

# Materials Design for Energy Storage and Conversion: Theory and Experiment

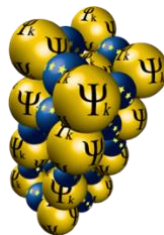


*March 02, 2021 - March 05, 2021*  
*Online event - hosted by CECAM-HQ*

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# 1. Description

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Detailed analysis of the various factors underlying the relevant properties and processes during energy conversion not only helps us to better understand the phenomenological implications of the fundamental concepts but also to uncover the important physical and chemical trends in energy systems that they produce. Electronic and atomic structure, microstructure, chemical and mechanical stability, electronic and ionic conductivity, as well as reactivity are examples of important parameters controlling the performance of energy materials. In principle, all these parameters can be characterized by applying experimental and/or theoretical techniques. Thus, designing high-performing energy storage and conversion systems requires combined theoretical/experimental efforts to screen materials in the search for optimal components.

First-principles density functional theory (DFT) calculation as well as ab initio thermodynamics, kinetics, and dynamics, and continuum-scale modelling have been applied to investigate electrode, electrolyte, and their interfaces in energy storages and conversion systems. For example, mechanism of lithiation/delithiation and potential profile for Li-based cathode materials have been simulated by calculating phase diagrams [1,2] using a combination of DFT and Monte Carlo simulations. The simulated staging mechanism was in fair agreement with experimental observation [3]. Moreover, electronic and atomic structure of surfaces as well as morphology of nanoparticles of cathode materials for Li-ion [4-5], Li-air [6], Li-S [7] batteries were characterized by DFT and ab initio atomistic thermodynamics approaches. The theoretical calculations were either combined with experimental observations or they were later confirmed by measurements. Stress distribution in primary [8] and secondary particles [9] of cathode materials was also simulated. The results of ref. 9, for example, explained the reason behind the higher stability of well-ordered core-shell cathode particles. Influence of atomic structure of grain boundaries in microstructures on Li-ion conduction in solid electrolyte materials for all-solid-state batteries (ASSBs) were also recently performed [10,11], which can be in future combined with experimental studied to understand and improve the performance of ASSBs. Electronic and atomic structure of complex solid-electrolyte/electrode interfaces have also been calculated [12,13]. On the other hand, continuum-scale modelling of solid/solid interfaces and space charge have been developed [14-18]. However, most of the computational parameters in these models have been either provided by experimental data or were considered as free parameters. Besides ideal interfaces, decomposition and secondary phase formation between solid electrolytes and electrodes were predicted by calculating Li and O grand potential phase diagrams [19,20]. Predicted interfacial phase formation were well correlated with experimental interfacial observations and battery performance.

Treatment of chemical reactions at the active interfaces in energy conversion devices, in particular solid-liquid interfaces in fuel cells, remain a great challenge for theory. Ab initio treatment of chemical reactions at solid-liquid interfaces under bias voltage was pioneered by Nørskov et al. [21]. The long-range effects of charge redistribution between electrodes, adsorbates, and electrolyte play an important role, and must be taken into account when modelling electrochemical reactions [17,22,23]. The main remaining problem is to account for statistical effects at finite temperatures, in particular configurational entropy and interplay of chemical reactions. The possibility of treating equilibrium (potential of zero charge and differential capacitance) and dynamic (chemical reactions) properties of the electrode/electrolyte interface in a unified theoretical framework remains unclear [24]. A prerequisite to address these problems from first principles is to accelerate the energy evaluation per structure without losing the accuracy. In the context of fuel cells, a combination of first-principles calculations with reactive force fields has been proposed for this purpose [25]. An interesting emerging prospect is to combine ab initio calculations with machine learning to find more transferrable interatomic potentials [26-28]. Data mining and machine learning can be also used to find descriptive parameters (descriptors) that establish correlations between easily computable or measurable properties of materials (e.g., properties of involved atoms

and interfaces) and their (electro)chemical and catalytic properties [29,30]. Therefore, we expect that bringing together experts with data mining/machine learning and energy conversion expertise will induce many interesting discussions and collaborations.

Characterization of primary particles and microstructures as well as interfaces and grain boundaries are a major challenge for both experimentalists and theoreticians. Multiscale modelling of these complex systems is still in its infancy stage and needs to be further developed. In particular, ab initio and/or atomistic-based multiscale modelling can help us to gain fundamental understanding of relation between (physical, chemical, and mechanical) properties and performance of materials. It is also an enormous challenge for experimentalist to characterize energy materials in atomic scale, in particular at realistic operational conditions.

The workshop aims to bring together theoreticians and experimentalists who are working on development of energy materials and systems. Recent advances in electronic-structure theory, continuum modelling, multiscale modelling, data analytics, as well as atomic-scale observation techniques will be presented and discussed. Applications of the theoretical and experimental methods for Li- and Na-ion batteries, all-solid-state batteries, and solid oxide fuel cells will be presented.

Topics:

- Data Mining and Machine Learning, Multiscale Modeling
- Combined Computational/Experimental Strategies for Energy Material Design
- All-Solid-State Batteries and Fuel Cells: Novel Electrolyte Materials, Interfaces and Interphases
- Novel Cathode and Anode Materials for Li-, Na-, K-, and Mg-based Batteries

## Key References

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# 2. Program

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## Day 1 - Tuesday March 02nd 2021

### Metal-ion Batteries: Theory and Experiment

- 13:00 to 13:10 - Welcome & Introduction
- 13:10 to 13:40 - **Yoshitaka Tateyama**  
DFT-based understanding of ion transfer at heterogeneous solid-solid interfaces in batteries
- 13:40 to 13:50 – Discussion
- 13:50 to 14:05 - **Randy Jalem**  
Computational design of novel solid electrolytes with inverse Ruddlesden-Popper structures
- 14:05 to 14:10 – Discussion
- 14:10 to 14:40 - **Artem Abakumov**  
Complexity of Li-rich layered battery cathodes: from point defects to grain boundaries.
- 14:40 to 14:50 – Discussion
- 14:50 to 15:10 - Coffee break
- 15:10 to 15:40 - **Qianli Ma**  
Some developments of solid-state sodium batteries in Forschungszentrum Jülich
- 15:40 to 15:50 – Discussion
- 15:50 to 16:20 - **Axel Gross**  
Atomic level understanding and descriptors for structures and processes in post-li batteries
- 16:20 to 16:30 – Discussion
- 16:30 to 16:45 - **Mohsen Sotoudeh**  
Descriptor and scaling relations for ion mobility in crystalline solids
- 16:45 to 16:50 – Discussion
- 16:50 to 17:05 - **Robert Mücke**  
Controlling mechanical stresses in cathode materials of lithium-ion batteries
- 17:05 to 17:10 - Discussion

## Day 2 - Wednesday March 03rd 2021

### Energy Materials: Characterization and Modelling

- 13:00 to 13:30 - **Keith Stevenson**  
Recent advances in energy storage: challenges and prospects
- 13:30 to 13:40 – Discussion
- 13:40 to 14:10 - **Michael Eikerling**  
Theory and computation of charged interfaces in electrochemical energy devices: challenges and approaches
- 14:10 to 14:20 – Discussion
- 14:20 to 14:50 - **Piotr Kowalski**  
Energy materials from joint theory, simulation and experimental studies
- 14:50 to 15:00 - Discussion
- 15:00 to 15:20 - Coffee break

### Electrochemistry

- 15:20 to 15:50 - **Timo Jacob**  
Chemistry electrified: from fundamental electrochemistry to energy research
- 15:50 to 16:00 – Discussion
- 16:00 to 16:15 - **Sung Sakong**  
Influence of electrochemical environments on oxygen reduction reaction
- 16:15 to 16:20 – Discussion
- 16:20 to 16:35 - **Laura Braunwarth**  
Reactive force field simulations of the electrochemical platinum-water interface
- 16:35 to 16:40 - Discussion

## Day 3 - Thursday March 04th 2021

### Electrochemistry (continued)

- 13:00 to 13:30 - **Minoru Otani**  
Recent applications of the DFT simulation combined with the classical liquid theory for energy storage devices
- 13:30 to 13:40 - Discussion

### (Electro)catalysis

- 13:40 to 14:10 - **Emiel J. M. Hensen**  
(Electro)catalyzing the energy transition

- 14:10 to 14:20 - Discussion
- 14:20 to 14:35 - **Vivek Sinha**  
A multiscale modelling approach to elucidate the mechanism of the oxygen evolution reaction at the hematite-water interface
- 14:35 to 14:40 – Discussion
- 14:40 to 15:00 - Coffee break

### High-Throughput Calculations and Machine Learning

- 15:00 to 15:30 – **Leonid Kahle**  
High-throughput computational screening for solid-state Li-ion conductors
- 15:30 to 15:40 – Discussion
- 15:40 to 16:10 - **Alexander Shapeev**  
Machine-learning interatomic potentials for computational materials design
- 16:10 to 16:20 – Discussion
- 16:20 to 16:35 - **Julia Romanova**  
Hunting for singlet fission chromophores by machine learning algorithms: the diradical character as a preselection rule
- 16:35 to 16:40 – Discussion

## Day 4 - Friday March 05th 2021

### High-Throughput Calculations and Machine Learning (Continued)

- 13:00 to 13:15 - **Sergey Levchenko**  
Single-atom alloy catalysts designed by first-principles calculations and artificial intelligence
- 13:15 to 13:20 - Discussion

### 2D Materials

- 13:20 to 13:50 - **Mina Yoon**  
Two-dimensional topological quantum materials by doping and strain
- 13:50 to 14:00 - Discussion
- 14:00 to 14:15 - **Wei Luo**  
Tuning the magnetism of MnBi<sub>2</sub>Te<sub>4</sub> by magnetoelectric coupling
- 14:15 to 14:20 - Discussion
- 14:20 to 14:40 - Coffee break

- 14:40 to 14:55 - **Lizhi Zhang**  
Prediction of two-dimensional quantum topological materials based on metal organic frameworks
- 14:55 to 15:00 – Discussion
- 15:00 to 15:15 - **Dino Novko**  
Nonadiabatic effects in Raman spectra of graphite-based batteries
- 15:15 to 15:20 – Discussion
- 15:20 to 15:35 - **Christian Tantardini**  
Computational modelling of 2D materials under high pressure and their chemical bonding: Silicene as possible field-effect transistor
- 15:35 to 15:40 – Discussion
- 15:40 to 15:50 - Closing Word

## 3. Abstracts

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### ***Recent Advances in Energy Storage: Challenges and Prospects***

**Keith Stevenson**

Skolkovo Institute of Science and Technology, Russian Federation

The development of advanced materials and technologies to efficiently convert and store energy directly into electricity is of urgent importance due to increasing energy demands of an ever-growing world population and the growing need to diversify to renewable energy alternatives. However, tremendous scientific challenges remain before successful implementation of any number of competing energy technologies such as solar cells, fuel cells, and batteries can be adopted. The materials, interfaces and device architectures currently being explored are very challenging to interrogate by ensemble-averaging, bulk experimental methods since they do not exhibit long-range order or homogeneity, contain unique nano-morphological features and possess non-uniform chemical compositions and defect chemistry. Additionally, these materials and interfaces are dynamically “reactive” and their performance degrades significantly during use which limits their cycle life and their ultimate commercialization prospects. This presentation will summarize a few materials design strategies for optimizing the performance of redox-active materials used in beyond Li<sup>+</sup> applications (i.e. for Na<sup>+</sup> and K<sup>+</sup> batteries). The development of high resolution spatially resolved tools for studying charge transfer processes at electrochemical interfaces will also be highlighted. Information obtained from these new tools enables the elucidation of complex electron and ion transfer mechanisms and degradation processes in existing and emerging materials considered for advanced electrochemical energy storage applications.

### ***Influence of electrochemical environments on oxygen reduction reaction***

**Sung Sakong, Axel Groß**

Ulm University, Germany

Based on density functional theory (DFT) calculations, we have addressed the influence of the electrochemical environment on the oxygen reduction reaction (ORR) on Pt-based surfaces [1]. The ORR occurs at the cathodes of hydrogen fuel cells and is thus one of the most crucial multistep electrocatalytic reactions. To describe the ORR scheme and activity in detail, the energetics of representative configurations at each reaction step is needed, in principle, under consideration of the presence of the electrolyte. The surrounding solvation layer can significantly influence the energy of each reaction intermediate on the cathode surface. Typically, upon adsorption on the cathode, a possible reaction intermediate has to replace molecules in the solvation layer. ¶Furthermore, these intermediates can become hydrogenated upon interaction with protons from the aqueous electrolyte, which might be effectively mediated via the Grotthuss mechanism. We have taken the statistical nature of the structure of the solvation layer at the electrode/electrolyte interface into account by extensive atomistic modeling using ab initio molecular dynamics (AIMD) simulations [2-4]. Based on these AIMD simulations, we have introduced a model solvation layer to address the ORR under reaction conditions. For each reaction step, by varying the numbers of O and H species in the model solvation layer within a grand canonical scheme, the stability of possible reaction intermediates in the ORR is assessable. Based on these calculations, the proposed reaction scheme globally agrees with the widely accepted four-electron process. Still, we show that the consideration of the electrochemical environment strongly influences the microscopic details of the rate-limiting step.

[1] Sakong et al., *J. Phys. Chem. C* **124**, 27604 (2020)

[2] Sakong et al., *J. Chem. Phys.* **144**, 194701 (2016)

[3] Sakong and Groß, *J. Chem. Phys.* **149**, 084705 (2018)

[4] Sakong and Groß, *Phys. Chem. Chem. Phys.* **22**, 10431 (2020)



## ***DFT-based understanding of ion transfer at heterogeneous solid-solid interfaces in batteries***

**Yoshitaka Tateyama**

National Institute for Materials Science, Japan

Interfacial charge-transfer processes in batteries govern their performance and stability, where both electronics and ionics play crucial roles. However, the electronic and atomistic understanding of such interface processes is still difficult in the conventional computational as well as experimental approaches.

We have addressed this issue by DFT-based calculations of free/potential energy profile across the interfaces [1,2], in conjunction with interface structure search technique utilizing CALYPSO method [2]. We clearly showed the energy profiles upon discharging. The results also provided several perspectives about possible origins of interfacial ionic resistance, picture of charging effect, and how to understand the electrochemical equilibrium [1-3].

[1] T. Baba, Y. Tateyama *et al.*, *Phys. Chem. Chem. Phys.* **22**, 10764 (2020)

[2] B. Gao, R. Jalem, Y. Ma, Y. Tateyama, *Chem. Mater.* **32**, 85 (2020)

[3] K. Leung, *Phys. Chem. Chem. Phys.* **22**, 10412 (2020)

## ***Hunting for singlet fission chromophores by machine learning algorithms: the diradical character as a preselection rule***

**Julia Romanova**, Lyuben Borislovov, Miroslava Nedyalkova, Joanna Stoycheva, Alia Tadjer  
Sofia University "St. Kliment Ohridski", Bulgaria

Singlet fission is a photophysical process in organic chromophores, in which one singlet exciton is split into two triplet excitons [1]. Therefore, implemented in solar cells, the singlet fission can lead to next generation organic-based photovoltaics with efficiency above the Shockley–Queisser limit. Nakano *et al.* proved recently that among the known chromophores, those with low to intermediate diradical character are candidates with high singlet fission potential [2]. Therefore, we developed a high-throughput procedure, combining quantum-chemical calculations and machine learning approaches, that serves as a prescreening tool for the discovery of new singlet fission chromophores based on their diradical character. Our approach is cheap in terms of computational resources and can be easily extended to explore new potential materials for organic photonics and spintronics. Statistical analysis allows us to explore the relationships between molecular structure and singlet fission propensity and to outline classes of prospective compounds for photovoltaics application.

[1] M. Smith, J. Michl, *Chem. Rev.*, **110**, 6891-6936 (2010)

[2] T. Minami, M. Nakano, *J. Phys. Chem. Lett.*, **3**, 145-150 (2011)

## ***A multiscale modelling approach to elucidate the mechanism of the oxygen evolution reaction at the hematite-water interface***

**Vivek Sinha**<sup>1</sup>, Dapeng Sun<sup>1</sup>, Evert J. Meijer<sup>2</sup>, Thijs J.H. Vlugt<sup>1</sup>, Anja Bieberle-Hütter<sup>3</sup>

<sup>1</sup>Delft University of Technology, Netherlands

<sup>2</sup>University of Amsterdam, Netherlands

<sup>3</sup>Dutch Institute for Fundamental Energy Research, Eindhoven, Netherlands

Photoelectrochemical (PEC) cells that produce hydrogen from water are highly relevant in transition to a clean energy economy. Hematite is a promising semiconductor for PEC cells due to its suitable band gap (2.1 eV), availability, stability and non-toxicity. The oxygen evolution reaction (OER) largely determines the energy efficiency in PEC cells. Computational modelling of OER, a multiscale modelling problem, can improve the efficiency of PEC cells by mechanistic insight which is unavailable from experiments. We present a multiscale computational model of OER which connects the thermodynamics and kinetics of elementary charge transfer reactions in OER to kinetics of OER at laboratory length and time scales (Figure 1). We couple density functional theory (DFT) and DFT based molecular dynamics (DFT-MD) simulations with solvent effects at an atomistic level with kinetic Monte Carlo (kMC) simulations at a coarse-grained level in our multiscale model. The time and applied bias

potential dependent surface coverage, which are experimentally not known, and the O<sub>2</sub> evolution rate during OER at the hematite-water interface are calculated by the multiscale model. Furthermore, the multiscale model demonstrates the effect of explicitly modelling the interaction of water with the electrode surface via direct adsorption.

[1] V. Sinha, D. Sun, E. Meijer, T. Vlugt, A. Bieberle-Hütter, *Faraday Discuss.*, (2020)

## **Atomic Level Understanding and Descriptors for Structures and Processes in Post-Li Batteries**

**Axel Gross**

Ulm University, Germany

Li-ion battery technology is confronted with limitations, including uncertainties regarding sustainable materials supply. Sustainable electrochemical storage systems based on Na, Mg, Zn, Ca, Al, Cl ions, so-called post-Li system, might become an alternative. Furthermore, finding chemical trends among these different battery chemistries might lead to design principles for safer and better batteries based on the so-called descriptor concept. We will show how first-principles electronic structure calculations can be used to derive such chemical trends in battery materials. In particular we will demonstrate how descriptors can be derived both for dendrite growth on metal anodes [1] as well as for the ion mobility in solid electrolytes and electrodes [2] based on calculated diffusion barriers. Furthermore, we will show how the comparison of measured and calculated Raman spectra can yield an almost complete picture of Na insertion and storage in hard carbon [3, 4].

[1] M. Jäckle, K. Helmbrecht, M. Smits, D. Stottmeister, A. Groß, *Energy Environ. Sci.*, **11**, 3400-3407 (2018)

[2] M. Dillenz, M. Sotoudeh, H. Euchner, A. Groß, *Front. Energy Res.*, **8**, (2020)

[3] M. Anji Reddy, M. Helen, A. Groß, M. Fichtner, H. Euchner, *ACS Energy Lett.*, **3**, 2851-2857 (2018)

[4] H. Euchner, B. Vinayan, M. Reddy, M. Fichtner, A. Groß, *J. Mater. Chem. A*, **8**, 14205-14213 (2020)

## **Reactive force field simulations of the electrochemical platinum-water interface**

**Laura Braunwarth, Timo Jacob**

Ulm University, Germany

A detailed understanding of the atomic-scale platinum electrode-electrolyte interface is mandatory for further advancements of the ongoing electrochemical processes on this interesting electrocatalyst. While valuable insights could be gained for the platinum-water interface by first-principles electronic structure calculations (e.g., on the formation and the characteristics of the electric double layer as well as surface charging phenomena), open questions still remain, for instance the influence of the aqueous electrolyte, requiring an explicit description to account for charge reorganization events, or the inclusion and control of the electrode potential.

Here, we aim to add another insightful puzzle piece by utilizing ReaxFF, a bond-order dependent reactive force field approach. ReaxFF enables studies on the dynamical evolution of a system, concatenated with a description of time- and geometry-dependent polarization effects as well as charge redistributions. Using a self-developed ReaxFF force field for Pt/Oxygen-systems that was previously applied to study the oxidation of the Pt(111) surface [1,2] and nanoparticles of various shapes [3], we now focus on the description of the electrode-electrolyte interface. Here, we circumvent the explicit modeling of the electrode potential by establishing a structure-potential-relation by patterning the potential-dependent free charge density in the double layer potential region as well as the composition of adsorbed oxygenated intermediates in the surface oxidation region.

For further exploration of the interface properties, we performed a thermodynamic characterization of the Pt-H<sub>2</sub>O interface via the Two-Phase Thermodynamics (2PT) method by evaluating molecular dynamics trajectories. Based on the obtained spatial entropy development that increases when moving away from the electrode into the electrolyte, an entropy reduction of nearly 50% in the layer of adsorbed H<sub>2</sub>O was observed. A distinct near-order in the inter-oxygen distribution as well as the occurrence of hexagonal motifs reason the significantly lowered entropy.

In order to deepen our microscopic understanding of electrode-electrolyte interfaces as well as the ongoing processes, our future aims are to investigate the oxygen reduction reaction and water formation

within our model of the electrified Pt-H<sub>2</sub>O interface in a grand-canonical molecular dynamics approach [4,5].

- [1] D. Fantauzzi, J. Bandlow, L. Sabo, J. Mueller, A. van Duin, T. Jacob, *Phys. Chem. Chem. Phys.*, **16**, 23118-23133 (2014)  
[2] D. Fantauzzi, J. Mueller, L. Sabo, A. van Duin, T. Jacob, *ChemPhysChem*, **16**, 2797-2802 (2015)  
[3] B. Kirchoff, L. Braunwarth, C. Jung, H. Jónsson, D. Fantauzzi, T. Jacob, *Small*, **16**, 1905159 (2019)  
[4] C. Jung, L. Braunwarth, T. Jacob, *J. Chem. Theory Comput.*, **15**, 5810-5816 (2019)  
[5] L. Braunwarth, C. Jung, T. Jacob, *Top Catal*, **63**, 1647-1657 (2020)

## **Theory and Computation of Charged Interfaces in Electrochemical Energy Devices: Challenges and Approaches**

**Michael Eikerling**, Rebekka Tesch, Piotr Kowalski  
Forschungszentrum Jülich, Germany

The interfacial region between metal surface and aqueous electrolyte lies at the heart of electrochemical energy technologies. The need to understand the properties of this region, also referred to as the electrochemical double layer (EDL), continues drive extensive research in electrochemistry.[1][2] However, with crucial aspects of the EDL not well understood, the existing interpretations of interfacial phenomena are often controversial. The fundamental task is to unravel the complex interplay of electronic structure effects, potential-induced variations of surface properties, local reaction conditions on the electrolyte side of the interface, and electrochemical kinetics of reactions of interest. This situation brings into focus theoretical models and computational studies. The presentation will recapitulate a recently presented theoretical framework that accounts for essential components and phenomena of the interface system, albeit in a simplified way. [3] We will discuss the two main challenges that hinder a predictive and transferrable theoretical description of the EDL. The first challenge concerns the atomic scale description of interactions between the liquid electrolyte and the metal surface; various computational approaches have been employed, from *ab initio* molecular dynamics (AIMD) [4] simulations to continuum electrolyte models. The other challenge is to properly account for the impact of the electrode potential on the interfacial energy landscape, which controls the kinetics of every electrochemical process. In recent attempts to address these challenges, we have adopted a computational scheme that employs the DFT/ESM-RISM method, developed by Otani and co-workers, [5] and applied it to simulate the Pt (111) surface with varying number of oxygen adatoms in acidic solution. [6] With some variation to the distance of closest approach between metal and electrolyte regions being done, the hybrid solvation method reproduced the peculiar non-monotonic charging relation of the Pt-electrolyte interface, in agreement with the theoretical prediction. The presentation will conclude with a discussion of practical implications of this charging relation.

- [1] O. Magnussen, A. Groß, *J. Am. Chem. Soc.*, **141**, 4777-4790 (2019)  
[2] M. Eslamibidgoli, M. Eikerling, *Current Opinion in Electrochemistry*, **9**, 189-197 (2018)  
[3] J. Huang, A. Malek, J. Zhang, M. Eikerling, *J. Phys. Chem. C*, **120**, 13587-13595 (2016)  
[4] S. Sakong, K. Forster-Tonigold, A. Groß, *The Journal of Chemical Physics*, **144**, 194701 (2016)  
[5] S. Nishihara, M. Otani, *Phys. Rev. B*, **96**, 115429 (2017)  
[6] V. Fernandez-Alvarez, M. Eikerling, *ACS Appl. Mater. Interfaces*, **11**, 43774-43780 (2019)

## **Computational Design of Novel Solid Electrolytes with Inverse Ruddlesden-Popper Structures**

**Randy Jalem**<sup>1</sup>, Yoshitaka Tateyama<sup>1</sup>, Kazunori Takada<sup>1</sup>, Masanobu Nakayama<sup>2</sup>

<sup>1</sup>National Institute for Materials Science, Japan

<sup>2</sup>Nagoya Institute of Technology, Japan

All-solid-state Li-ion batteries (Li-ASSBs) are now widely regarded as one of the most promising next-generation energy conversion and storage devices for the realization of low-carbon society. The solid electrolyte (SE) is one of the crucial components in Li-ASSBs which is required to satisfy multiple property criteria during battery work operation (e.g., high Li ionic conductivity, large electrochemical window, and good chemical stability). In this talk, we present our recent first-principles DFT calculation results on the material space of more than 500 *in-silico*-generated compounds with inverse Ruddlesden-

Popper structure. We evaluated the compounds in terms of thermodynamic stability, chemical stability, Li ionic conductivity, and surface property and found a number of promising novel compounds for SE use. We also determined that a modified formulation of the Goldschmidt tolerance factor is a good descriptor for thermodynamic stability and electronic band gap energy of inverse Ruddlesden-Popper compounds.

## **Energy materials from joint theory, simulation and experimental studies**

**Piotr Kowalski**

Forschungszentrum Juelich, Germany

Advancements in computational and experimental methods allow for superior, joint characterization of materials for energy applications. Rapid progress in performance of computational resources allows for *ab initio* investigation of complex metal and metal oxide energy materials. Structural arrangements, atomic-scale ordering-disordering phenomena, electronic and catalytic properties as well as phase transitions or thermodynamic parameters of complex solid solutions can be effectively studied with the aid of first principle calculations and state-of-the-art thermodynamic modeling. We apply superior supercomputing resources of Forschungszentrum Jülich to enhance understanding of performance of energy materials and provide much-needed interpretation of the experimental data. The talk will focus on derivation and understanding of structural and thermodynamic parameters, and interplay between these. As test case examples, we will discuss our results on orthophosphates and zirconates as well as selected other oxides investigated jointly with experimental partners [1-5]. A key to the successful research is application of reliable and feasible computational approach. The DFT+*U* is a widely used method for computation of strongly correlated transition metals or lanthanide/actinide elements. However, the proper choice or derivation of the Hubbard *U* parameter is crucial for computation of electrochemical reactions and materials structures. We will demonstrate performance of the linear response method for derivation of the Hubbard *U* parameter for *d* cations in different redox states and discuss dramatic improvement in the method's performance by correct selection of projectors for estimation of *d* states occupations (e.g. *Wannier-type orbitals*) [2-3]. The reported findings were enabled only through close cooperation with experimental partners and access to excellent data on ionic and electronic structures of investigated materials [1-4].

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## **Complexity of Li-rich layered battery cathodes: from point defects to grain boundaries.**

**Artem Abakumov**

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Li-rich layered oxides and chalcogenides demonstrate extremely rich electrochemical behavior being probed as positive electrode (cathode) materials for Li-ion batteries. Their record electrochemical capacity of > 270 mAh/g and energy density approaching 1000 Wh/kg are plagued with voltage and capacity fade, voltage hysteresis and kinetic hindrances intrinsic in these materials. The response of the crystal and electronic structure of the Li-rich layered cathodes toward Li<sup>+</sup> extraction/insertion is still hotly debated involving a competition between cationic and anionic redox processes, exact nature of the anionic oxidized species, type of the chemical bonding ensuring their reversible oxidation/reduction, atomistic origin of sluggish kinetic behavior and other challenging aspects. In this contribution, the ultimate complexity of these materials will be demonstrated at different spatial scales using advanced transmission electron microscopy. Because of specific synthesis techniques, the Li-rich layered oxides possess hierarchical microstructure with highly textured arrangement of the nanosized primary grains and anisotropic Li<sup>+</sup> diffusion pathways across the grain boundaries which differ substantially in composition and structure from the bulk. The interior grain structure is characterized with abundant planar defects and short-range ordering which demonstrate dynamical partially reversible behavior

upon charge/discharge. At the atomic scale these changes are tightened to transition metal cation migration, cooperative deformations of the anion framework, formation and annihilation of oxygen vacancies and structure “densification”. In total, this family of cathode materials poses extreme challenge for establishing unequivocal relationships between their electrochemical response and evolution of atomic and electronic structure. The work is supported by RSF (20-43-01012).

### ***Machine-learning interatomic potentials for computational materials design***

**Alexander Shapeev**

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### ***Descriptor and scaling relations for ion mobility in crystalline solids***

**Mohsen Sotoudeh, Axel Groß**

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Ion-mobility is a significant transport parameter for designing new functional materials with a variety of applications, including electrochemical energy storage and conversion. Here, using periodic density functional theory calculations, we identified a descriptor for the ion mobility based on the ionic radii, the oxidation states, and the difference in the Pauling electronegativities of the involved species. These physical factors describe the activation energy of binary and ternary phases of Mg cation through linear scaling relations. These scaling relations indicate that a purely classical description of ionic bonding is inadequate to explain the migration barriers, even for the highest known ionic character such as magnesium fluoride. The deviation from the purely ionic picture is taken into account by including Pauling's electronegativity difference which reflects the bond character between migrating cations and the anions of the host lattice. This work identifies the crucial factors underlying ion mobility and will thus be instrumental in guiding experimentalists and theoreticians towards the accelerated discovery of materials with improved migration properties.

### ***Two-dimensional topological quantum materials by doping and strain***

**Mina Yoon**

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Our understanding of two-dimensional (2D) materials has developed rapidly with the recent development of theoretical and computational capabilities, as well as the development of experimental synthesis and characterization tools. Now it has become feasible to perform atomic-level manipulations of 2D materials, such as introducing atomic defects and dopants, and engineering boundaries by applying strains and interfaces by growing them on different substrates. The rich parameter space of 2D materials makes them appealing candidates for new types of flexible electronic devices, such as topological electronics, that are stable at room temperature and operate at ambient conditions. Theoretical/computational modeling is desirable to guide experimental efforts in exploring the vast parameter space of 2D material and to identify desirable parameter space. On the other hand, modeling 2D materials in realistic experimental conditions is challenging; not only do these materials contain many atoms (exceeding thousands of atoms) but also the materials' parameter space is highly complex. Using a prototypical topological 2D system as an example—e.g., graphene, transition metal chalcogenides, and stanene—schemes combining various levels of theoretical/computational approaches were developed, including a tight-binding approach, high-throughput density functional theory calculations, data analytics, and large-scale ab initio dynamics using supercomputer capabilities at the Department of Energy's Oak Ridge National Laboratory. The developed schemes are targeted to enhance theoretical predictability for topological materials while significantly narrowing the gap with the experimental effort toward robust topological materials.

## **Recent applications of the DFT simulation combined with the classical liquid theory for energy storage devices**

**Minoru Otani**

National Institute of Advanced Industrial Science and Technology, Tsukuba, Japan, Japan

Electric double-layer capacitors (EDLCs) are attractive energy storage devices to address uneven power demand in sustainable energy systems. To improve an efficiency and durability of electrode materials we need to understand the precise electronic structures and electrochemical properties of EDLC. In this study, we focus on MXene electrode which is an emerging class of two-dimensional materials with chemical formula  $M_{n+1}X_nT_x$  (where M is a transition metal, X is carbon or nitrogen, T is surface termination groups) [1-3].

Using a DFT-based simulation technique which is combined with the classical liquid theory [4-7], we revealed significant electrochemical properties of MXene electrode such as the capacitive/pseudocapacitive behavior [1] and the negative dielectric constant [3].

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## **(Electro)catalyzing the energy transition**

**Emiel J. M. Hensen**

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We have far more oil, coal and gas than we can safely burn. Tackling global warming implies that we must leave most of our fossil reserves in the ground. This will require a completely different way of running our economy, shifting from liquid fuels for mobility and deriving our chemicals and materials as much as possible from renewable resources. I will discuss the challenges and opportunities that arise from these changes and the enabling role that catalysis can play in the energy transition using examples of our own research.

A view on our “catalysis by design” approach will be presented, which starts from molecular level understanding of reaction mechanism in relation to the atomic scale structure of catalytic surfaces. This approach is applied to the conversion of small molecules such as carbon monoxide, carbon dioxide and methane. A case study is the hydrogenation of CO<sub>2</sub> with “green” hydrogen to CH<sub>4</sub> as a storage chemical by Mn-promoted Ni nanoparticle catalysts. The combination of theoretical modelling based on density functional theory and microkinetics simulations, advanced characterization and reactor modelling allows optimizing Ni catalysts for small-scale Sabatier reactors. I will also show how to apply the tools, techniques and lessons learned from thermal (heterogeneous) catalysis to the field of electrocatalysis. I will first discuss the relevance of different polymorphs of MoS<sub>2</sub> as hydrogen evolution reaction (HER) catalysts with the potential to replace more expensive Pt standard HER electrocatalysts. A second example pertains to microkinetic simulations of electrochemical reactions, viz. the electrochemical reduction of CO<sub>2</sub> to C<sub>1</sub> products on Cu electrodes, in which mass transfer limitations will be taken into account in a first-principles based electro-chemokinetic network of the underlying elementary reaction steps.

## **Chemistry Electrified: From Fundamental Electrochemistry to Energy Research**

**Timo Jacob**

Ulm University, Germany

Electrochemistry is at the heart of many important industrial processes or systems, ranging from electrolysis and hydrogen evolution over corrosion, metal deposition up to batteries, fuel cells and photoelectrochemical devices. Here, the interface between the electrolyte and the electrode, which can be considered the site where electrocatalytic processes occur, is of particular interest. Due to its complexity and multi-component environment even after many decades of research, our understanding of the properties of this interface is still scarce. In this talk we will discuss how modern theoretical multi-scale methods in conjunction with *in-situ* experiments on well-defined model systems is capable to unravel the structure and composition of these interfaces as well as the ongoing electrochemical processes.

We will concentrate on metal-ion based energy storage systems, including Li-ion and Mg-ion batteries. In our experimental work we concentrated on realizing and investigating the deposition of these metal ions on model electrodes in order to understand apparent nucleation and growth processes (e.g. dendrite formation/growth). By combining different *in-situ* structure sensitive techniques we could resolve the initial stages of dendrite formation, which we believe is prerequisite to avoid or control their appearance [1]. The experimental work was accompanied with corresponding theoretical studies [2] on the self-diffusion on transition metal electrodes, battery-relevant materials and finally the dynamics of alloy-based electrocatalysts under operation conditions, showing the importance and urgent need for *in-operando* experimental studies.

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## **Single-atom alloy catalysts designed by first-principles calculations and artificial intelligence**

**Sergey Levchenko**<sup>1</sup>, Zhongkang Han<sup>1</sup>, Debalaya Sarker<sup>1</sup>, Runhai Ouyang<sup>2</sup>, Aliaksei Mazheika<sup>3</sup>, Yi Gao<sup>4</sup>

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Single-atom metal alloy catalysts (SAACs) have recently become a very active new frontier in catalysis research. The simultaneous optimization of both facile dissociation of reactants and a balanced strength of intermediates' binding make them highly efficient and selective for many industrially important reactions. However, discovery of new SAACs is hindered by the lack of fast yet reliable prediction of the catalytic properties of the sheer number of candidate materials. In this work, we address this problem by applying a compressed-sensing data-analytics approach parameterized with density-functional inputs. Our approach is faster and more accurate than the current state-of-the-art linear relationships. Besides consistently predicting high efficiency of the experimentally studied Pd/Cu, Pt/Cu, Pd/Ag, Pt/Au, Pd/Au, Pt/Ni, Au/Ru, and Ni/Zn SAACs (the first metal is the dispersed component), we identify more than two hundred yet unreported promising candidates. Some of these new candidates are predicted to exhibit even higher stability and efficiency than the reported ones. Our study demonstrates the importance of breaking linear relationships to avoid bias in catalysis design, as well as provides a recipe for selecting best candidate materials from hundreds of thousands of transition-metal SAACs for various applications.

## **Nonadiabatic effects in Raman spectra of graphite-based batteries**

**Dino Novko**

Institute of Physics, Croatia

In this contribution we demonstrate the importance of dynamical electron-phonon interaction in predicting and simulating the precise vibrational Raman spectra of graphite-based batteries. Raman spectroscopy has been readily used to understand the staging mechanisms of anions and cations into graphite-based electrodes. By combining the density functional theory and dynamical phonon self-energy calculations [1] we investigate the Raman peaks for AlCl<sub>4</sub>-intercalated graphite in Al batteries [2]. We successfully correlate the Raman peaks of the G phonon in AlCl<sub>4</sub>-graphite with experiment for intercalation stages 1, 2, and 4, while stage 3 appears to be absent. In addition, the same methodology is used to simulate the Raman-active modes in Li-doped graphite. In this highly and negatively doped case we show how electron-hole scattering rates due to electron-phonon coupling are important to understand the vibrational peak positions and the corresponding linewidths.

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## **Prediction of Two-Dimensional Quantum Topological Materials Based on Metal Organic Frameworks**

**Lizhi Zhang**

University of Tennessee, Knoxville, United States

Two-dimensional topological materials, such as quantum spin/anomalous Hall insulators (QSHI/QAHI), are a highly promising class of materials for spintronic devices and quantum computations. To date, the investigations of topological materials are mainly focused on inorganic systems. Compared to the inorganic materials, organic materials have potentially the advantages of low cost, easy fabrications, and mechanical flexibility. Here, we propose several 2D organic topological materials in three families of 2D metal (M) organic frameworks (MOF): M<sub>2</sub>(DCA)<sub>3</sub> (M = Cu, Au), M<sub>2</sub>(DCB)<sub>3</sub> (M = Cu, Au) and M<sub>2</sub>(C<sub>2</sub>O<sub>4</sub>)<sub>3</sub> (M = Re, Pt). First-principles calculations show that M<sub>2</sub>(DCA)<sub>3</sub> (M = Cu, Au) are intrinsic QSHI which have been synthesized in experiment, M<sub>2</sub>(DCB)<sub>3</sub> (M = Cu, Au) are intrinsic QAHI, and M<sub>2</sub>(C<sub>2</sub>O<sub>4</sub>)<sub>3</sub> (M = Re, Pt) are room-temperature QAHI with the spin-orbit coupling gaps >120meV and Curie temperature >300 K. Based on the orbital and tight-binding model analysis, we also confirm their topological properties and explain the origin of these properties. Our findings not only enrich the families of the topological materials, but also provide a new avenue to track these materials in the 2D MOF systems.

## **Computational Modelling of 2D Materials under High Pressure and their Chemical bonding: Silicene as Possible Field-Effect Transistor**

**Christian Tantardini**<sup>1</sup>, Xavier Gonze<sup>1</sup>, Boris Yakobson<sup>2</sup>, Carlo Gatti<sup>3</sup>, Alexander Kvashnin<sup>1</sup>

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To study the possibility for silicene to be employed like field-effect transistor pressure sensor we explore the chemistry of monolayer and multilayered silicene focusing on change in hybridization under pressure. *Ab initio* computations show that the effect of pressure depends greatly on the thickness of silicene film, but also reveals the influence of real experimental conditions, where the pressure is not hydrostatic. For this purpose, we introduce anisotropic strain states. With pure uniaxial stress applied to silicene layers, a path for *sp*<sup>3</sup> silicon to *sp*<sup>3</sup>*d* silicon is found, unlike with pure hydrostatic pressure. Even with mixed-mode stress (in-plane pressure half of the out-of-plane one), we find no such path. Our results, in addition to introduce a new theoretical approach to study 2D materials, show how the hybridization change of silicene under pressure makes it a good FET pressure sensor.



## ***Tuning the magnetism of MnBi2Te4 by magnetoelectric coupling***

**Wei Luo**, MAOHUA DU, Fernando Reboledo, MINA YOON  
Oak Ridge National Lab, United States

Using first principle calculations, we investigate the heterostructure between MnBi2Te4 and a two-dimensional ferroelectric material. We identify the most stable stacking order between MnBi2Te4 and ferroelectric material and find that the magnetic moments are coupling with the dipole moments in the ferroelectric materials. When we switch the dipole moment, the magnetic order of MnBi2Te4 changes and a topological phase transition happen. Our work gives a new route to tune the magnetism in MnBi2Te4 by electric field.

## ***Controlling mechanical stresses in cathode materials of lithium ion batteries***

**Robert Mücke**

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Approaches to minimise the detrimental mechanical stresses inside cathode materials during electrochemical cycling due to volume change include the development of low strain active materials, the adaptation of microstructure, the mixture of active materials with opposite expansion, and the inclusion of elastomers. We calculated the stresses in core-shell cathode particles for liquid electrolytes as well as the stresses of different materials and microstructures of mixed cathodes of all-solid-state batteries. An appropriate microstructural texture is one key factor to minimise the stress. The stresses inside a multi-compositional particulate core-shell cathode with radially aligned crystals (Li[Ni<sub>0.9</sub>Co<sub>0.05</sub>Mn<sub>0.05</sub>]O<sub>2</sub>) was found to be more favourable than those inside traditional randomly packed aggregates. Herefor, experimental TEM images were used to reconstruct the real microstructures (from group Prof. Yang-Kook Sun, Hanyang University, Seoul).

For all-solid-state batteries (Li<sub>7</sub>La<sub>3</sub>Zr<sub>2</sub>O<sub>12</sub> [LLZO] / LiCoO<sub>2</sub> [LCO]) the crystallographic alignment of LCO was also found to be of major importance including the stress at the cell level (bulk, thin and thick separator): Compared to randomly oriented crystals, the in-plane textured crystallographic alignment of LCO grains, introduced by the manufacturing process, has a significant effect and yields considerably better stress distributions in all cell configurations investigated. The design of optimised all-solid-state cells with reduced separator thickness led to a significantly more favourable stress state than a typical lab scale separator-supported cell. These results can be transferred to most currently used layered-oxide cathode materials such as NMC. When LLZO fibres and infiltrated LCO particles are used instead of sintered particles, the stresses even halve. Using additional elastomer binders (such as PEO) in the infiltrated cathode, the stresses become negligible.

## ***High-throughput computational screening for solid-state Li-ion conductors***

**Leonid Kahle**<sup>1</sup>, Aris Marcolongo<sup>1</sup>, Nicola Marzari<sup>2</sup>

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We present a computational screening of experimental structural repositories for fast Li-ion conductors, with the goal of finding new candidate materials for application as solid-state electrolytes in next-generation batteries. We start from ~1400 unique Li-containing materials, of which ~900 are insulators at the level of density-functional theory. For those, we calculate the diffusion coefficient in a highly automated fashion, using extensive molecular dynamics simulations on a potential energy surface (the recently published pinball model) fitted on first-principles forces. The ~130 most promising candidates are studied with full first-principles molecular dynamics, including an estimate of the activation barrier for the most diffusive structures. The results of the first-principles simulations of the candidate solid-state electrolytes found are discussed in detail.

## **Some developments of solid-state sodium batteries in Forschungszentrum Jülich**

**Qianli Ma**<sup>1</sup>, Tu Lan<sup>2</sup>, Chih-Long Tsai<sup>2</sup>, Frank Tietz<sup>2</sup>, Dina Fattakhova-Rohlfing<sup>2</sup>, Olivier Guillon<sup>2</sup>

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Compared to their lithium counterpart, solid-state sodium battery (SSNB) is regarded to have similar properties but is a much less mature technology because it is much less addressed. Besides their well-known natural endowment like high element abundance, low price etc., in the present study, some technological advantages of SSNBs are discussed in comparison with solid-state lithium batteries (SSLBs). Very recently, Na<sub>3.4</sub>Zr<sub>2</sub>Si<sub>2.4</sub>P<sub>0.6</sub>O<sub>12</sub> (NZSP) ceramics were reported to have total conductivity of  $5 \times 10^{-3} \text{ S cm}^{-1}$  at 25 °C, higher than previously reported polycrystalline Na-ion conductors.<sup>[1]</sup> Inhibition of dendrite growth in SSLBs and SSNBs has long been a challenge to the field. In the present study, with simply sticking sodium metal to NZSP ceramic pellets and without external pressure applied during operation, the critical current density of Na/NZSP/Na symmetric SSNBs reaches  $9 \text{ mA cm}^{-2}$  at 25°C. The cells can be stably operated at areal capacity of  $5 \text{ mAh cm}^{-2}$  (per half cycle, with  $1.0 \text{ mA cm}^{-2}$ ) at 25°C for 300 h in a galvanostatic cycling measurement without any dendrite formation. This critical current density is much higher than those of existing SSLBs operated at similar conditions. The influence of metal self-diffusion on the dendritic plating is the main explanation of the high dendrite tolerance of SSNBs. In this report, the inter-ceramic contact problems in the cathode are also solved by combining the infiltration of a porous electrolyte scaffold by precursor solution with *in situ* synthesis of electrode active material.<sup>[2,3]</sup> The resulting full cells using Na<sub>3</sub>V<sub>2</sub>P<sub>3</sub>O<sub>12</sub>, NZSP and Na as the positive electrode, electrolyte and negative electrode materials, respectively, can be stably operated with a capacity of  $0.55 \text{ mAh cm}^{-2}$  at high rate of  $0.5 \text{ mA cm}^{-2}$ . This is the first successful example showing that contact problems between rigid electrolyte and electrode materials can be solved without using any soft phase (liquid, polymers, ionic liquids etc.) as an accommodation or wetting medium. Since SSNBs have these advantages while SSLBs have not, the future roadmap of the development of solid-state batteries may shift from SSLBs towards SSNBs despite the higher molar weight of the sodium compounds in comparison to the Li analogues.

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# Notes:

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