experimental synthesis, and they can be chemically exfoliated into 2D sheets, and selectively prepared in multilayer form, as delaminated single-layer sheets in colloidal suspension, or as additive-free filtered films. For example, Ti_4MoSiB_2 *o*-MAB can be used to derive 2D TiO_xCl_y of high yield. $(Mo_{2/3}Y_{1/3})_2AlB_2$ and $(Mo_{2/3}Sc_{1/3})_2AlB_2$ *i*-MAB can be used for realization of so called boridene in the form of single-layer 2D sheets with ordered metal vacancies, $Mo_{4/3}B_{2-x}T_z$(T_z = -F, -O, -OH). The present talk will summarize the results to date of our predictive theoretical approach that have led to 2D materials synthesis from 3D quaternary metal borides, the mechanisms behind the realization of these 3D/2D materials, and evaluation of selected properties.

2:00pm **TS4-ThA-3 Machine Learned Moment Tensor Potentials for Hard Coatings**, ***Ferenc Tasnádi (ferenc.tasnadi@liu.se)***, *F. Bock, M. Odén, I. Abrikosov,* IFM Linköping University, Sweden

Refractory nitrides (TiN, Hf N , NbN etc.) nitride alloys (Ti_{1-x} Al_xN, etc.) or high-entropy alloys, such as TiZrHfTa are materials with high industrial relevance for hard coatings o f cutting tools [1] even for superconducting and plasmonic-based devices. M ajor objectives for their performance are the high-temperature the rmodynamic, dynamic and elastic properties. A recently developed combination of quantum mechanical calculations with machine-learning interatomic potentials (MLIP) [2], is utilized to calculate high temperature properties with high accuracy. On-the-fly training of moment tensor potentials allows us to perform the calculations with more than two orders less computational effort than using state-of-the-art ab initio molecular dynamics simulations. The calculated elastic constants are used to simulate surface acoustic waves and Brillouin light scattering (BLS) spectra. The results are compared with experiments. Furthermore, we investigate high-temperature bcc phase of titanium and predict very weak temperature dependence of its elastic moduli [3], called Elinvar effect, similar to the behavior observed for the so-called GUM metals . The effect in bcc-Ti is intrinsic and therefore unique.

[1] See, for example, F. Tasnádi et al., Phys. Rev. B 85, 144112 (2012); F. Tasnádi et al., Appl. Phys. Lett. 97, 231902 (2010); D. Holec et al. Phys. Rev. B 90, 184106 (2014); F. Tasnádi et al., Mater. Des. 114, 484 (2017); H. Huang et al., Adv. Mater. 5, 1701678 (2017). [2] I. S. Novikov et al., Mach. Learn.: Sci. Technol. 2 025002 (2021). [3] A. Shapeev et al., New. J. Phys. 22, 113005 (2020).

2:20pm **TS4-ThA-4 High-Throughput Rapid Experimental Alloy Development (HT-READ)**, ***Kenneth Vecchio (kvecchio@eng.ucsd.edu)***, UC San Diego, Dept. of NanoEngineering, USA **INVITED**

The development of high-throughput materials development strategies in the thin-film field have moved forward more quickly than bulk material high throughput strategies, primarily due to the need in bulk materials to account for microstructure effects on properties.In addition, the current bulk materials discovery cycle has several inefficiencies from initial computational predictions through fabrication and analyses. Much of the information and knowledge generated existed in isolated data silos making integrated approaches more challenging. This was the motivation for the 2011 Materials Genome Initiative, which sparked advances in many high-throughput computational techniques related to materials development. However, computational techniques ultimately rely on experimental validation.However, bulk materials are generally evaluated in a singular fashion, relying largely on human-driven compositional choices and analysis of the volumes of generated data, thus also slowing validation of computational models. Thus, increasing the rate of materials experimentation is fundamental to improving materials research, and requires parallelizing, automating, and miniaturizing key steps in experimental materials research, including computation, synthesis, processing, characterization, and data analysis.To overcome these limitations, we developed a High-Throughput Rapid Experimental Alloy Development (HT-READ) platform and methodology that comprises an integrated, closed-loop material screening process inspired by broad chemical assays and modern innovations in automation. Our method is a general framework unifying computational identification of ideal candidate materials, fabrication of sample libraries in a configuration amenable to multiple tests and processing routes, and analysis of the candidate materials in a high-throughput fashion. An artificial intelligence agent is used to find connections between compositions and material properties.New experimental data can be leveraged in subsequent iterations or new design objectives. The sample libraries are assigned unique identifiers and stored to make data and samples persistent, thus preventing institutional knowledge loss.This integrated approach paves the way for compositionally accurate and microstructurally informed bulk materials development in a highly-accelerated manner.

3:00pm **TS4-ThA-6 Finding Thermally Robust Superhard Materials with Machine Learning**, ***Jakoah Brgoch (jbrgoch@Central.UH.EDU)***, University of Houston, USA **INVITED**

Superhard materials with a Vickers hardness >40 GPa are essential in applications ranging from manufacturing to energy production. Finding new superhard materials has traditionally been guided by empirical design rules derived from classically known materials. However, the ability to quantitatively predict hardness remains a significant barrier in materials design. To address this challenge, we constructed an ensemble machine-learning model capable of directly predicting load-dependent hardness. The predictive power of our model was validated on eight unmeasured metal disilicides and a hold-out set of superhard materials. The trained model was then used to screen compounds in Pearson's Crystal Data (PCD) set and combined with our recently developed machine-learning phase diagram tool to suggest previously unreported superhard compounds. Finally, industrial materials often experience tremendous heat during application; thus, we are building a method for predicting hardness at elevated temperatures.

3:40pm **TS4-ThA-8 Rational Composition Optimization: Coupling Mixture Designs, Combinatorial Methods and Machine Learning**, ***Elise GAREL (elise.garel@grenoble-inp.fr)***, *H. VAN LANDEGHEM, J. PAROUTY, M. VERDIER, S. COINDEAU, R. MARTIN, F. ROBAUT, R. BOICHOT,* SIMAP, Grenoble-INP, CNRS, France

Multinary optimization has been at the heart of recent developments, either for High Entropy Alloy or for metallic glasses, amongst others. It represents a challenge that requires overcoming the usual method of "one sample at a time". Combinatorial approaches have been applied many times to N-element systems, and this study proposes to couple it to mixture design, in order to guarantee a uniform and systematic screening of the composition space, by elaborating and characterizing magnetron sputtered films with controlled gradients of composition.

This method was applied to the refractory high entropy alloy system Nb-Ti-Zr-Cr-Mo, on as-grown and annealed samples, as well as on a nitride pseudo-ternary, Ti-Al-Nb-N. The mechanical properties were measured by nanoindentation for both, while crystallinity was assessed using XRD — and EBSD in the case of the HEA. Conductivity measurements were performed on the nitride system. This high-throughput screening resulted in an experimental database covering the properties of 460 HEA compositions and 140 nitride compositions that was used to train Machine Learning models linking compositions, structures and properties.

In order to explore the possibilities offered by Machine Learning, several databases and different models were used. Raw experimental data and statistically processed database allowed delineating the performances of Machine Learning. Models with increasing complexity were tested: multilinear regression with interactions, Support Vector Machine, Random Forest and Neural Network, with previously adjust hyper-parameters. Random Forest and Neural Network show a very good accuracy, either on

regression or on classification, for property prediction over the entire composition space. Multilinear regression, performing adequately as well, allows a rapid understanding of the weights of each component on the properties, as well as their interactions, which can be used to rationally examine the largely discussed hypothesis of the cocktail effect. For instance, the elastic modulus of as-grown RHEA system is well fitted by multilinear regression and coefficients show that it almost only results from a linear combination of the effect of pure elements. Hardness presents on the contrary much more complexity, with a high positive effect of binaries and principally a negative effect of ternaries and quaternaries, leading to higher values at the edges of the composition space than at its center.

Based on Machine Learning predictions, isovalue hulls and Pareto-optimal domains can be determined inside the composition space and used to identify the composition sets that show the best property compromises.

4:00pm **TS4-ThA-9 Transfer Learning of Thermodynamic and Elstic Properties of Hard-Coating Alloys**, ***Henrik Levämäki (henrik.levamaki@liu.se)***, *F. Tasnadi, D. Sangiovanni,* Linköping University, IFM, Sweden*; L. Johnson,* Sandvik Coromant, Sweden*; R. Armiento, I. Abrikosov,* Linköping University, IFM, Sweden

Accelerated design of novel hard-coating materials requires state-of-the-art computational tools, which include data-driven techniques, building databases, and training machine learning (ML) models against the databases.

We present a development of a heavily automated high-throughput workflow to build a database of industrially relevant hard coating alloys, such as disordered binary and ternary nitrides [1]. We use Vienna Ab initio Simulation package (VASP) as the density functional theory calculator and the high-throughput toolkit (httk) to automate the calculation workflow.

One of the key quantities in the computational study of hardness is the elastic tensor, and the challenge we face is that calculating the elastic tensor for disordered supercells is resource intensive, which makes building a large database of disordered hard-coating alloys slow. We therefore explore ways for ML techniques to support and complement our databases.

We find that the crystal graph convolutional neural network (CGCNN) model [2] trained on ordered compounds from the Materials Project [3] has sufficient prediction accuracy for the disordered nitrides. This suggests that the existing public or commercial databases provide important data for predicting mechanical properties of qualitatively different types of material systems, which in our case are disordered hard-coating alloys that are not included in the original dataset.

[1] Can be found in arXiv under the title: "Predicting properties of hard-coating alloys using ab-initio and machine learning methods"

[2] Xie, Tian and Grossman, Jeffrey C., "Crystal Graph Convolutional Neural Networks for an Accurate and Interpretable Prediction of Material Properties", Physical Review Letters 120, 145301 (2018), doi: 10.1103/PhysRevLett.120.145301

[3] Jain, Anubhav and Ong, Shyue Ping and Hautier, Geoffroy and Chen, Wei and Richards, William Davidson and Dacek, Stephen and Cholia, Shreyas and Gunter, Dan and Skinner, David and Ceder, Gerbrand and Persson, Kristin A., "Commentary: The Materials Project: A Materials Genome Approach to Accelerating Materials Innovation", APL Materials 011002 (2013), doi: 10.1063/1.4812323

4:20pm **TS4-ThA-10 Data-Driven Search for Thermal Insulators Guided by Anharmonicity: From First Principles to Machine Learning**, ***Florian Knoop (florian.knoop@liu.se)***, Linköping University, IFM, Sweden*; M. Langer,* Technical University of Berlin, Germany*; C. Carbogno,* NOMAD Laboratory at the Fritz Haber Institute of the Max Planck Society, Germany*; M. Rupp,* University of Konstanz, Germany*; M. Scheffler,* NOMAD Laboratory at the Fritz Haber Institute of the Max Planck Society, Germany

We present a systematic first-principles search for thermal insulators in materials space which covers hundreds of compounds, five lattice types and seven space groups, including simple rocksalt and zinc blende structures, up to complex perovskites. Using the high-throughput framework FHI-vibes [1] and a recently developed measure for the strength of anharmonicity [2], we identify 120 candidate materials with potential for low thermal conductivity at room temperature. We investigate the 60 most promising candidates with the ab initio Green Kubo method (aiGK) [3], enabling data-driven extraction of design principles for bulk materials with low thermal conductivity. The aiGK method provides an accurate framework to obtain thermal conductivities for materials, in particular strongly anharmonic ones such as thermal barrier coating ceramics like zirconia, since all anharmonic effects responsible for low thermal conductivity are included.We subsequently demonstrate how the first principles calculations can be complemented with the help of machine learning potentials to remove the computational bottleneck. For this task, we use message passing neural networks, a class of models that can accommodate implicit long-range interactions as well as directional information [4]. We present a systematic account of their performance for calculating the thermal conductivity of solid semiconductors and insulators and discuss implications for high-thoughput heat transport simulations and the discovery of novel thermal insulators.

[1] F. Knoop et al., J. Open Source Softw. 5, 2671 (2020)
[2] F. Knoop et al., Phys. Rev. Mater. 4, 083809 (2020)
[3] C. Carbogno, R. Ramprasad, and M. Scheffler, Phys. Rev. 118, 175901 (2017)
[4] K.T. Schütt et al., J. Chem. Phys. 148 241722 (2018)

4:40pm **TS4-ThA-11 2D Phase Mapping of Hf-Al-Si Refractory Complex Concentrated Alloy Produced using High-Throughput Magnetron Sputtering**, ***Sophia Cooper (sophiacooper@my.unt.edu)***, *M. Dockins, M. Young, A. Voevodin,* University of North Texas, USA*; A. Ghoshal, V. Blair,* U.S. Army Futures Command, USA*; S. Aouadi,* University of North Texas, USA

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