Advanced Characterization, Modelling and Data Science for Coatings and Thin Films
Room Palm 5-6 - Session CM4-2-TuM

Simulations, Machine Learning and Data Science for Materials Design and Discovery II

Moderators: Po-Liang Liu, National Chung Hsing University, Taiwan , **Ferenc Tasnadi**, Linköping University, Sweden

8:00am **CM4-2-TuM-1 DFT + ML + Calphad: From Qualitative to Quantitative Phase Stability Predictions***, **Moritz to Baben (mtb@gtt-technologies.de)**, P. Keuter, C. Früh, B. Reis, F. Tang,* GTT-Technologies, Germany **INVITED**

Today, phase stability predictions using quantum mechanical calculations can be considered state-of-the-art for metallurgical coatings and thin films. However, these predictions are usually qualitative in nature, partly because of missing data and partly because the processes are complex and not in thermodynamic equilibrium.

Here, it is shown how

(I) data gaps concerning phase stability can be closed by a the combination of 0 K DFT calculations, machine learning to cheaply extend the data validity to relevant temperatures and Calphad methodology to describe phase stability of solid solutions and thus solid-gas equilibria.

(II) phase stability data can then be used to make quantitative predictions for magnetron sputtering, i.e. a process that is usually considered to be far from equilibrium, using the para-equilibrium approach (nitrogen stoichiometry of TiAlN, to Baben et al., MRL 5 (2017) 158) or using a small process model based on the Hertz-Knudsen equation (stoichiometry of Mg-Ca Thin Films, Keuter et al, Materials 16 (2023) 2417).

8:40am **CM4-2-TuM-3 Cu-Zr-Al Thin Film Metallic Glasses in a Wide Range of Compositions and Growth Conditions***, **Jiri Houska (jhouska@kfy.zcu.cz)**, P. Zeman,* University of West Bohemia, Czechia

Cu-Zr-Al thin film metallic glasses are investigated by a combination of simulations of their atom–by-atom growth with magnetron sputtering. We fulfill all requirements which maximize the usefulness of the results: mutual support of calculated and experimental data, simulation algorithm which exactly reproduces what is happening in the experiment, wide compositional range (from pure Cu to pure Zr and from [Al] = 0% to 20%), wide range of growth conditions (energy of arriving atoms, temperature, growth template). We focus on the homogeneity, densification, short-range order (bonding preferences and coordination numbers), medium-range order (common neighbor and network ring statistics) and functional properties. Special attention is paid to the key building blocks of Cu-Zr-Al: not only icosahedral clusters (12 vertices) but also newly identified supraicosahedral clusters (16 vertices).

First, we identify crystalline Zr-rich compositions (on any growth template) and Cu-rich compositions (with a strong effect of growth template), and glasses (as homogeneous as what result from a random distribution of atoms) at [Cu] = 20% to 80-85%. Increasing [Cu] in the glassy compositions leads to increasing coordination of both Cu and Zr, packing factor and icosahedron-like medium-range order. Second, increasing [Al] in glassy $Cu_{0.46}Zr_{0.54-x}Al_x$ preserves the homogeneity (at a very low preference to form Al-Al bonds) and once again leads to increasing coordination of all elements, packing factor and concentration of icosahedral clusters (around smaller Cu and Al) and supraicosahedral clusters (around larger Zr). All of that is achievable at low energy delivered into the growing films, while delivering too much energy (by energetic bombardment or by ohmic heating) may be even harmful.

While the atomic-scale simulations provide a lot of information not accessible experimentally, they are correlated with and explain experimental data including increasing hardness, Young's modulus, glass transition temperature and crystallization temperature with increasing [Cu]/[Zr] and [Al]/[Zr]. Collectively, the results [1,2] are important for understanding the structures and properties of this class of metallic glasses, and for optimizing their compositions and pathways for their preparation.

[1] J. Houska, P. Machanova, M. Zitek, P. Zeman, J. Alloys Compd. 828, 154433 (2020)

[2] J. Houska, P. Zeman, Comp. Mater. Sci. 222, 112104 (2023)

9:00am **CM4-2-TuM-4 Transformation Plasticity and Fracture in MB_2 (M=Ti, Ta, W, Re) Diborides via Ab-Initio and Machine-Learning-Potential Molecular Dynamics***, **Shuyao Lin (shuyao.lin@tuwien.ac.at)**,* TU Wien, Institute of Materials Science and Technology, Austria*; T. Leiner,* Montanuniversität Leoben, Leoben, Austria*; Z. Chen,* Austrian Academy of Sciences, Austria*; R. Janknecht,* TU Wien, Institute of Materials Science and Technology, Austria*; F. Tasnadi,* Linköping University, Sweden*; Z. Zhang,* Austrian Academy of Sciences, Austria*; L. Hultman,* Linköping University, Sweden*; P. Mayrhofer,* TU Wien, Institute of Materials Science and Technology, Austria*; D. Holec,* Montanuniversitat Leoben, Austria*; D. Sangiovanni,* Linköping University, Sweden*; N. Koutná,* TU Wien, Institute of Materials Science and Technology, Austria

In this contribution we employ ab-initio molecular dynamics (AIMD) and machine learned interatomic potential molecular dynamics (ML-MD) simulations to elucidate trends and typical patterns in the mechanical response of transition metal diborides. Four representative diboride systems, MB_2, are selected, with M from the group IV (Ti), V (Ta), VI (W), and VII (Re) of the periodic table. The AIMD simulations serve to find finite-temperature equilibrium lattice parameters of the chosen diborides and to estimate their tensile and shear response at the atomic scale. The thereby produced *ab initio* dataset is used to fit and validate machine-learning interatomic potentials for ML-MD (within the moment tensor potential, MTP, formalism), providing a basis to study deformation behavior at the nanoscale. By controlling the phase structure (the AlB_2, WB_2, and ReB_2-prototype phase), supercell size (few to dozens of nm^3), and imposing well-defined loading conditions (tensile or shear deformation with various loading directions and temperatures), our ML-MD simulations allow assessing similarities as well as fundamental differences between the studied diborides. Considering a nanoscale model with a pre-indent on the surface, we go one step further and discuss ML-MD predictive power and limitations in the light of experimental results for an indented TiB_2 thin film.

9:20am **CM4-2-TuM-5 Fracture Toughness: Atomistic Understanding of Directional and Temperature Dependence for the case of $Ti_{1-x}Al_xN_y$***, **Davide Sangiovanni (davide.sangiovanni@liu.se)**,* Linköping University, Sweden

The fracture toughness (K_{Ic}) of single-crystal lattices and interface structures is a physical property that depends on temperature and crystallographic orientation. For ceramic thin films, experimental characterization of K_{Ic} is complicated by the presence of grain boundaries or structural inhomogeneities. Narrow scatter among measured K_{Ic} values (1-to-5 MPa $\sqrt{m}$), combined to relatively large statistical uncertainties (±1 MPa $\sqrt{m}$), vanifies attempts to rank different hard ceramics according to their effective fracture resistance.

Taking B1-structure $Ti_{1-x}Al_xN_y$ as representative ceramic systems, I present results of atomistic fracture-mechanics simulations carried out at different temperatures (*T*) and for diverse crystallographic orientations of the fracture plane (hkl) / crack front [h'k'l']. The approach — based on K-controlled *nanoscale* loading, implemented with anisotropic *T*-dependent elastic responses **[1]** — can reliably forecast *observable* mechanical responses.

Direct atomistic observations of localized transformation-induced or slip-induced plasticity in flawed $Ti_{1-x}Al_xN_y$ lattices allow understanding and quantifying the impact of small-scale yielding on K_{Ic} and fracture strength values calculated as a function of *T* and (hkl)[h'k'l']. Moreover, the simulation results evidence limits of Griffith (^{G}) and Rice (^{R}) criteria for predictions of stress intensities that lead to brittle-fracture (K_{Ic}^G) and dislocation emission (K_{Ie}^R). Alternative descriptors — based on properties evaluated by homogeneous deformation of defect-free crystals **[1,2]** — are proposed as convenient means to rapidly screen mechanical strength, tendency to undergo plastic deformation, and fracture resistance at any temperature of interest.

The talk will also briefly cover our recent developments in machine-learning interatomic potentials for cutting-edge description of materials subject to deformation at realistic conditions **[3]** and *ab initio* database of ceramic properties computed from 0 K to elevated temperatures **[4]**.

[1] Physical Review Materials (2023) https://doi.org/10.1103/PhysRevMaterials.7.103601
[2] Science Advances (2023) https://doi.org./10.1126/sciadv.adi2960
[3] Preprint (2023) https://doi.org/10.48550/arXiv.2309.00996
[4] npj Computational Materials (2022) https://doi.org/10.1038/s41524-022-00698-7

9:40am **CM4-2-TuM-6 Exploring Surface Energy and Work Function Changes in ZnGa2O4(111) via Ab Initio Studies***, Po-Liang Liu* ***(pliu@dragon.nchu.edu.tw)****, Y. Lin,* National Chung Hsing University, Taiwan

The metal oxide semiconductor gas sensor holds promise as the primary component for environmental monitoring within artificial intelligence-based systems designed for household and industrial gas detection. The fabrication of ZnGa2O4 thin films has been advanced due to their capacity to operate within sensor temperature ranges and discern the composition and concentration of mixed gases. The n-type semiconductor nature of ZnGa2O4 enables the detection of NO2 and H2S molecules. This semiconductor exhibits rapid and robust sensing responses along with high signal intensity towards NO2 and H2S, thereby anticipating an enhancement in sensor operational efficiency, particularly in terms of elevated temperature utilization. Hence, this study employs ab initio calculations based on the density functional theory to determine the surface energy of ZnGa2O4(111). The analysis reveals that the ZnGa2O4(111) surface comprises Ga, Zn, and O elements. Findings indicate that the surface energy for Zn-Ga-O-, O-, and Ga-terminated ZnGa2O4(111) range between 0.0516 to 0.2335 eV/Å2, 0.0516 to 0.7789 eV/Å2, and 0.0464 to 0.5918 eV/Å2, respectively. The Ga-terminated ZnGa2O4 has the lowest surface energy of 0.0464 eV/Å2 in a Ga-rich environment, showing the Ga-terminated ZnGa2O4(111) is the most favorable surface. The work function change of Zn-Ga-O-, O-, and Ga-terminated ZnGa2O4(111) are 3.70 eV, 0.48 eV, and 6.35 eV, respectively. This highlights that the Ga surface atoms demonstrate a maximum work function change, consistent with previously experimental observations

Author Index

Bold page numbers indicate presenter

— C —
Chen, Z.: CM4-2-TuM-4, 1
— F —
Früh, C.: CM4-2-TuM-1, 1
— H —
Holec, D.: CM4-2-TuM-4, 1
Houska, J.: CM4-2-TuM-3, **1**
Hultman, L.: CM4-2-TuM-4, 1
— J —
Janknecht, R.: CM4-2-TuM-4, 1
— K —
Keuter, P.: CM4-2-TuM-1, 1
Koutná, N.: CM4-2-TuM-4, 1
— L —
Leiner, T.: CM4-2-TuM-4, 1
Lin, S.: CM4-2-TuM-4, **1**
Lin, Y.: CM4-2-TuM-6, 2
Liu, P.: CM4-2-TuM-6, **2**
— M —
Mayrhofer, P.: CM4-2-TuM-4, 1
— R —
Reis, B.: CM4-2-TuM-1, 1
— S —
Sangiovanni, D.: CM4-2-TuM-4, 1; CM4-2-TuM-5, **1**
— T —
Tang, F.: CM4-2-TuM-1, 1
Tasnadi, F.: CM4-2-TuM-4, 1
to Baben, M.: CM4-2-TuM-1, **1**
— Z —
Zeman, P.: CM4-2-TuM-3, 1
Zhang, Z.: CM4-2-TuM-4, 1