Advanced Characterization, Modelling and Data Science for Coatings and Thin Films
Room Town & Country C - Session CM4-1-MoM
Simulations, Machine Learning and Data Science for Materials Design and Discovery I

Moderator: Davide G. Sangiovanni, Linköping University, Sweden

10:00am **CM4-1-MoM-1 High-Throughput Rapid Experimental Alloy Development (HT-READ)**, ***Kenneth Vecchio (kvecchio@ucsd.edu)***, University of California at San Diego, 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.Furthermore, 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 truly compositionally-accurate and microstructurally-informed bulk materials development in a highly accelerated manner.This overall strategy has enabled our group to achieve the ability to design, fabricate, and fully characterize more than 800 bulk alloy samples per year with a single researcher.

10:40am **CM4-1-MoM-3 Fundamental Investigation for Film Quality Prediction Based on Zone Model in Magnetron Sputtering**, ***Kohei Kuroshima (kohei-kuroshima@osakavacuum.co.jp)***, *I. Ikeda,* Osaka Vacuum, Ltd., Japan*; Y. Gotoh,* Department of Electronic Science and Engineering, Kyoto University, Japan*; M. Iguchi, S. Sugimoto,* Osaka Vacuum, Ltd., Japan

There are diagrams for film structures that are closely related to film properties, called the structure zone model. Thornton's model[1] uses a pressure for one axis, so the model may vary depending on the sputtering apparatus. Anders[2] argued the structure zone diagram (SZD) for the films deposited under presence of energetic particles. In the SZD, the axes are the normalized energy of the particles incident on the substrate, E* and the generalized temperature, T*.

Although it is said that this model cannot be used to predict film structure at points on the E*-T* plane, a trial to predict the structure on the SZD was done in this study. We focused on magnetron sputtering and calculated E* using only the kinetic energy when sputtered particles are incident on the substrate, and with measured substrate temperature, and verified whether Anders' SZD could be applied. We developed a sputter particle transport simulation software and calculated E*. This simulation includes the thermal motion effect of the process gas[3]. For validation, two common types of magnetron sputtering equipment were used to deposit film samples at several combinations of sputtering pressure (0.3-4.0 Pa), target-to-substrate distance (50-100 mm), and substrate temperature (RT-450 ℃). The input power was constant at 300W DC. We observed the surface and cross section of the samples by Scanning Electron Microscope (SEM). It was confirmed whether the structure of the film deposited with the condition which is represented by a certain point on the SZD was consistent with the structure represented by the ZONE.

As a result, under the conditions corresponding to the film structures of ZONE 1 and ZONE 2 on the SZD, the SEM images of the film had the characteristics of each ZONE. On the other hand, under conditions corresponding to the ZONE T, which is the transition region, it was found that the film structure did not show the texture of ZONE T so much, but strongly showed the structure of the adjacent ZONE, especially in the area near the boundaries of the zones.

Following SZD, we are developing a new diagram which exhibits film properties instead of film structure[4].

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References

[1] J. A. Thornton, J. Vac. Sci. Technol. 11, 666 (1974).

[2] A. Anders, Thin Solid Films, 518, 4087 (2010).

[3] T. Nakano, Appl. Surf. Sci., 113/114, 642 (1997).

[4] I. Ikeda et al., International Conference on Metallurgical Coatings & Thin Films 2024, Sandiego, May 19-24, 2024 (submitted).

11:00am **CM4-1-MoM-4 Are ML Potentials Useful to Understand Deformation and Fracture of Ceramics?**, ***Nikola Koutná (nikola.koutna@tuwien.ac.at)***, *S. Lin,* TU Wien, Austria*; L. Hultman,* Linköping University, Sweden*; P. Mayrhofer,* TU Wien, Austria*; D. Sangiovanni,* Linköping University, Sweden **INVITED**

Theoretical understanding of atomic-scale mechanisms underlying deformation and crack growth in ceramics enables rational design of alloys, superlattices, or nanocomposites with optimized combination of hardness and toughness. Simulations represent an important counterpart to experiment, being relatively inexpensive and allowing to impose well-defined loading conditions, thus making fair comparisons within one material class. Certain comprehension of how ceramics behave subject to mechanical loads can be achieved by *ab initio* methods, however, experimentally-relevant predictions require a combination of finite-temperature effects and large-enough models to consider extended crystallographic defects. In this talk I will discuss the exciting and rapidly growing field of machine-learning interatomic potentials (MLIPs) for molecular dynamics and how these can be used to study the onset of fracture. Transition metal diborides and MAB phases (i.e. atomically-thin laminates of ceramic/metallic-like layers) will serve as model materials to showcase a possible training strategy for the MLIP development and challenges upon up-scaling beyond length scales of *ab initio* reach. Furthermore, simulations of crack initiation in TiB_2 as well as the formation of ripplocations in Ta_2AlB_2 and other MAB phases under certain loading conditions will be interpreted in the light of experimental data available via collaborators. The ML potentials will turn out to be quite useful.

11:40am **CM4-1-MoM-6 Impact of TM Elements on Structural, Thermodynamic and Mechanical Properties of CrN**, ***David Holec (david.holec@unileoben.ac.at)***, Montanuniversität Leoben, Austria*; P. Mayrhofer,* TU Wien, Institute of Materials Science and Technology, Austria

CrN belongs to a family of transition metal nitrides used as protective coatings in automotive, aerospace, tooling and other applications. It poses a specific challenge for *ab initio* modelling due to its genuine magnetic properties: it has an antiferromagnetic (AFM) orthorhombic structure below the Néel temperature (T_N~room temperature) and adopts paramagnetic (PM) cubic B1 above T_N.

In this contribution, we use *ab initio* calculations to study the alloying impact of TM elements (Sc, Y, Ti, Zr, Hf, V, Nb, Ta, Mo, W) on CrN in its application-relevant PM-B1 phase. Apart from V, all other elements increase the lattice parameter of CrN consistently with the corresponding binary TMNs. The enthalpy of isostructural mixing is the largest for Y due to large internal strains, followed by the group IVB elements. Interestingly, $H_{mix}$is negligible in the whole compositional range for Sc and V and becomes even negative (stable solid solutions) for $Cr_{1-x}Ta_xN$. Nb, W and Mo exhibit more complicated, composition-dependent behaviour. As quantified by the *B/G* ratio and the Cauchy pressure (C_{12}-C_{44}), the mechanical properties

strongly correlate with the valence electron concentration of the ternary $Cr_{0.89}TM_{0.11}N$ solid solutions. While Mo and W are the most potent ductility enhancers, Sc and Y are predicted to embrittle CrN.

12:00pm **CM4-1-MoM-7 Machine-Learning Potential for Accurate Predictions of Elastic Properties in Amorphous W-B-C**, ***Pavel Ondracka (ondracka@mail.muni.cz)***, *J. Ženíšek,* Masaryk University, Czechia*; G. Nayak,* RWTH Aachen University, Germany*; D. Holec,* Montanuniversität Leoben, Austria*; P. Vašina,* Masaryk University, Czechia

Amorphous tungsten boron carbide is a prospective material for protective coatings, with superior ductility and crack resistance [1], and yet the subtle details of its atomistic structure and origin of its excellent mechanical properties are still unclear. Due to the small sizes of representative models and limited timescales of *ab initio* molecular dynamics based on density functional theory, it is difficult for this standard methodology to reliably predict structural and mechanical properties of amorphous W-B-C. Such predictions lead to strongly anisotropic mechanical properties and large uncertainties in the results. We solved the issues by fitting interatomic potential in a general nonlinear atomic cluster expansion (ACE) form [2] to this material system using an active learning approach to sample the amorphous configuration space and the D-optimality criterion and MaxVol algorithm to efficiently construct the training set [3]. The potential was trained for the W content between 10 and 60 at. % and C:B ratios between 4:1 and 1:4. Subsequently, we employed a melt & quench procedure to generate amorphous structural models containing more than 10000 atoms which yielded an isotropic mechanical response and revealed trends with respect to the system composition, density and quenching rates. The thus obtained values of Young's modulus were successfully validated against experimental data.

[1] S. Mirzaei et al., Surface and Coatings Technology 383, 125274 (2020).
[2] Y. Lysogorskiy et al., npj Computational Materials 7, (2021).
[3] Y. Lysogorskiy et al., Physical Review Materials 7, (2023).

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