# Accelerated Discovery of New Materials

787. WE-Heraeus-Seminar

15 – 18 May 2023

at the Physikzentrum Bad Honnef, Germany



#### Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

#### Aims and scope of the 787. WE-Heraeus-Seminar:

The development of new materials is critical for all industrialized societies, as it touches upon virtually all technology relevant application fields including energy, efficient use of resources and devices for information technologies.

One of the major challenges in this field is the sheer number of conceivable materials, which is far more than can ever be experimentally realized and characterized. Digitalization strategies for materials research, which blend advanced computational approaches with sophisticated experimental techniques, offer a way forward. Traditional trial and error research needs to be replaced by computer-based simulations carried out in parallel with novel experimental processes.

In this seminar, we will put a main - but not exclusive - focus on two novel classes of materials: "high entropy alloys" (HEA) and "metal organic frameworks" (MOFs). HEAs are alloys with roughly equal proportions of five or more elements, yielding e.g. mechanical properties that are often substantially different from conventional alloys. HEA oxides display interesting properties in catalysis and energy storage. MOFs are porous, crystalline compounds where organic linkers connect metal nodes. Originally developed for gas storage and separation, recently MOFs have found widespread applications in optoelectronics and sensorics. We will also not exclude other materials - in particular perovskites and battery materials.

The program will include a series of lectures by leading researchers from experiment, theory, and computer science. Students and early career researchers are encouraged to participate with posters and flash presentations. To facilitate trans- and interdisciplinary exchange we will hold open discussion sessions on a variety of topics.

#### **Scientific Organizers:**

Prof. Dr. Christof Wöll	Karlsruher Institut für Technologie, Germany E-mail: christof.woell@kit.edu
Prof. Dr. Martin Aeschlimann	Universität Kaiserslautern, Germany E-mail: ma@physik.uni-kl.de
Prof. Dr. Roser Valentí	Goethe-Universität Frankfurt am Main, Germany E-mail: valenti@ito.uni-frankfurt.de

### Introduction

#### Administrative Organization:

Dr. Stefan Jorda Elisabeth Nowotka Marion Reisinger	Wilhelm und Else Heraeus-Stiftung Kurt-Blaum-Platz 1 63450 Hanau, Germany Phone +49 6181 92325-12 Fax +49 6181 92325-15 E-mail nowotka@we-heraeus-stiftung.de Internet: www.we-heraeus-stiftung.de
<u>Venue:</u>	Physikzentrum Hauptstrasse 5 53604 Bad Honnef, Germany Conference Phone +49 2224 9010-120 Phone +49 2224 9010-113 or -114 or -117 Fax +49 2224 9010-130 E-mail gomer@pbh.de Internetwww.pbh.de Taxi Phone +49 2224 2222
<u>Registration:</u>	Marion Reisinger (WE Heraeus Foundation) at the Physikzentrum, reception office Monday (10:00 h – 15:00 h)

#### Monday, 15 May 2023

11:00 – 15:00	Registration	
12:30 – 13:45	LUNCH	
14:00 – 14:15	Scientific organizers	Opening remarks
14:15 – 15:00	Claudia Draxl	The FAIRmat approach to accelerated discovery of new materials
15:00 – 15:30	COFFEE BREAK	
15:30 – 16:15	Pascal Friederich	Machine learning for simulation, understanding and design of molecules and materials
16:15 – 17:00	Jonathan Schmidt	Machine learning discovery of materials
17:00 – 17:45	Manuel Tsotsalas	Accelerated MOF synthesis via automated data extraction and machine learning
18:30 – 20:00	DINNER	
20:00 – 20:45	Berend Smit	Big data in chemistry and chemical engineering: Science beyond understanding
20:45 – 21:00	Hartmut Gliemann	Present status and future of fully automated production and application of surface-anchored metal-organic framework (SURMOF) based coatings at IFG – an overview
21:00 – 21:15	Dinga Wonanke	A step towards predicting synthesis conditions of metal-organic frameworks
21:15 – 21:30	Qiang Zhang	Exploring the Mechanical Properties of a 2D MOF: A Combined MD Simulation and Experimental Study

#### Tuesday, 16 May 2023

08:00	BREAKFAST	
09:00 – 09:45	Larry Lüer	Autonomous discovery of semiconducting materials for photovoltaics
09:45 – 10:30	Seda Keskin	Computational modeling of MOFs for gas separations
10:30 – 11:00	COFFEE BREAK	
11:00 – 11:45	Regine Willumeit- Römer	Towards a digital twin for degradable Mg-implants
11:45 – 12:30	Ziyuan Rao	Machine learning-enabled high-entropy Invar alloy discovery
12:30– 12:40	Conference Photo (in	n the front of the lecture hall)
12:40 – 14:00	LUNCH	

#### Tuesday, 16 May 2023

14:00 – 14:15	Anna Kornyushchenko	Formation of porous metal nanosystems under near-equilibrium condensation conditions in plasma- condensate system
14:15 – 14:30	Tomasz Ossowski	Superconductivity in high entropy alloys
14:30 – 14:45	Andreas Terfort	Parallel electrochemical investigation of combinatorial catalytic system
14:45 – 15:30	Poster Flash 1 (poster	presenters will be announced)
15:30 – 16:00	COFFEE BREAK	
16:00 – 18:00	Poster session 1	
18:30 – 20:00	DINNER	
20:00 – 20:45	Giorgio Sangiovanni	Dirac fermions on the triangular lattice
20:45 – 21:00	Brian Richard Pauw	Synthesizing a library of 1000+ reproducible MOFs

#### Wednesday 17 May 2023

#### 08:00 BREAKFAST

09:00 – 09:15 Roland A Fischer

Structure-activity relationships in metal-organic frameworks' conversion for Oxygen evolution reaction

- 09:15 10:00 **Poster flash 2** (poster presenters will be announced)
- 10:00 10:30 COFFEE BREAK
- 10:30 12:30 **Poster session 2**
- 12:30 14:00 LUNCH
- 14:00 17:00 **Excursion**
- 18:30 HERAEUS DINNER (social event with cold & warm buffet with complimentary drinks)

#### Thursday, 18 May 2023

08:00	BREAKFAST	
09:00 – 09:45	Helge Stein	Progress, prospects and the future of autonomous chemistry
09:45 – 10:30	Axel Groß	The use of descriptors in the accelerated discovery of battery materials with improved properties
10:30 – 11:00	COFFEE BREAK	
11:00 – 11:45	Wolfgang Wenzel	Virtual materials design
11:45 – 12:30	Scientific organizers	Poster prize and closing remarks
12:30	LUNCH	

#### End of the seminar and departure

NO DINNER for participants leaving on Friday; however, a self-service breakfast will be provided on Friday morning

## Posters

		Posters
1	Thomas Ackstaller	Automated ink sampling for high-throughput research on ink-based electrocatalysts
2	Gun Deniz Akkoc	Accelerated optimization of water-splitting materials via automation
3	Mushtaq Ali	Improved solubility prediction on most diverse dataset
4	Katharina Bleher	Immobilization of enzyme models in zeolites
5	Ben Breitung	High-throughput techniques and applications for multi-component mofs for electronic device applications
6	Hilal Aybike Can	Robot-accelerated material development for perovskite-based multijunction solar cells
7	Jiangming Cao	Design guidelines for two-dimensional transition metal dichalcogenide alloys
8	Christian Clausen	High-entropy alloys for catalyst discovery
9	Silvia Cristofaro	Mechanical response to strain of semiconducting polymers
10	Nathan Daelman Jose Maria Pizarro Blanco	FAIR materials science research using NOMAD
11	Johannes Döhn	Computational screening of oxide perovskites as insertion-type cathode materials
12	Joshua Edzards	Structure-property relationships in functionalized metal organic framework from first-principles high-throughput screening

#### Posters

13	Aaron Flötotto	Surfaces and interfaces of semiconductor systems for solar energy conversion
14	Kristýna Gazdová	Magnetism in TiO $_2$ from ab initio perspective
15	Malte Grunert	Search for novel optical materials
16	Vasyl Haramus	Application of Ti and Mg hybrid material as partly degradable implant
17	Arthur Hardiagon	High-throughput screening workflow combining synthesis conditions and physicochemical properties of MOFs for gas separation applications
18	Noor Titan Putri Hartono	Perovskite solar cells' ageing test data clustering
19	Mohamed Hendy	Alchemical perturbation density functional theory for accelerated screening of high-entropy alloys catalysts
20	Aysenur Iscen	Investigation of structure-property relationships in amyloid-like supramolecular peptide nanofibrils
21	Ali Ismael	Green energy harvesting in endohedral- fullerene, single-molecule junctions
22	Mehrdad Jalali	Social network analysis (SNA) in material science for the prediction of material properties
23	Jan G. Korvink	Efficient NMR screening of chemical parameter spaces
24	Denis Kramer	Integrated design methodology for advanced functional materials

#### Posters

25	Aakash Kumar	2D Heterostructure design of novel energy storage devices
26	Yi Luo	MOF synthesis prediction enabled by automatic data mining and machine learning
27	Neil MacKinnon	Nuclear magnetic resonance supports accelerated materials discovery
28	Lukas Meier	The H@GB puzzle
29	Artem Musiienko	Unravelled 143-year secret in physics gives access to explore charge transport parameters and limiting factors of the materials unreachable otherwise
30	Carsten Natzeck Lena Pilz	Machine learning based multi-parameter and multi-objective optimization in metal-organic framework thin films
31	Salih Okur	SURMOF e-tongue: Highly stable functionalized UiO-66-X sensor array for discrimination in liquid
32	Dhanshree Pandey	Cluster expansion extended to W-Ta- vacancy multi-component alloy
33	Matthias A. Popp	The squeezable nanojunction: A multi- physics experimental platform for accelerated geometry manipulation of nano- scale systems
34	Holger-Dietrich Saßnick	Bulk and surface properties of Cs-based photocathode materials via high-throughput density functional theory calculations
35	Shabbir Tahir	Exploring the potential of laser ablation in liquid for high-throughput synthesis of CoCrFeMnNi nanoparticles

		Posters
24		Functional design and investigation of Mar
30	Znixuan vvei	ion conductors for solid-state Mg batteries
37	Alexander Wieczorek	Accelerating the development of stable vapor-deposited perovskite thin-films via combinatorial UV–Vis degradation studies

## **Abstracts of Talks**

(in alphabetical order)

# The FAIRmat approach to accelerated discovery of new materials

#### **Claudia Draxl**

Department and IRIS Adlershof, HU Berlin, Berlin Germany

The enormous amounts of research data produced every day in the field of materials science represent a gold mine of the 21st century. How can we turn these data into knowledge and value? Here, a FAIR (Findable, Accessible, Interoperable, and Re-usable) data infrastructure plays a decisive role as this gold mine is of little value if the data are not comprehensively characterized and made available. Only then, data can be readily shared and explored by data analytics and artificial-intelligence (AI) methods. Making data Findable and AI Ready (another interpretation of the acronym) will change the way how science is done today.

In this talk, I will discuss how the NFDI consortium FAIRmat [1] is approaching these goals [2], making data from sample synthesis, various experimental probes, and computational materials science FAIR. A particular emphasis will be on the *I*, the interoperability. With selected examples, I will also show how knowledge can be gained from these data, be it with unsupervised [3] or supervised [4] machine-learning techniques.

- [1] https://fairmat-nfdi.eu
- M. Scheffler, M. Aeschlimann, M. Albrecht, T. Bereau, H.-J. Bungartz, C. Felser, M. Greiner, A. Groß, C. Koch, K. Kremer, W. E. Nagel, M. Scheidgen, C. Wöll, and C. Draxl, Nature 604, 635 (2022).
- [3] M. Kuban, S. Gabaj, W. Aggoune, C. Vona, S. Rigamonti, and C. Draxl, MRS Bulletin **47**, 991 (2022).
- [4] T. Bechtel, D. Speckhard, J. Godwin, and C. Draxl, preprint.

#### Structure-Activity Relationships in Metal-Organic Frameworks' Conversion for Oxygen Evolution Reaction

#### **Roland A. Fischer**

Technical University of Munich; TUM School of Natural Sciences, Department of Chemistry, Chair of Inorganic and Metal-Organic Chemistry; Catalysis Research Center (CRC), Garching, Germany

Metal-organic frameworks (MOFs) have been reported to catalyze the oxygen evolution reaction (OER).[1] Of prime importance appears the choice of MOFs and how their compositions relate to the catalyst stability and in turn to the reconstruction mechanisms to the active species under OER conditions.[2-3] An isoreticular series of Ni-MOFs [Ni<sub>2</sub>(OH)<sub>2</sub>L] was chosen to elucidate the effects of the carboxylate linker length expansion and modulation of the linker-linker pi-pi interactions. (L = 1,4benzodicarboxylate, 2,6-napthalenedicarboxylate, biphenyl-4,4'-dicarboxylate and pterphenyl-4,4"-dicarboxylate). The linker controls the transformation of Ni-MOF into distinct nickel hydroxide phases, and the conversion from alpha-Ni(OH)<sub>2</sub> to beta-Ni(OH)<sub>2</sub>, thus correlating the Ni-MOFs composition with the OER activity of the Ni-MOF-derived metastable nickel hydroxide phase mixture. We will report on screening the parameter space of the described system. In particular, we found a stabilization effect on the Ni-MOF structure combined with an spectacular increase of the electrocatalytic performance data (current densities, overpotential, mass and area specific activities) when an excess of linker was added to the alkaline electrolyte. Opportunities to translate the so-far linear experimental work-flow into a parallel, high throughput version will be discussed.

- S. Yuan, J. Peng, B. Cai, Z. Huang, A. T. Garcia-Esparza, D. Sokaras, Y. Zhang, L. Giordano, K. Akkiraju, Y. G. Zhu, R. Hübner, X. Zou, Y. Román-Leshkov, Y. Shao-Horn, Nature Materials **21**, 673 (2022)
- S. Hou,W. Li, S. Watzele, R. M. Kluge, S. Xue, S. Yin, X. Jiang, Markus Döblinger, A. Welle, B. Garlyyev, M. Koch, P. Müller-Buschbaum, C. Wöll, A. S. Bandarenka, R. A. Fischer, Adv. Mater. 33, 2103218 (2021)
- [3] W. Li, S. Xue, S. Watzele, S. Hou, J. Fichtner, A. L. Semrau, L. Zhou, A. Welle, A. S. Bandarenka., R. A. Fischer, Angew. Chem. Int. Ed. 59, 5837 (2020)

#### Machine Learning for Simulation, Understanding and Design of Molecules and Materials

Pascal Friederich<sup>1,2</sup>

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Machine learning can accelerate the screening, design and discovery of new molecules and materials in multiple ways, e.g. by virtually predicting properties of molecules and materials, by extracting hidden relations from large amounts of simulated or experimental data, or even by interfacing machine learning algorithms directly with for autonomous decision-making automated high-throughput experiments. In this talk, I will focus on our research activities on graph neural networks for property prediction [1,2] and understanding of structure property relations [3], as well as on the use of graph neural networks for accelerated atomistic simulations [4,5]. Application areas range from superconductors, metal-organic frameworks and organic semiconductors to photochemical reactions. Furthermore, I will go beyond virtual design and simulations, and give a brief outlook on the use of machine learning methods for data-analysis and decision-making processes in automated materials science and chemistry labs [6,7].

- [1] Reiser *et al.*, Communications Materials **3** (1) (2022), https://www.nature.com/articles/s43246-022-00315-6
- [2] KGCNN library, https://github.com/aimat-lab/gcnn\_keras
- [3] Teufel et al., NeurIPS Workshop (2022), https://arxiv.org/abs/2211.13236
- [4] Friederich *et al.*, Nature Materials **20** (6) (2021), https://www.nature.com/articles/s41563-020-0777-6
- [5] Li *et al.*, Chemical Science **12** (2021), https://pubs.rsc.org/en/content/articlehtml/2021/sc/d0sc05610c
- [6] Luo *et al.*, Angewandte Chemie International Edition **61** (19) (2022), https://onlinelibrary.wiley.com/doi/full/10.1002/anie.202200242
- [7] Velasco *et al.*, Advanced Materials **33** (43) (2021), https://onlinelibrary.wiley.com/doi/full/10.1002/adma.202102301

#### Present status and future of fully automated production and application of surface-anchored metal-organic framework (SURMOF) based coatings at IFG – an overview

#### H. Gliemann, C. Wöll

Institute of Functional Interfaces (IFG), Karlsruhe Institute of Technology (KIT), Eggenstein-Leopoldshafen, Germany

Surface-anchored metal-organic frameworks (SURMOFs) represent an innovative material platform consisting of organic linker molecules coordinated to inorganic nodes by forming highly porous and crystalline materials. SURMOFs can be used as molecular filter membranes, host materials for functional molecules (luminescence, conductivity) or are per se functional materials in terms of optically and catalytically active materials [1]. The SURMOF pores can be tailored in terms of size, shape and physicochemical properties (hydrophobic/hydrophilic, polar/none-polar etc.) by choosing the corresponding linker molecules. As SURMOFs are grown on supporting substrates (metals, oxidic materials, glass, polymers) in a layer-by-layer approach, the thickness of the SURMOF layers can be exactly controlled on the nm scale, depending on the numbers of deposition cycles. Here we present the strategy of fully automated robot-supported layer-by-layer (LBL) growth of SURMOFs and some selected application examples. In this context we point out the development starting from the classical LBL method via first attempts in implementing evaluation algorithms, to optimize experimental parameters to the future plans, where we will use artificial intelligence combined with machine learning approaches to setup a fully autonomous robot coating system producing SURMOFs with optimized properties including e.g. low roughness, high crystallinity or homogenous layer thickness [2]. The activities are flanked by the consequent use of different electronic lab notebooks (ELNs), where the experimental preparation parameters and analysis data are archived in a findable manner, so that we can use it for future data mining and to identify correlations between preparation parameters and SURMOF properties.

- [1] D.-H. Chen, H. Gliemann, C. Wöll, Chem. Phys. Rev. 4, 011305 (2023)
- [2] L. Pilz, C. Natzeck, J. Wohlgemuth, N. Scheuermann, P.G. Weidler, I. Wagner, C. Wöll, M. Tsotsalas, Adv. Mater. Interfaces 10, 220177 (2023)

# The use of descriptors in the accelerated discovery of battery materials with improved properties

#### Axel Groß<sup>1,2</sup>

<sup>1</sup>Institute of Theoretical Chemistry, Ulm University, 89069 Ulm/Germany <sup>2</sup> Helmholtz-Institute Ulm (HIU), 89081 Ulm/Germany

Currently, the market for high-performance batteries for stationary and mobile applications is dominated by the Li-ion battery technology. However, the high demand for Li-ion batteries leads to challenges and risks, in particular as far as the availability of the raw materials for these type of batteries is concerned. Hence there is a growing interest in alternative battery technologies, in which Li is replaced by alternative charge carriers such as Na, K, Mg, Ca or Al. However, most of these alternative battery types require an entirely new design with respect to their components such as electrodes and electrolytes. The search for such components necessitates also increased fundamental research efforts with respect to basic scientific aspects of materials properties.

In this talk, I will focus on atomistic studies based on first-principles electronic structure calculations aiming at a fundamental understanding of battery properties. I will particularly focus on so-called descriptors which represent correlations between some basic materials properties and desired or non-desired functional properties of these materials [1,2]. Once reliable descriptors are established, they can significantly speed up the search for new materials with promising properties. This concept will be applied to the understanding of dendrite growth in batteries which represents the main cause for battery fires [1]. Furthermore, the mobility of charge carriers in electrodes and electrolytes will be addressed which is a critical performance parameter of batteries [2]. In addition, further data-driven computational studies aiming at the accelerated discovery of battery materials will be presented [3].

- M. Jäckle, K. Helmbrecht, M. Smits, D. Stottmeister, A. Groß, Self-Diffusion Barriers: Possible Descriptors for Dendrite Growth in Batteries?, Energy Environ. Sci. 11, 3400-3407 (2018)
- [2] M. Sotoudeh and A. Groß, Descriptor and scaling relations for ion mobility in crystalline solids, JACS Au **2**, 463–471 (2022)
- [3] M. Dillenz, M. Sotoudeh, C. Glaser, J. Janek, A. Groß, H. Euchner, Unravelling charge carrier mobility in d0-metal-based spinels, Batteries Supercaps 5, e202200164 (2022)

#### Computational Modeling of MOFs for Gas Separations

#### S. Keskin

Koç University, Istanbul, Turkey

We have witnessed the quick growth of metal organic frameworks (MOFs) and covalent organic frameworks (COFs), porous materials with multifunctional properties offering great potentials for a large variety of chemical applications, in the last two decades. We already know millions of different types of MOFs and COFs, either synthesized or computer-generated. The number of MOFs and COFs increases at an unpredictable rate, and it is very challenging to test every single material for all possible applications using trial-and-error experiments. Thus, computational methods that can accurately evaluate the properties and potentials of MOFs and COFs in different applications are very valuable. High-throughput computational screening based on molecular simulations has been widely used to study MOFs and COFs as adsorbents, membranes, sensors etc. Molecular simulations enable researchers to screen large number of materials in a time-efficient manner to identify the highperforming materials for a target application and detailed analysis of the molecular features of the promising materials leads to the design and discovery of new materials. A recent approach is to integrate data science, specifically machine learning to molecular simulations, to better understand complex and very large structure-performance data obtained from simulations. In this talk, I will describe the state-of-the art in molecular simulations and machine learning modeling of MOFs for gas storage and gas separation applications by discussing various examples from our recent studies such as computational discovery of the most promising MOF adsorbents, MOF membranes, and MOF-based mixed matrix membranes for gas separations in addition to addressing the opportunities and challenges in the field of computational modeling of MOFs.

#### Formation of porous metal nanosystems under nearequilibrium condensation conditions in plasmacondensate system

## <u>A. Kornyushchenko<sup>1,2</sup>, V. Perekrestov<sup>2</sup>, Y. Kosminska<sup>2</sup>, S. Shevchenko<sup>2</sup>, V. Natalich<sup>2</sup>, G. Wilde<sup>1</sup></u>

<sup>1</sup>Institute of Materials Physics, University of Muenster, Muenster, Germany <sup>2</sup>Sumy State University, Sumy, Ukraine

It is known, that porous structures depending on morphology can possess unique physical properties which can determine areas of their application. In the proposed work a new technique for synthesizing metal porous micro- and nanostructures has been developed. This approach is based on the phase transition of sputtered substances into the condensed state under conditions close to thermodynamic equilibrium. The low dimensional metal systems (Cr, Zn, Cu, Ti, Ni, Al) have been obtained in the different morphological forms, such as network structures, nanowires, agglomerations of weakly-bound crystals, columnar structures consisting of prolonged crystals with approximately identical habitus, etc. The results confirm the important new opportunities for size, shape and physical property tuning of nanostructured materials that are given by deposition near thermodynamic equilibrium conditions. It has been established that the growth mechanism under conditions close to thermodynamic equilibrium possesses principally new peculiarities and possibilities in comparison with traditional methods of condensation from vapor state and consequently can contribute to a new zone in the structure zone model.

- [1] A. Kornyushchenko, Y. Kosminska, S. Shevchenko, V. Natalich, V. Perekrestov. Journal of Nano- and Electronic Physics **13**, 02034 (2021).
- [2] A. Kornyushchenko, Y. Kosminska, S. Shevchenko, G. Wilde, V. Perekrestov. Journal of Electronic Materials **50**, 2268 (2021).

#### Autonomous Discovery of Semiconducting Materials for Photovoltaics

Christoph J. Brabec<sup>1,2,3</sup> and L. Lüer<sup>1</sup>

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<sup>2</sup> Helmholtz-Institute Erlangen-Nürnberg (HI ERN), Forschungszentrum Jülich , Immerwahrstrasse 2a, 91058 Erlangen, Germany
<sup>3</sup> Zernike Institute, University of Groningen, Groningen, Netherlands e-mail: christoph.brabec@fau.de

The development of complex functional solar materials poses a multi-objective optimization problem in a large multi-dimensional parameter space. Solving it requires reproducible, user-independent laboratory work and intelligent preselection of experiments. However, experimental materials science is a field where manual routines are still predominant, although other domains like pharmacy or chemistry have introduced robotics and automation long before. Human interaction in the process of data acquisition is seen critical due to incomplete assessment of meta-data or hidden processing correlations which complex reproducibility. Materials Acceleration Platforms (MAPs) are regarded as an enabling technology for Data-Driven Material Science, leading to an increased number of concepts and a dynamic evolution of MAP lines. In this talk, I will present our approach to laboratory automation in materials science with a strong focus on fully functional solar devices. AMANDA (Autonomous Materials and Device Application Platform - www.amandaplatform.com) was developed as a generic platform for distributed materials research comprising a self-developed software backbone and several MAPs. However, one realizes that accelerating a whole technology requires more than accelerated materials research. It also takes devices and process development to truly accelerate a PV technology. These are concepts which are summarized under Technology Acceleration Platforms (TAP).

This talk will stepwise introduce the current concepts and technologies to accelerate solar technologies: from the material to the device and to the process. The outlook will discuss how these platforms can be made communicative to each other in order to transform them into autonomously acting TAP with the power to accelerate the learning curve for a whole solar cell technology.

#### Superconductivity in High Entropy Alloys P. Sobota<sup>1,2</sup>, W. Nowak<sup>1,2</sup>, R. Topolnicki<sup>1,3</sup>, <u>T. Ossowski<sup>1</sup></u>, A. Pikul<sup>2</sup> and R. Idczak<sup>1</sup>

<sup>1</sup>Institute of Experimental Physics, University of Wrocław, Wrocław, Poland <sup>2</sup>Institute ofLow Temperature and Structure Research, Polish Academy of Science, Wrocław, Poland <sup>3</sup>Diosuri Center in Topological Data Analysis, Institute of Mathematics, Polish Academy of Sciences, Warsaw, Poland

Superconductivity is the set of physical properties observed in certain materials where electrical resistance vanishes and magnetic flux fields are expelled from the material. Any material exhibiting these properties is a superconductor. Unlike an ordinary metallic conductor, a superconductor has a characteristic critical temperature (T<sub>c</sub>) below which the resistance drops abruptly to zero. In our project we try to find new superconducting high entropy alloys (HEAs) materials, based on different elements as e.g. Nb, Ta, Mo, Hf, W, Th, Ti, Zr, and to develop a method of finding of new HEAs superconductors using theoretical modelling. The results of our combined theoretical and experimental study of superconducting HEAs will be presented. An example can be find in [1] where superconductivity in the (NbTa)<sub>0.67</sub>(MoHfW)<sub>0.33</sub> HEA is discussed.

#### References

[1] P. Sobota, R. Topolnicki, T. Ossowski, T. Pikula, A. Pikul, R. Idczak, PHYSICAL REVIEW B **106** (2022) 184512.

#### Synthesizing a library of 1000+ reproducible MOFs.

#### G. J. Smales<sup>1</sup>, A. P. A. Chambers<sup>1,2</sup>, I. Bressler<sup>1</sup> and <u>B. R. Pauw<sup>1</sup></u>

<sup>1</sup>BAM Bundesanstalt für Materialforschung und -Prüfung, Berlin, Germany <sup>2</sup> University of Birmingham, Birmingham, UK

While the synthesis of Metal-Organic Framework (MOF) particles can be as easy as adding two solutions together, *reproducibly* obtaining the same particles, time and time again, is a lot harder. As laboratory-independent reproducibility is a cornerstone of the scientific method, we must put effort into finding and controlling all necessary parameters to achieve this.

An open-source Python/EPICS-controlled robotic platform (see picture) was adapted to systematically explore this for a 20 ml MOF synthesis of the Zeolitic Imidazole Framework-8 (ZIF-8) chemistry in methanol. Parameters that were explored included: 1) addition sequence, 2) addition speeds, 3) reaction times, 4) source chemicals, 5) stirring speeds, 6) stirring bar choice, 7) starting concentrations, and 8) workup methodologies. It was found that, by controlling these parameters, highly reproducible syntheses are obtained. Secondly, the variation of these parameters alone led to a dramatic difference in volume-weighted particle size means, which exceeds an order of magnitude as investigated by our in-house X-ray scattering instrument [1].

The syntheses thoroughly are documented in an automated fashion, and the synthesis libraries as well as analyses libraries will become available in batches soon. With this library, it will be possible to extract previously unknown correlations, and other laboratories produce can specific particles by following the exact procedures of the particles of their choice.



#### References

[1] Glen J. Smales and Brian R. Pauw, JINST 16 P06034 (2021)

## Machine learning-enabled high-entropy Invar alloy discovery

Ziyuan Rao<sup>1</sup>, Ye Wei<sup>2</sup>, Po-Yen Tung<sup>3</sup>, Ruiwen Xie<sup>4</sup>, Hongbin Zhang<sup>4</sup>, Stefan Bauer<sup>5</sup> and Dierk Raabe<sup>1</sup>

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High-entropy alloys are solid solutions of multiple principal elements, capable of reaching composition and feature regimes inaccessible for dilute materials. Discovering those with valuable properties, however, relies on serendipity, as conventional thermodynamic alloy design rules alone often fail in high-dimensional composition spaces. Here, we propose an active-learning strategy to accelerate the design of novel high-entropy Invar alloys in a practically infinite compositional space, based on very sparse data (shown in Fig. 1). Our approach works as a closed-loop, integrating machine learning with density-functional theory, thermodynamic calculations, and experiments. After processing and characterizing 17 new alloys (out of millions of possible compositions), we identified 2 high-entropy Invar alloys with extremely low thermal expansion coefficients around  $2 \times 10^{-6}$  K<sup>-1</sup> at 300 K. Our study thus opens a new pathway for the fast and automated discovery of high-entropy alloys with optimal thermal, magnetic and electrical properties.



Fig. 1. Overview of the active learning framework for the composition design of highentropy alloys. The framework combines machine learning models, density functional theory calculations, thermodynamic simulations and experimental feedback <sup>[1]</sup>.

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#### **Dirac fermions on the triangular lattice**

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In the hunt for room-temperature quantum spin Hall insulators, bismuthene [1] has demonstrated the impressive advantage of a local spin-orbit coupling experienced by the in-plane *p*-orbitals. This alternative to  $\pi$ -bond graphene can be pushed to a conceptually even more essential level upon halving the honeycomb lattice, i.e. considering chiral *p*-orbitals on a triangular lattice [2]. Here, we theoretically conceive and experimentally realize for the first time a triangular QSHI, indenene, an indium monolayer exhibiting non-trivial valley physics and a large gap. We identify an interference mechanism of the Bloch functions and the emergence of a hidden honeycomb pattern in the charge localization, which makes the topological classification accessible to bulk experiments, without the necessity of quantum edge transport.

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#### **Machine Learning Discovery of Materials**

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Graph neural networks for crystal structures typically rely on atomic species and positions as input. We develop crystal-graph attention networks (CGATs) that substitute precise bond distances with graph distance embeddings. This approach enables high-throughput investigations based on both compositions and crystal structure prototypes. By integrating a newly curated dataset of 3 million materials with CGATs, we have efficiently explored over two thousand prototypes, covering a space of 13 billion materials. As a result, the size of the convex hull has more than doubled to over 100,000 materials.

To enhance the accuracy of density functional theory (DFT) calculations regarding thermodynamic stability, we offer SCAN and PBEsol databases for stable and metastable materials. We research in detail the dependence of the prediction error when transfer learning from PBE to these datasets.

Moreover, transfer learning enables us to extend our high-throughput searches to two-dimensional materials.

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#### Big Data in Chemistry and Chemical Engineering: Science Beyond Understanding

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In this lecture, we give examples of how data science can be used to tackle some questions in chemistry for which theory has no practical solution yet.<sup>1</sup> The focus of this lecture will be on Metal-Organic Frameworks (MOFs). The chemical design space of MOFs and related materials is enormous. In principle, one can combine an infinite number of possible linkers with many different metal nodes assembled in all kinds of MOFs. This makes MOFs an ideal playing ground for data-science methods.<sup>2</sup> Examples of questions we tried to address include the colour of MOFs <sup>3</sup>, the oxidation state of the metal in MOFs,<sup>4</sup> and predictions of the adsorption properties.<sup>5</sup> In this lecture, we also discuss recent developments using large language models.<sup>6,7</sup>

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# Progress, prospects and the future of autonomous chemistry

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This talk feature the history, frameworks and failures and resulting lessons learned that lead to the development of the first localized materials acceleration platform for battery research<sup>1</sup> and the first pan-European Materials Acceleration Platform that is both asynchronous and intention agnostic<sup>2</sup>. I will showcase different tools from our set of methods<sup>3</sup> including how to automate entire research laboratories with one modular framework, integrate research tasks across research institutions, analyze data autonomously, and how to plan the most effective experiment. This will include an opinionated selection of promising research directions in machine learning for materials science. I will conclude with a new framework on the engineering of research for better understanding, greater throughput, and experiment orchestration to achieve accelerations beyond 100x

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## Parallel electrochemical investigation of combinatorial catalytic system

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The oxidation of phenols to quinones is a keystep in the production of several fine chemicals, including some vitamins. Up to date, hazardous chemicals, like Cr(VI) are employed, which are not sustainable. The catalytic aerobic oxidation is a very green alternative, as in an optimal process no waste would be produced. Unfortunately, no excellent catalysts were identified yet.

In our project, we use so-called salene complexes, which are formed from salicylaldehydes, diamines and a metal salt, opening a threedimensional chemical space for catalyst optimization. For catalyst testing, the electrochemical observation of product formation was chosen. This method can be extended to a highly parallel process by using electrode arrays fabricated by micro-contact printing, permitting the testing of several hundred catalysts per day.

As these data do not show simple structure/efficiacy correlations, we now start to employ AI approaches to further improve catalyst performance, with the aim to reduce the number of necessary experiments to a reasonable amount.

# Accelerated MOF synthesis via automated data extraction and machine learning

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The chemical space of metal-organic framework (MOF) materials offers an almost infinite space of compositions, structures, properties, and applications. [1-2] Computerassisted methods have been applied to for the discovery [3-4] and the modification [5] of MOF. However, the potential of using machine learning (ML) methods to suggest parameters in MOF synthesis experiments is not well explored. In this presentation, I will initially I will focus on ML-based approach to quickly identify optimized growth conditions for SURMOFs with high crystallinity and uniform orientation, which can subsequently be used to optimize growth on other types of substrates. The approach offers insights into the factors governing the growth of MOF thin films and highlights the potential of ML for multi-parameter optimization problems.[6] Later, I will show how automated data extraction can be combined with machine learning to accelerate the synthesis of MOFs with desired crystal structure. In this project we could establish the first MOF synthesis database and trained ML models to predict synthesis conditions for new MOF structures. The results demonstrate that the ML models outperform human expert predictions and enable the rationalization and acceleration of the MOF discovery process.[7] Overall, these results demonstrate the potential of ML approaches to support the discovery and optimization of MOFs for diverse applications.



**Figure1.** Overview of our approach to rationalize and accelerate MOF discovery by directly predicting the synthesis conditions of a MOF based on its crystal structure.

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# Printed combinatorial libraries of emerging semiconductors for optoelectronics

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Accelerating the optimization, exploration, and discovery of energy conversion material for a future relying on purely sustainable energy technology is imperative. Material Science is starting to increasingly exploit high-throughput and autonomous approaches to identify new and improved materials with optimal key performance indicators for a specific application. What is required to succeed, is to combine high-throughput materials synthesis with analytical methods that provide direct insight and allow the prediction of the technological potential.

As a team, we are focussing on solution-based methods for scaled fabrication of optoelectronic devices. We developed combinatorial fabrication methods, both, based on inkjet-printing and slot-die coating. The combinatorial methodologies have now been validated by demonstrating compositional engineering metal-halide perovskite semiconductors.<sup>1,2,3</sup> We are now looking to establish strategic collaboration opportunities with groups specializing in high-throughput advanced analytics of material libraries.

In parallel, we have started to set up data infrastructure to capture the rapid global progress in halide perovskite photovoltaics and are working on setting up connected data infrastructure and workflows to facilitate managing and disseminating research data generated from high-throughput approaches.

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#### **Virtual Materials Design**

#### <u>W. Wenzel<sup>1</sup></u>

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In recent years a number of experimental and theory groups at KIT joined forces to form an initiative to develop, validate and apply multiscale methods for virtual materials design. In this presentation I will summarize recent results regarding the structure property relations of metal organic frameworks<sup>1,2</sup>, organic light emitting diodes<sup>3–5</sup> and lithium ion batteries<sup>6</sup>. We will discuss the impact of workflow technologies<sup>7,8</sup> towards the realization of materials acceleration platforms.

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#### Towards a digital twin for degradable Mg-implants R. Willumeit-Römer

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Mg and its alloys are a new class of load bearing metal implants because they degrade under physiological conditions. This amazing property offers significant advantages over classical, non-degrading implants which have to be removed upon the completion of the healing process. However, it is not easy to deal with Mg-based implant materials because the degradation in a biological environment is extremely complicated. The close the interplay between microstructure, material degradation and biological response is not yet fully understood. In vitro studies with sufficient spatial, temporal and chemical resolution are challenging, and longitudinal in vivo monitoring with equally good resolution is typically even impossible. Therefore, we are aiming at the development of a Digital Twin for in silico studies which link the material development and processing with the degradation and tissue response. To calibrate this digital twin, we use not only reliable electrochemical data but also in vitro and in vivo imaging data with highest resolution (e.g. synchrotron radiation-based micro and nano tomography or transmission electron microscopy). This presentation will summarize the status of the Digital Twin development for this new class of implants and provides aspects how that can be used to accelerate the approval process of these medical devices.

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#### A step towards predicting synthesis conditions of metal-organic frameworks.

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The process of synthesising metal-organic frameworks (MOFs) falls under a branch of chemistry known as reticular chemistry. Here, well defined crystalline compounds are synthesised from a well thought out design principle by linking predefined building blocks under specific conditions. Although, this approach appears to be intuitive, the synthesis of any novel MOF still follows the conventional synthetic approach that begins with a thorough literature survey to explore reagents and reactions conditions, which are then proceeded by risk assessment, calculations of aliquots and a series of time-consuming stressful trial-and-error syntheses. Consequently, this seems to be the reason for the rather slow industrialisation of MOFs despite their potentials for a plethora of applications. For instance, although millions of stable hypothetical MOFs with interesting properties have been predicted, only approximately 100 thousand crystal structures of MOFs currently exist in the Cambridge Structural Database (CSD). This indicates a significant bottleneck in the intelligent design and synthesis of novel stable MOFs with targeted properties.

For this reason, we are interested in designing a bottom-up approach for an effective crystal engineering of high performing MOFs with details on their experimental synthetic conditions. However, in this talk, we will provide only details on our first step, which involves the creation of a comprehensive MOF database that maps every MOF to their experimental synthetic conditions.
# Exploring the Mechanical Properties of a 2D MOF: A Combined MD Simulation and Experimental Study

### Hafeesudeen Sahabudeen<sup>1</sup>, <u>Qiang Zhang<sup>1</sup></u>, Yue Liu<sup>1</sup>, Matthias Heuchel<sup>1</sup>, Rainhard Machatschek<sup>1\*</sup>

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Assessing the mechanical properties of 2D-Metal Organic Frameworks (2D-MOFs) is crucial for their perspective applications, where they can be subjected to various modes of mechanical stresses and strains. Despite the rapid developments focused on preparing ultrathin 2D-MOF materials, their mechanical and degradation behavior is not well understood.

Here, we assess how the Langmuir-Blodgett (LB) technique can be used to measure the Young's modulus of 2D-MOFs, since this method provides a non-destructive and straightforward in-situ approach to determine the Young's modulus of any 2D material formed or assembled at the air-water interface. Using the well-established 2D-MOF PdTCPP-Cu (NAFS-13) as a model system, we find that the Young's modulus measured *in-situ* is 1 - 2 orders of magnitude lower than the one determined with *ex-situ* AFM nano-indentation.

In order to better understand the difference between the two load scenarios, we perform MD simulations based on UFF4MOF force fields while varying structural defects, deformation mode and humidity.

As a result, we are able to conclude that AFM-nano-indentation is well suited to measure the tensile modulus, whereas the LB technique provides the compression modulus, which, in the case of NAFS-13, is orders of magnitude smaller.

It is further demonstrated that the LB-technique can be used to predict how the mechanical behavior of 2D-MOFs is affected by chemical degradation. Upon acidic hydrolysis of the 2D-MOF film, the interfacial 2D shear modulus decreases in line with the first order kinetics expected for the hydrolysis reaction.

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# **Abstracts of Posters**

(in alphabetical order)

# Automated Ink Sampling for High-Throughput Research on Ink-based Electrocatalysts

### T. Ackstaller<sup>1</sup> and D. Dworschak<sup>1</sup>

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Computation and machine learning are propelling the prediction of high-performance catalysts for oxygen evolution reaction (OER) in electrolyzers. [1] Much time is needed from the discovery of novel materials to their application in systems. As materials are introduced into applications, complexity increases. Research often tries to focus on a small parameter space while keeping many others fixed. However, in electrochemical applications many different parameters show correlations, that are not well understood yet. Many experiments must be carried out to get a handle on the numbers. To spare time here, automation can come in handy to cover a huge parameter space faster than with manual lab work.

In this work, we present a tool to optimize inks for catalyst layers. For this layer, novel materials are discovered and researched with liquid electrochemical tests as RDE testing. From these materials, dispersions are formulated before an electrode layer is cast. Such a layer is then deposited on both sides of a membrane material to form the catalyst coated membrane (CCM). One step up, this CCM is introduced to a full electrochemical cell. Each level of complexity adds more parameters and more time for research interactions. In addition, the performance measured for the pure catalyst and its performance in an electrolyzer CCM differ and the correlation is not yet understood. [2] The inks' characteristics depend on the materials and processes used. Systematic researching consumes large resources in lab space and time.

To overcome this bottleneck, we have developed a robotic setup for the highthroughput synthesis of catalyst ink samples. The robotic system can receive a file containing a series of compositions along with all key processing parameters. After starting the setup, each sample is produced sequentially from powders and solutions. The exact processing results are stored in an output file. Parameters such as the exact ambient conditions are also monitored. These are often neglected by human operators. This work is the first step in the development of a robotic high-throughput laboratory for electrolyzer research. Step by step, the sample complexity will be elevated and lead to a better understanding of the processes within the cell.

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# Accelerated Optimization of Water-Splitting Materials via Automation

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With the carbon neutral conversion cycle and high energy density, hydrogen production through water-electrolysis is an excelling candidate for green energy storage[1]. While there are already commercially available water electrolyzers, the dependence on rare-earth element based elements (i.e., Ir and Pt) as catalyst material, combined with efficiency and stability requirements limits the scalability of water electrolyzers. Moreover, each step of manufacturing a catalyst layer, from catalyst ink formulation to the coating parameters affects both catalytic activity and stability[2]. Therefore, the optimization of the catalyst layer with the smallest amount of precious metals requires extensive testing of the experimental design space which is only possible with speeding up both production and testing of these materials due to the very large number of all possible materials and production parameters[3].

For the characterization of catalyst candidates, Scanning Flow Cell(SFC) with Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) is an indispensable and high-throughput(HT) ready tool for assessing both activity and stability thanks to its ability to observe in operando dissolution of catalyst materials with ppb level accuracy while simultaneously performing electrochemical measurements. Towards automation of SFC-ICP-MS, we are augmenting the cobot with a custom swappable tool where SFC, a machine vision camera as well as a force sensor are mounted for precise surface approach and targeting of catalyst spots. Similarly, using image processing, the catalyst surfaces can be quantitatively evaluated to detect cracks, inhomogeneities and other faults. By combining these tools to create tabulated data stored in a database and taking advantage of machine learning techniques to guide the experiments, we ultimately aim to create a self-driving laboratory to optimize water-electrolyzers with minimum number of experiments.

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#### Title : Improved solubility prediction on most diverse dataset

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#### Abstract

The aqueous solubility is one key property of a chemical compound which determines its possible use in different applications such as drug development. Although there are many efforts made over the last decades, there are still several challenges which need to be solved, in particular for a generalization of the model to be applied to the full chemical space. The goal of this work is to develop a model which should have the capability to predict the solubility of a broad range of diverse organic molecules. We collected 7 different datasets from the previous paper and used statistical analysis to find the quality of the data based on standard deviation. We have tested a finalized model on the diverse set of organic compounds dataset and achieved improved results compared to the previous paper from Sorkun with R2 0.98 and RMSE 0.29. We explored several combinations of the descriptors along with fingerprints with diverse modeling techniques to improve the existing performance. We also performed feature analysis to identify the most suitable descriptors to predict the solubility

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## Immobilization of enzyme models in zeolites

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Enzyme models are chemical compounds that can catalyze a large number of reactions by mimicking the active site of enzymes.<sup>[1,2]</sup> However, compared to enzymes, they have the advantage of being easier to produce and therefore less expensive, and they are less sensitive to different reaction conditions such as organic solvents.

To date, intensive fundamental research has been carried out in the field of enzyme models.<sup>[3]</sup> The technical applicability of these systems can be highly improved by immobilization. This not only facilitates the separation of the reaction mixture, but also improves the reusability of the enzyme models or enables it in the first place.

In this study, we first investigated the enzyme model, a copper(II)bispidine complex, in the homogeneous reaction of styrene to the corresponding aziridine. The influence of the counterions used, the solvent and the reaction conditions were investigated. In the next step, we want to immobilize the enzyme models in zeolites. For this purpose, we want to implement the model in NaY zeolite using flexible ligand method, as this zeolite has the suitable pore size. Subsequently, the reaction rates and yields of the free and immobilized enzyme models will be compared to determine whether this increases the lifetime of the catalyst.

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# High-throughput techniques and applications for multi-component mofs for electronic device applications

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Metal-organic frameworks (MOFs) comprising regular and high-entropy structures are a captivating class of materials with vast potential for diverse applications, such as printed memristors and batteries.1-7 As their complexity increases, the number of possible compositions grows, necessitating the synthesis and characterization of numerous MOFs. An automated high-throughput synthesis and characterization platform can accelerate these tasks and enable the discovery of optimized MOF and high entropy materials for specific applications.8,9

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### Robot-Accelerated Material Development for Perovskite-based Multijunction Solar Cells

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Halide perovskites have emerged as one of the most promising and diverse material systems in the history of photovoltaics. Intense interest in these materials is a result of several favorable optoelectronic characteristics, while being solution-processable using abundant elements at low temperatures. Furthermore, bandgap tunability from the UV to the NIR makes them ideally suited for multi-junction solar cells. Within the generic three-dimensional structure ABX<sub>3</sub>, a constantly expanding list of options exist for all three lattice sites, with common choices for A being cesium, formamidinium, methylammonium, dimethylammonium, while the B-site metal is frequently Pb or Sn, and the X-site is typically occupied by the halides CI, Br, and I. Alloying has been demonstrated, which, in addition to the possibility of including additives (dopants, passivating agents, capping molecules), makes for a vast space of technologically interesting compositions. The exploration of this space is therefore a task ripe for high-throughput and automated material synthesis and characterization.

Despite their advantages, most perovskite compositions used in highly efficient PSCs exhibit comparatively poor thermal stability due to the presence of organic cations, particularly the volatile methylammonium. Inorganic Cs-based perovskites are an intriguing exception, and have recently been demonstrated to exhibit excellent operational stability in single-junction solar cell devices. An attractive next step would be to find a stable composition for use in multi-junction devices. The optimization problem therefore is therefore to find a stable material with a suitable bandgap that can be processed at temperatures compatible with a multi-junction substrate.

Here we report on a project to develop a robotic high-throughput system for exploring the compositional space of perovskite top-cell absorbers. The system will include capability to fabricate samples via spin-coating and/or drop casting, and a separate apparatus for automatic characterization. In the latter, optoelectronic properties (e.g., bandgap, luminescence) will be recorded before and during ageing under elevated temperatures and intense illumination. Analytical tools will be used to understand the relationships between process parameters, material composition and film performance. The gathered information will be used to develop a material database, and computer-aided decision-making (e.g., Bayesian inference) will be used to model the hyperspace and iteratively improve the model with each new experiment. In combination we expect this system to dramatically accelerate the pace of optimization and the discovery of commercially relevant perovskite compositions.

## Design Guidelines for Two-Dimensional Transition Metal Dichalcogenide Alloys

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Two-dimensional (2D) materials and transition metal dichalcogenides (TMD) in particular are at the forefront of nanotechnology. To tailor their properties for engineering applications, alloying strategies -- used successfully for bulk metals in the last century -- need to be extended to this novel class of materials. Here we present a systematic analysis of the phase behavior of substitutional 2D alloys in the TMD family on both the metal and the chalcogenide site. The phase behavior is quantified in terms of a metastability metric and benchmarked against systematic computational screening of configurational energy landscapes from First-Principles. The resulting Pettifor maps can be used to identify broad trends across chemical

spaces and as starting point for setting up rational search strategies in phase space, thus allowing for targeted computational analysis of properties likely on thermodynamically stable compounds. The results presented here also constitute useful а guideline for synthesis of binary metal 2D TMDs alloys via a range of synthesis techniques.



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# High-Entropy Alloys for Catalyst Discovery C. M. Clausen<sup>1,</sup> J. K. Pedersen<sup>1</sup> and J. Rossmeisl<sup>1</sup>

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High-entropy materials have lately gathered attention as a potential discovery platform for novel catalyst materials [1,2]. This is due to the incredibly vast chemical space that these materials potentially span; however, this also introduces new challenges to be overcome as experimental screenings quickly become infeasible.

To address these challenges, we have created computational frameworks to predict the catalytic performance of high-entropy alloys (HEAs) for different reactions. The prediction is based on established theory within the field of catalysis, but the highly complex surfaces of HEAs necessitate machine learning-assisted regression of binding site properties, new approaches to simulating adsorbate coverage, and the use of optimization strategies to accelerate the screening of a high-dimensional space [3,4].

This poster will highlight our recent progress in this pursuit using oxygen reduction as reaction framework. We employ graph neural networks to describe the characteristic distribution of adsorption energies of \*OH and \*O on an HEA surface and how these relate to the expected catalytic activity. These predictions are validated using high-throughput experiments carried out by our collaborators, providing additional insight into the future development of prediction models [4,5].

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# Mechanical response to strain of semiconducting polymers

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Within the last decades, semiconducting polymers strongly contributed to the development of the organic electronics industry. Thus, significant efforts in molecular design are made to enhance their optical, mechanical, and electronical properties. While early strategies focused on the molecular tailoring of highly ordered crystalline structure, as for the case study poly(2,5-bis(thiophen-2-yl)thieno[3,2-b]thiophene) (C14-PBTTT), nowadays, it is emerging a new class of intrinsically stretchable semiconducting polymers, identified by short-range positional order and effective molecular charge transport, where indacenodithiophene–benzothiadiazole (C16-IDT-BT) copolymer is one of the most promising candidates<sup>[1],[2]</sup>.

We investigated the mechanical properties of C14-PBTTT and C16-IDTBT amorphous phases, through the calculation of a full elastic response to normal strains. We ran a set of uniaxial strain molecular dynamics (MD) simulations performed with the NAMD software. Starting from equilibrated samples (T = 298K, p = 1 atm), we measured the pressure tensor and derived the stiffness tensor using linear elasticity equations. Finally, we managed to convert the stiffness tensor to the engineering constants, i.e., Young modulus and Poisson ratio, and we compared our findings with the experimental data<sup>[3]</sup>. We applied the same protocol to compute the mechanical response to strains for the corresponding crystalline phases as well. We are confident that our research would provide an insight on how uniformity influences the semicrystalline polymeric film performances in terms of flexibility and electrical response in a scenario where semiconducting polymers are increasingly implied in nano scaled devices.

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### FAIR Materials Science research using NOMAD

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The NOMAD Archive and Repository [nomad-lab.eu] is an online, open data-infrastructure tailored towards searching and publishing materials science data. Supporting over 40 atomistic codes, NOMAD has already accumulated over 12.5 million Density Functional Theory (DFT) contributions.

FAIRmat is a new NFDI initiative whose main objective is to extend the NOMAD Archive to other computational techniques and experiments, in line with the FAIR principles [1], i.e., by making materials science data Findable, Accessible, Interoperable, and Reusable. The platform is a low-barrier starting point for materials exploration and analysis, including machine learning (ML)-accelerated materials discovery.

We will show an interactive poster with a hands-on demo of NOMAD in which we will show (a) how to explore the materials space via our graphical user interface (GUI), (b) user customization with a local setup of our NOMAD Oasis solution, (c) analyses and use-cases with a special focus on *high-entropy alloys* and *metal-organic frameworks*, and (d) solve all the doubts and questions you might have.

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# Computational Screening of Oxide Perovskites as Insertion-Type Cathode Materials

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The intermittency of wind and solar power – the solely sustainable energy sources which are considered to be abundantly available – leaves only one consequence: For the transition towards renewable energy systems, efficient and reliable storage technologies are needed. Batteries are one of the most widely used storage devices but current technology based on the transfer of Li-ions faces several challenges including their dependence on critical materials with respect to both, scarcity and toxicity.

In our contribution we will discuss atomic-scale investigations of potential future battery materials carried out using density functional theory (DFT). We employed a high-throughput approach in order to screen the well-known material class of oxide perovskites as insertion-type cathode materials and we derived several crucial battery properties including voltage, volume change during charge/discharge, theoretical energy density and chemical stability for in total 280 compounds. For those candidate materials with promising properties, we evaluated additional features such as voltage profile, the band gap, and diffusion barriers for ionic transport.

Such in-silico investigations significantly narrow down the potential materials space for experimental coworkers and thereby *accelerate the discovery of materials* for green, cheap and reliable energy storage.



# Structure-property relationships in functionalized metal organic framework from first-principles high-throughput screening

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Metal organic frameworks (MOFs) are novel materials with high potential in many fields of application, ranging from gas storage and catalysis to optoelectronics<sup>1,2</sup>. MOFs are constituted by metal atoms bound together by linker molecules which can be modified by functional groups. This leads to an enormous variety of MOFs with different structural properties, which can in turn affect their band structures, projected density of states, charge density distribution, etc. In this project, we perform highthroughput first-principles calculations on MOF-5 a well- known member of the MOF family. We scan all possible structures that arise by exchanging the metal ion Zn with metals, and by functionalizing the linker molecule alkaline-earth (1, 4benzodicarboxylate) with Br, CH<sub>3</sub>, Cl, COOH, NO<sub>2</sub>, NH<sub>2</sub> and OH. We investigate the electronic properties of the stable structures and assess how they are influenced by the choice of the metal ion and by the functionalization of the linkers. The investigation shows that the metal atoms have an impact on the structural properties, the stability of the materials and the charge distribution at the edges of the linkers. Furthermore, the functional groups contribute to the stability. COOH and OH form hydrogen bonds which stabilizes the structure even more. The charge distribution at the linker changes significantly with the exchange of the functional groups which is due to the electron donating or withdrawing character. The band structure and density of states is influenced mostly by the functional groups. The contribution at the valance band maximum or conduction band minimum is mostly formed by carbon and oxygen. The influence of the metal atoms is negligible. NH2 and OH shifts the band gap in visible light region. These functional groups form a band gap state in between the bands. Thus, the band gap is decreased from approximately 3.5 eV to just below 1.8 eV.

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# Surfaces and interfaces of semiconductor systems for solar energy conversion

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Highly efficient systems for solar energy conversion - either to electrical power or directly to green hydrogen - almost always involve semiconductor hetero interfaces and surfaces. While DFT-based high-throughput calculations for materials discovery yielded considerable advances in a variety of fields, progress in solar energy conversion comes most likely not only from a better choice of bulk material but from a deep understanding of the involved surfaces and interfaces. E.g. regarding direct solar energy conversion to hydrogen, photocatalytic processes at the interface and further restraint of surface degradation bear considerable potential for improvement of device efficiency. So far, the screening of, for example, possible reconstructions of a given semiconductor surface is usually done by manually preselecting structures using common electron counting arguments. For interfaces, additionally possible orientations to align the two materials are identified prior to the selection of atomic structures. The energetically favorable surface or interface can then be determined by DFT calculations on the preselected structures. Often many possible adsorption sites for typically hydrogen or oxygen are then explored in order to get insights into photocatalytic processes at these surfaces. For the identification of the detailed structure of experimentally observed surfaces, quantities like reflection anisotropy spectra, STM images, or surface vibrational modes are determined additionally, often using post-DFT methods, after several possible surface structures have been identified theoretically.

We present examples of our work on surface and interfaces of semiconductor systems for solar energy conversion with special emphasis of what steps should be automated for large scale screening with minimum manual inputs and our ideas on how to do so.

### Magnetism in TiO<sub>2</sub> from ab initio perspective

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We present a combined experimental and theoretical study of surface-related magnetic states in TiO<sub>2</sub>. Experiments performed on nano-sized thin films of pure TiO<sub>2</sub> have suggested that the observed room-temperature magnetism originates from defects, in particular, from the surface of thin films as well as from point defects, such as oxygen vacancies located mainly at the surface. Since verifying this issue is very important in order to manipulate the magnetic properties of pristine TiO<sub>2</sub> films for future spintronic applications, the quantum-mechanical density functional theory (DFT) calculations were performed for (i) bulk anatase TiO<sub>2</sub>, (ii) bulk-like TiO<sub>2</sub>terminated vacancy-free (001) surfaces, (iii) TiO-terminated vacancy-containing (001) surfaces, (iv) TiO<sub>0.75</sub>-terminated vacancy-containing (001) surfaces (with additional 25% surface oxygen vacancies), as well as (v) oxygen-terminated (001) surfaces. Our fixed-spin-moment calculations identified both the bulk and the bulk-like TiO<sub>2</sub>-terminated vacancy-free (001) surfaces as non-magnetic. On the contrary, oxygen vacancies in the TiO-terminated and TiO<sub>0.75</sub>-terminated (001) surfaces led to ferromagnetic and rather complex ferrimagnetic states, respectively. The spinpolarized atoms are the Ti atoms (due to the d-states) located in the surface and subsurface atomic planes. Lastly, the O-terminated surfaces are also magnetic due to the surface and sub-surface oxygen atoms and sub-surface Ti atoms (however, their surface energy is high).

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# Search for novel optical materials

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Novel optical applications require novel optical materials. As an example, so called epsilon-near-zero (ENZ) materials have recently attracted significant attention: These materials possess a vanishing real part of the dielectric permittivity, which frequently results in extreme optical nonlinearities. We discuss promising applications in photonic communication, sensing and computation based on the literature [1]. The basic mechanism of bulk ENZ materials relies on the continuity of the normal component of the electric displacement field. A vanishing dielectric permittivity thus leads to a giant enhancement of the electric field inside the material.

However, only few practical ENZ materials are currently available, mostly degenerate semiconductors such as tin-doped indium oxide, aluminum-doped zinc oxide and dysprosium-doped cadmium oxide, which were not developed for ENZ applications specifically [2]. Furthermore, each possible ENZ application requires different material properties, e.g., operating wavelength and optimal loss. Similarly complex design considerations occur for many optical applications.

The search for novel materials with such properties has just begun; to which we plan to contribute via high throughput DFT and Machine Learning. We propose the creation of a database comprised of optical properties to aid the search of ENZ and other optical materials using a combination of existing databases and high-throughput DFT calculations. Designers would then be able to develop a figure-of-merit for each specific application [3] and extract the optimal material from the database. We critically discuss material characteristics for preselection and evaluate material properties in regards to computational accuracy and cost. In the second phase of research, the database will be used to train a predictive neural network to significantly expand the size of the database. Finally, we aim to investigate the use of generative neural networks to propose novel material compositions for guided materials design [4].

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# Application of Ti and Mg hybrid material as partly degradable implant

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Multifunctional materials based on a combination of permanent and degradable metals open new perspectives for medical implants combining osseo-conductivity and drugdelivery functions, which can significantly decrease the number of implants' revision. In this work, the hybrid magnesium-titanium materials were produced via sintering and the properties of the permanent titanium component before and after the degradation of the temporary magnesium part were evaluated. The changes of chemical composition and mechanical parameters were determined. Loading of hydrogen into the titanium part at room temperature was observed, which deteriorates the mechanical characteristics, but could also simultaneously improve the biocompatibility of the permanent titanium implant. The control of degradation of the magnesium part and the modification of the titanium part is required for the development of partly degradable hybrid implants.

# High-throughput screening workflow combining synthesis conditions and physicochemical properties of MOFs for gas separation applications

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MOFs are the subject of intense research due to their potential for various industrial applications in gas separation, catalysis, and decontamination [1]. With around 10,000 synthesized materials and one million hypothetical MOFs in databases, the challenge is determining whether hypothetical MOFs can actually be synthesized, hindering the development of ultra-high performance materials. It is therefore important to expand the databases by developing strategies that couple automated synthesis of novel compounds by synthesis robots with systematic characterization by high-throughput computational screening.

Based on the high-throughput screening techniques already developed in the laboratory, especially for rare gas separation [2], we extend the methodology to target in a first step the separation of molecular gases (N2, O2, CH4, H2). Our goal is to expand current databases beyond structural descriptors by incorporating diverse physical and chemical properties to enhance their predictive capabilities.

Once the computational screening and experimental characterization workflow is tested on a family of well-known MOFs, a high-speed hydrothermal synthesis robot will be set up to generate a series of known MOFs with a wide structural and chemical diversity, in order to obtain a quality set for future model training. The results of the computational screening, the experimental characterization data as well as the synthesis parameters will be used to feed the Machine Learning models to determine the key descriptors for gas separation [3], while taking into account novel parameters such as synthesis feasibility.

The results of this strategy can guide the synthesis of new MOFs using the flexibility of the robot. Characterization tests can then validate the strategy and extend it to other applications by comparing them with targeted values from the predictive model.

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## Perovskite Solar Cells' Ageing Test Data Clustering

# <u>Noor Titan Putri Hartono<sup>1</sup>, Hans Köbler<sup>1</sup>, Paolo Graniero<sup>2,3</sup>, Mark Khenkin<sup>2</sup>, Rutger Schlatmann<sup>2</sup>, Carolin Ulbrich<sup>2</sup>, Antonio Abate<sup>1</sup></u>

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Perovskite solar cell (PSC) technology has reached beyond 25% efficiency within the past decade,<sup>1</sup> comparable to silicon solar cells. Despite PSC's promising efficiency, its lifetime is still much shorter than silicon solar cells, because PSC undergoes complex, intertwined degradation mechanisms in the presence of illumination, oxygen, and water from humidity.<sup>2</sup> As a result, PSCs ageing test results are more challenging to analyze. We use clustering methods, for instance self-organizing map (SOM),<sup>3</sup> to help cluster the ageing test shapes and perform a large data analysis to look at the general trends of how efficiency loss due to degradation is related to maximum efficiency of the cells.

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# Alchemical Perturbation Density Functional Theory for Accelerated Screening of High-Entropy Alloys Catalysts

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High-entropy alloys (HEAs) have attracted significant interest in recent years due to their exceptional properties over conventional alloys. Recently, HEAs have shown remarkable catalytic performance especially for oxygen reduction reaction (ORR) and carbon dioxide reduction reaction (CO2RR) [1]. First principle calculation methods such as density functional theory (DFT) are typically used to explore active and stable electrocatalysts. However, the ability to use explore HEAs for catalytic application using conventional DFT is hindered by the vast configuration space due to the large number of possible arrangements of surface sites. This requires a huge number of DTF calculations, which can be infeasible. Machine learning (ML) approaches have been recently used for catalysis prediction. However, a significant drawback of ML is that large data sets are needed for training and validating the ML models. Besides, ML models are ill-placed to extrapolate catalysis reactions and energies to new systems. To tackle this issue, robust approaches to efficiently screen the configurational space of catalytic HEA materials need to be developed and employed. An efficient method to navigate the configuration space of HEA alchemical perturbation density functional theory (APDFT) [2]. A key advantage of APDFT is that a single Density functional theory (DFT) calculation of the adsorbate's binding energy (BE) can be used to predict many hypothetical catalysts surface structures' BE at a negligible additional computational cost. This characteristic makes APDFT an appealing technique to explore the configurational space of catalytic HEAs at significantly less computational cost compared to conventional brute-force DFT calculations. The accuracy of APDFT was assessed for different adsorbates on AuAgPtPdCu HEA surface. The results have shown that APDFT can capture the BE trends with minimal computational cost, suggesting a fast-screening approach with less number of brute-force DFT calculations required. The accuracy of APDFT can be further improved using gaussian regression models.

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# Investigation of structure-property relationships in amyloid-like supramolecular peptide nanofibrils

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Assemblies of peptides and proteins through specific intermolecular interactions set the basis for macroscopic materials found in Nature. When small peptides or parts of proteins fold in a specific way through hydrogen bonding interactions between amino acids, a highly ordered  $\beta$ -sheets structure is formed. This hydrogen bonding network is commonly observed in biology when peptides self-assemble into supramolecular fibrillar structures. Many researchers have taken advantage of this highly stable and easily tunable interaction to develop supramolecular peptide nanofibrils (PNFs), that can mimic biological structures in their ability to act as scaffolds, provide stability, transport materials, and signal cells. With small modifications in the structure, these nanofibers can be used in many different applications, such as promoting neuron regrowth, retroviral transduction, and carbon dioxide capture. While there is extensive research in the development of biomimetic materials with improved properties, experimental methods rely heavily on trial-and-error approaches. Inspired by our earlier work [1], here we provide a microscopic view of the self-assembled PNFs, their secondary structure, interaction with solvent and other fibrils in solution, to determine the relationship between structure, morphology and biological properties. By employing a multi-scale modeling approach, we investigate how different modifications, such as sequence truncation, amino acid substitution and sequence re-ordering, affect the self-assembly behavior, β-sheet formation and dynamic properties of peptides in supramolecular fibrils. In the future, we aim to utilize machine learning methods for data-based analysis of simulation data to better make connections and predictions between structure-property relationships of materials. The models developed will provide vital insights into the microscale processes associated with amyloid formation and how we can use it to the advantage of rationally developing strong, biocompatible, dynamic, functional materials.

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# Green energy harvesting in endohedral-fullerene, single-molecule junctions.

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For the purpose of creating single-molecule junctions, which can convert a temperature difference  $\Delta T$  into a voltage  $\Delta V$  via the Seebeck effect, it is of interest to screen molecules for their potential to deliver high values of the Seebeck coefficient  $S = -\Delta V / \Delta T$ . Here we demonstrate that insight into molecular-scale thermoelectricity can be obtained by examining the widths and extreme values of Seebeck histograms. Using a combination of experimental scanning-tunnelling-microscopy-based transport measurements and density-functional-theory-based transport calculations, we study electrical conductance and Seebeck coefficient of three endohedral the metallofullerenes (EMFs) Sc<sub>3</sub>N@C<sub>80</sub>, Sc<sub>3</sub>C<sub>2</sub>@C<sub>80</sub>, and Er<sub>3</sub>N@C<sub>80</sub>, which based on their structures, are selected to exhibit different degrees of charge inhomogeneity and geometrical disorder within a junction. We demonstrate that standard deviations in the Seebeck coefficient  $\sigma_s$  of EMF-based junctions are correlated with the geometric quantity  $\sigma$  and the charge inhomogeneity  $\sigma_a$ . We benchmark these molecules against C<sub>60</sub> and demonstrate that both  $\sigma_{a}, \sigma_{s}$  are the largest for Sc<sub>3</sub>C<sub>2</sub>@C<sub>80</sub>, both are the smallest for  $C_{60}$  and for the other EMFs, they follow the order  $Sc_3C_2@C_{80}$  $>Sc_3N@C_{80} > Er_3N@C_{80} > C_{60}$ . A large value of  $\sigma_s$  is a sign that a molecule can exhibit a wide range of Seebeck coefficients and if orientations corresponding to high values can be selected and controlled, then the molecule has the potential to exhibit high-performance thermoelectricity. For the EMFs studied here, large values of  $\sigma_s$  are associated with distributions of Seebeck coefficients containing both positive and negative signs, which reveals that all these EMFs are bi-thermoelectric materials.

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# Social network analysis (SNA) in material science for the prediction of material properties

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#### ABSTRACT

The number of new materials and applications of these materials is experiencing rapid growth. Today, increased computational power and the established use of automated machine learning approaches make the data science tools available, which provide an overview of the chemical space, support the choice of appropriate materials, and predict specific properties of materials for a desired application. Among the different data science tools, graph theory approaches, where data generated from numerous real-world applications are represented as a graph (network) of connected objects, has been widely used in a variety of scientific fields such as social sciences, health informatics, biological sciences, agricultural sciences, and economics. In this work, we describe the application of a particular graph theory approach known as social network analysis (SNA) to the metal-organic framework (MOFs) and high-entropy alloys (HEAs) materials. As a first demonstration of MOFs materials, we construct a social network called MOFSocialNet from geometrical MOFs descriptors in the CoRE-MOFs database. The MOFSocialNet is an undirected, weighted, and heterogeneous social network; following the construction of this graph, a set of social network analysis processes is conducted to extract valuable knowledge from the MOFs data using graph machine learning algorithms. Community detection is one of the well-known SNA techniques employed on the MOFSocialNet to extract the most similar MOFs communities. To evaluate whether the properties of new MOFs can be predicted using MOFs communities, we randomly chose three MOFs from the CoRE MOFs database. For these MOFs, we excluded the crystal density as input during featurization and placed the MOFs within the MOFS ocialNet. The crystal density of the new MOFs is predicted by simply averaging the crystal density of the ten nearest neighbors. The results show an outstanding prediction accuracy for the crystal density of 99.69% for MOF ABAYOU, 99.79% for ABIXOZ, and 99.96% for ACOLIP. Additionally, communities extracted from MOFSocialNet can be leveraged to predict MOFs gas adsorption properties for CO2 and CH4. To evaluate the performance, the prediction performance of MOFSocialNet is compared with three standard ML models, namely K Nearest Neighbor (KNN), Gradient Boosting Regression, and Deep Learning. The results indicate that MOFSocialNet significantly improved prediction over the baseline ML algorithms. As a second demonstration, we are attempting to build interaction networks based on HEAs descriptors to discover HEAs communities with similar functionalities. The communities are extracted on 90 HEAs and six descriptors using a community detection algorithm like the Louvain algorithm. The results demonstrate 13 alloy communities, and the accuracy of the results is validated through modularity. In addition, specific HEAs characteristics, such as phase composition, could be predicted by extracted communities. Moreover, the proposed method predicted the HEAs phase composition and achieved about 93% precision.

# Efficient NMR screening of chemical parameter spaces

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The advantages of experimenting with microlitre sized chemical samples are numerous, including large diverse sample collections such as needed for drug screening, control over explosive exothermal reactions, efficient implementation of externally enacted perturbations, application of high electrochemical fields, rapid switching of experimental parameters, including titrations, application of heat and cooling, and so on. Microlitre campaigns beneficially result in at least 10<sup>3</sup> times less waste than for conventional NMR sample tubes. But small samples also have their challenges, such as handling precision, and access to characterisation. Using sophisticated NMR probe engineering, which implements high signal-to-noise ratio multi-nucleus micro-NMR detection in combination with MEMS lab-on-a-chip techniques, we achieve liquid state NMR observation in a hyphenated setup that includes photon cs and electrochemistry [1,2]. Our approach, which has been commercialised [3], achieves an analysis speedup of up to 200× in comparison to conventional sequential NMR spectrometry, whilst retaining all of the capabilities of a conventional double resonant diffusion-NMR probehead, augmented with the required additional experimental conditions placed under full spectrometer control.



B) Beverage NMR spectra from a triggered-flow high-throughput NMR experiment [1].

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## Integrated Design Methodology for Advanced Functional Materials

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Engineering innovation and technological leadership increasingly depend on the timely availability of advanced functional materials. Development times of technology enabling materials, however, are still typically measured in decades. Within the Digital Materials Foundry of the Digitalization and Technology Research Center of the Bundeswehr (dtec.bw), we develop and implement an integrated design methodology that equally draws on advanced high-throughput quantum mechanical simulations and automated combinatorial synthesis to dramatically accelerate the optimization of materials for a range of energy applications. Here we present our recent infrastructure advances as well as selected initial results from our research on functional materials.



Figure 1: Integrated Design Methodology for accelerated optimisation of functional materials

# 2D Heterostructure design of novel energy storage devices

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Understanding material interfaces is vital to the design of novel energy and electronic devices for national energy security. Modern Li-ion batteries are based on the principle of Li intercalation involving insertion/removal of Li ions into/from the electrode materials during operation. These intercalation sites are preset by the crystal structure of these electrode materials that limit their maximum storage capacity. Additionally, these electrodes are prone to cracking due to their volumetric expansion to accommodate Li ions in the process. Quasi-2D materials such as graphene, transition metal dichalcogenides (e.g. MoS<sub>2</sub>), and hBN are promising materials that circumvent these issues due to their ability to intercalate Li at multiple sites in their interlayer gap without any expansion. Heterostructures formed by stacking different 2D materials can further maximize the storage capacity and allow for greater control of intercalation induced kinetics in these materials. We demonstrate the varying nature of Li electron donation in MoS<sub>2</sub>/hBN and MoS<sub>2</sub>/graphene heterostructures using *ab initio* density functional theory calculations. We show that the intercalation process in MoS<sub>2</sub> is considerably impeded when graphene is used as a substrate than when hBN is used, confirmed by experiments. Our results show that heterostructure design is an excellent pathway to tune novel energy storage devices.

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## MOF Synthesis Prediction Enabled by Automatic Data Mining and Machine Learning

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The metal–organic framework (MOF) synthesis and modification offer an almost infinite space of compositions, structures, properties, and applications. [1-2] And computer-assisted methods have been used to been applied to for the discovery [3-4] and the modification [5] of MOF. However, the potential of using machine learning (ML) methods to suggest parameters in MOF synthesis experiments is not yet still explored. Here, we show an approach of data mining and ML method for rationalization and acceleration of the MOF discovery process by directly predicting the synthesis condition of a MOF based on its crystal structure. There are three steps in our approach: 1) The first MOF synthesis database (SynMOF) was constructed via automatic extraction of synthesis information from the scientific literature; 2) multiple ML models were then trained on the SynMOF database; 3) synthesis conditions for MOF structures were predicted by the ML models. These early-stage ML models, exhibit a good prediction performance, surpassing human expert predictions, which was shown through a synthesis survey from 11 MOF experts worldwide.

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# Nuclear magnetic resonance supports accelerated materials discovery

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Nuclear magnetic resonance (NMR) is an information rich analytical method, offering insight into material structure and dynamics non-invasively, non-destructively, and with atomic resolution. NMR is also well known to be an insensitive technique, and thus measurement time is relatively long (minutes to hours per sample). We propose to accelerate the information extracted from NMR experiments using three approaches: transitioning sample handling from individual sample tube to plug-flow based delivery, measurement automation along the entire experiment from sample preparation to measurement analysis, and measurement parallelization to further increase sample throughput.



Figure 1 A) Samples prepared in plug-flow format increases sample throughput by a factor of 50 [1]. B) The sample preparation lab at IMT, to enable high throughput from well-plate to plug-flow.

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# The H@GB Puzzle

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According to the European hydrogen backbone report,<sup>1</sup> 53,000 km of hydrogen pipelines will be installed by 2040 to connect production, storage, and consumption sites in Europe. However, when exposed to hydrogen, pipeline steel suffers from a degradation process known as *hydrogen embrittlement*, which leads to a weakening of the metal bonds and can cause a fatal material failure.

To understand the atomic processes involved and thus, accelerate the discovery of mitigation techniques, computational modelling is commonly used to assist in the interpretation of experimental findings. While *ab initio* methods allow for highly accurate predictions on a quantum-mechanical level of accuracy, they are restricted to relatively small system sizes due to high computational costs involved.

As a result, grain boundaries remain one of the last frontiers in material science that lack a sufficient description with quantum accuracy. While highly intricate at the atomic scale, this environment is highly relevant to the surface of iron and steel in real-world applications as grain boundaries present a major point of onset for hydrogen embrittlement. However, recent advances in the field of machine learning have opened up new possibilities to tackle such large problems while maintaining a quantum mechanical accuracy.

The presented H@GB (Hydrogen at Grain Boundaries) project aims to leverage *machine learning potentials* to explore a prototype problem: Hydrogen uptake at iron surfaces that intersect grain boundaries – a precursor to hydrogen embrittlement.

If we can effectively demonstrate quantum mechanical accuracy for models of this complexity, it will yield unprecedented insights into the physics of hydrogen/steel interactions and the role of grain boundaries therein, with the potential to contribute to the development of more robust and resilient materials in various industrial settings in the long run.



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## Unravelled 143-year secret in Physics gives access to explore charge transport parameters and limiting factors of the materials unreachable otherwise.

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The development of novel and improvement of existing semiconductor and semi-insulating materials relies on the deep knowledge of free-charge transport properties. In particular, knowledge of majority charge carrier minority and diffusion lengths, lifetimes, mobilities, and concentrations allows tailoring [1] the design of semiconductor devices and effectiveness of solar control cells. transistors, detectors, sensors, and LED [2].



Figure 1. Resolving electron and hole transport properties in halide perovskite by Constant Light Induced MAgneto Transport.

At the moment, the detection of minority and majority charge (electrons or holes) properties is challenging due to the natural limitation of the experimental methods. The broadly spread methods such as time-resolved photoluminescence (trPL), Theraherz conductivity, and photoconductivity decay can detect lifetime for only a minority of carriers, which control the fast decay component of the signal. The additional drawback of the commonly used methods is that they probe charge carriers' properties in a transient regime which is not matched with device or material operation conditions in a steady state [3].

In this study, we developed the Constant Light Induced MAgneto Transport (CLIMAT) method, which resolves electron and hole mobility, lifetime, diffusion coefficient, and diffusion lengths separately. We demonstrated the implication of the CLIMAT for two different materials used in solar cell applications -both organic and inorganic solar cells. We have shown that a combination of light and magnetic field resolves charge transport properties of electrons and holes and predicts the material's effectiveness for solar cell application without making the full device. Our method opens new horizons for tailoring electron and hole transport in optoelectronic devices.

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# SURMOF e-Tongue: Highly Stable Functionalized UiO-66-X Sensor Array for Discrimination in Liquid

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The applications of QCM widely used in the fields of biotechnology, polymers, lipids, proteins, electrochemistry, environmental studies, etc. However, viscoelastic films and environmental conditions cause dissipation of oscillation energy due to mechanical losses in the flexible mass and to viscous liquid environment. But QCM can be operated in liquids, if proper measures are taken to overcome the large damping. This makes QCM platform to work as a taste sensor array for liquid/ion detection and discrimination called electronic tongue (e-tongue) [1].

Three functional UiO-66-X SURMOFs sensors with (X=NH2, N3 and CI) have been fabricated successfully using layer by layer pump method with real-time QCM control and monitoring. Their stability of the functional UiO-66-X SURMOF films were tested in harsh conditions such as: @100%RH / @ 90 °C and inside water @ pH between 1-7, @18% HCl in liquid phase for short period of time less than 30min.

Our findings demonstrate that NH2 groups in UiO-66-NH2 MOF structure facilitate the use of a post-synthetic alteration to tune the functional properties of MOFs while preserving the Zr-based extremely stable framework structure. This improves the selectivity of the highly stable UiO-66-X films for use as an e-tongue in liquid phase sensor applications. Machine learning methods with three different statistical analysis such as Linear discriminant analysis, k-Nearest Neighbor and Neural Network analysis were used to discriminate six different liquids such as ethanol, n-Hexane, 2-Proponal, p-Xylene, toluene, water. More than 99% discrimination accuracies have been obtained. QCM-D results of three highly stable functionalized UiO-66-X SURMOF sensors will be presented in six different liquids including water as a poster.

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### Cluster expansion extended to W-Ta-Vacancy Multi-component Alloy

### <u>Dhanshree Pandey</u><sup>1</sup>, Nicola Seriani<sup>1</sup>, Ralph Gebauer<sup>1</sup>, Christian Hill<sup>2</sup>, Kalle Heinola<sup>2</sup>

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Tungsten (W) is considered to be the leading plasma facing materials (PFM), with high melting temperature, low erosion rates, etc. Unfortunately the above merits are coupled with very low fracture toughness characterized by brittle failure regimes, etc. A possible way of overcoming these problems consists of the identification of alloying elements [1]. In alloys, however, the formation energy depends on the local arrangement of different atoms around the vacant sites. Hence, the vacancy concentration should be a function of alloy composition. Thus, in the present study, we treat the mono-vacancies as an extra 'chemical' component of the W-based alloy. Hence, in this study, the phase diagram of W-Ta-Vacancy (WTaVac) is calculated using a combination of first-principles energy methods and Monte Carlo simulations. First-principles total-energy calculations of different ordered arrangements (more than 400 structures) of W/Ta and mono-vacancy within the bcc W host are performed. These energies are then used to parametrize a cluster expansion that describes the energy of the WTaVac system in terms of an Ising-like Hamiltonian. The resulting CE contains 24 pairs, 6 triplets, and 6 quartet. The structures that are actually stable at low temperature out of this large set are found by constructing the convex hull of the formation energies. A well-converged cluster expansion with a cross-validation of around 8 meV/atom is then used in Monte Carlo (MC) simulations to calculate thermodynamic properties. The MC simulation results suggest the clustering of the vacancies when concentrations of all the constituent elements are fixed during the simulations, with Ta atoms primarily surrounding the vacancy clusters...

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# Machine learning based multi-parameter and multiobjective optimization in metal-organic framework thin films

### Lena Pilz<sup>1</sup>, Carsten Natzeck<sup>1</sup>, Jonas Wohlgemuth<sup>1</sup>, Manuel Tsotsalas<sup>1</sup> and Christof Wöll<sup>1</sup>

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In recent years the wide topic of machine learning has found application in chemistry. Often it is used for predicting structures, binding situations or finding new compounds. But there is also a practical approach using genetic algorithms to predict synthesis conditions. In 2019 Moosaavi et. al. published a tool called SyntesisCondition finder (SyCo finder) consisting of three consecutive steps: The diverse set, where an initial data set is created on statistically spread experiments in a former defined parameter space, the machine learning step, where a genetic algorithm optimizes the parameters according to a given goal and an evaluation step, handing over the information about the importance of the used variables. This tool has been developed for Metal-Organic Frameworks (MOFs), versatile applicable 3-dimensional structures built from metal nodes and organic linker molecules, to optimize them target-oriented in one or more properties.[1]

Using machine learning in practical chemical approaches enables the possibility of multiparameter optimizations. Compared to classical optimizations where only one parameter at a time can be optimized, this method can achieve very good results with quite few experiments varying several parameters simultaneously. Since varying multiple parameters would lead to an immense number of experiments if all possible combinations in the resulting parameter space are to be considered, this is a very powerful tool.

We have applied this method successfully on several MOF-systems, for example in the optimization of oriented growth and crystallinity of surface-anchored MOFs (SURMOFs). After finalizing an optimization there is the additional possibility of gaining valuable understanding of the system by analyzing the produced data considering the importance of variables.[2]

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# The squeezable nanojunction: A multi-physics experimental platform for accelerated geometry manipulation of nano-scale systems

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The outstanding feature of (a) Squeezable the Nanojunction (SNJ) is its ability to approach two chip surfaces with atomic precision and thereby manipulate geometries on the nano-scale. The potential of the SNJ as a multi-physics experimental platform has been demonstrated by its application to two fields of physics: Thermoelectricity



Figure 1: The SNJ as tunable light-matter interface [2]

of nanojunctions and lightmatter interaction of nano-objects in varied electromagnetic environments. Physical insight into thermoelectricity of nanojunctions has been gained by exploiting the exceptional stability of the SNJ. Molecular junctions have been tuned by slowly squeezing their length. The combination with tailored electrical measurement techniques enabled the identification and characterization of nanojunctions in the near-resonant tunneling regime. In this regime, states of (comparatively) high thermoelectric conversion efficiencies are expected and high Seebeck coefficients have been measured. Investigations on light-matter interaction in varied electromagnetic environments (with varied SNJ gap distances of several light wavelengths) illustrate fundamental relationships of photoluminescence as well as thermal radiation. Systems under investigation have been 2D-materials such as MoS2, thin films of molecules and thermal point-emitters made from epitaxial graphene (on silicon carbide).

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# Bulk and Surface Properties of Cs-based Photocathode Materials via High-Throughput Density Functional Theory Calculations

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Cesium-based photocathodes such as CsTe2 and CsSb3 are commonly used as electron sources in particle accelerators; one relevant issue hindering the control over these systems, and hence their photoemission performance, is the poor control of their crystal structure and stoichiometry during synthesis, which usually occurs upon deposition. To predict which compounds are more likely to form and to control their properties, we apply an efficient high-throughput workflow based on density functional theory calculations and explore the compositional phase space of cesium-based materials.

Firstly, we calculate the formation energies as well as the electronic properties of crystalline phases obtained from computational databases employing the meta-GGA functional SCAN, which is known to provide accurate results for these systems [1]. By including additional crystal structures based on chemical similarity, prediction we can enrich the initial structure pool. Our results indicate that a larger number of different crystal structures may be formed and thus contribute to the macroscopic material's properties [2].

In a second step, surface properties, which play a fundamental role in the material's performance, are calculated for a selected set of stable bulk crystals. The studied surfaces exhibit diverse properties ranging from semiconducting to metallic character with the latter being formed by metal terminated facets. In addition to the diminishing band gap, the work function is tremendously reduced for those metallic facets.

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## Exploring the Potential of Laser Ablation in Liquid for High-Throughput Synthesis of CoCrFeMnNi Nanoparticles

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High entropy alloys (HEAs) have garnered considerable global attention owing to their remarkable mechanical, catalytic, and magnetic properties. In particular, the cantor alloy (CoCrFeMnNi) has demonstrated exceptional fracture toughness and ductility even at extremely low temperatures. At the nanoscale, these alloys have been utilized as an additive to enhance the catalytic performance, mechanical strength, and corrosion resistance of other materials.

In the present work, CoCrFeMnNi nanoparticles are synthesized by laser ablation in ethanol using different equiatomic CoCrFeMnNi alloy targets. The targets were prepared via three routes. 1) mixing of elemental micropowders, pressed into cylinder discs and sintered in a vacuum furnace (HEA-PP) [1], 2) alloying of elemental micropowders by high energy ball milling and consolidation by spark plasma sintering (HEA-BM) [2] and 3) casting of a bulk CoCrFeMnNi alloy in cylindrical shape and direct solidification in [001] direction using Bridgeman method to form a single crystal alloy (HEA-SC) [3]. The elemental distribution within the target surface and subsurface is homogeneous for HEA-BM and HEA-SC, while large stoichiometric deviations were observed for the HEA-PP.

The nanoparticles generated from the individual targets exhibit an average elemental composition of circa 20 at. % with large compositional variations between nanoparticles, independent of the target surface and subsurface composition. The NPs produced have an FCC structure with an overall oxygen composition of 20 at.%. The productivity, composition, particle size distribution and magnetic properties are similar for every target, regardless of the preparation method, which makes laser ablation in liquid a versatile and economically feasible technique for the production of HEA nanoparticles from elemental powder pressed targets.

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# Functional Design and Investigation of Mg-Ion Conductors for Solid-State Mg Batteries

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Secondary batteries using multivalent cations as charge carrier have attracted increasing attention in recent years due to the high theoretical energy density given by multi-electron redox reactions. Nevertheless, the large charge density of these cations inevitably causes sluggish kinetics of the ion migration under room temperature, which is challenging for the development of multivalent-ion solid-state batteries.<sup>[1]</sup> Taking Mg<sup>2+</sup> as a case study, we investigated a series of functional materials to figure out the design principle for Mg-ion solid-state conductors. First of all, an ionogel electrolyte using a MOF (metal-organic framework) as skeleton was prepared.<sup>[2]</sup> An ionic liquid was incorporated into UiO-66, which has a large inner surface area as well as a rich porous structure. Comparably high ionic conductivities of  $5.7 \cdot 10^{-5}$  S cm<sup>-1</sup> and  $2.4 \cdot 10^{-4}$  S cm<sup>-1</sup> could be achieved at room temperature and at mildly elevated temperature (60 °C), respectively. Apart from the good ionic prepared electrolyte also exhibits good conductivity, the chemical and electrochemical stability against magnesium metal. By pairing the magnesium metal anode with an aromatic organic material as cathode material, a proof-of-concept quasi-solid-state Mg battery can work reversibly at 60 °C. Secondly, we replaced the ion-insulating MOF framework by a new Mg-ion conducting NASICON-structured material, Mg<sub>0.5</sub>Sn<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub>. By combining it with a small amount of Mg ionic liquid to improve the Mg<sup>2+</sup> migration at grain boundary, the prepared Mg-ion conducting hybrid electrolyte shows superior room-temperature solid ionic conductivity of  $1.1 \cdot 10^{-4}$  S cm<sup>-1</sup> and an activation energy of 0.36 eV. Through in situ scanning electron microscopy, for the first time we observed the room temperature Mg growth inside a solid-state cell by the evidence of clear-cut metal particle formation and electrolyte particle cracking. The results shown here can act as a good starting point for the understanding of Mg transport behavior in solid-state batteries.

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## Accelerating the development of stable vapordeposited perovskite thin-films via combinatorial UV–Vis degradation studies

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High-throughput experimentation (HTE) is increasingly being employed to accelerate perovskite thin-film development.<sup>[1]</sup> A particular emphasis is often put on linking larger compositional variations of solution-deposited thin-films to resulting stabilities.<sup>[2,3]</sup> However for large-area applications, vapor-based thin-film deposition may be more attractive and further allows to circumvent the usage of toxic solvents.<sup>[4]</sup> For vapor-deposited thin-films, combinatorial screening platforms have been proven to be effective for accelerated materials development.<sup>[5]</sup> However, combinatorial stability studies have yet been scarce.

In this work, we present a novel method for high-throughput screening of optical properties and materials stability. We perform a rapid screening of the transmission and reflectance in a broad wavelength range under accelerated aging conditions. Specifically, the samples are stored in inert gas or synthetic air and exposed to elevated temperatures as well as bias-light illumination. With a temperature variation of  $\pm 1$  K and light intensity variation of <2% across combinatorial libraries, meaningful combinatorial stability screening is enabled.

We investigate the effect of residual PbX<sub>2</sub> on the stability of two-step deposited perovskite thin-films grown on vapor-deposited templates. As a result, we discuss strategies to derive guidelines for the two-step synthesis of stable hybrid perovskite thin films.

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