MM 22: Materials for the Storage and Conversion of Energy

Time: Wednesday 15:45–16:45 Location: H23

MM 22.1 Wed 15:45 H23

Understanding Stability of Ni-rich NMC cathodes using beyond DFT many-body calculatations — •Hrishit Banerjee[1,2,3,4], Clare Grey[2,4], and Andrew Morris[3,4] — [1]University of Dundee — [2]University of Cambridge — [3]University of Birmingham — [4]The Faraday Institution

High energy density Ni-rich $\mathrm{LiNi}_a\mathrm{Mn}_b\mathrm{Co}_c\mathrm{O}_2$ (NMC) cathodes undergo degradation in the form of O loss from the surface of NMC particles. O loss increases with Ni content and higher voltages. Our first-principles study examines the redox behavior of transition metals and O in NMC cathodes as a function of (de)lithiation. Despite Ni, Mn, and Co K-edges calculated using GW Approximation showing an excellent match with experimentally obtained XANES, we demonstrate that the ionic model of ascribing shifts in the XANES to changes in metal oxidation states is inappropriate. In these cases, which are characterised by strong covalency between the strongly correlated transition metal and oxygen, Dynamical Mean-Field Theory calculations are essential to calculate charges and hence assign oxidation states accurately. Due to a charge transfer from O p to Ni d, a ligand hole forms on O in Ni-rich regions. The individual Ni charge remains fairly constant throughout the charging/discharging process. In contrast, O has dual redox behavior, showing greater involvement in redox in Ni-rich regions while showing negligible redox involvement in Ni-poor regions. The dual behaviour of O in terms of participation in the redox process explains the overall higher relative stability of lower Ni content NMCs compared to Ni-rich NMCs or LiNiO_2 in terms of O loss.

MM 22.2 Wed 16:00 H23

Splitting Water Without Falling Apart: Accelerating the Understanding of NiFeV LDH via Design of Experiments and Machine Learning — •Juan Manuel Lombardi, Charles Pare, Karsten Reuter, and Christoph Scheurer — Fritz-Haber-Institut der MPG, Berlin

The development of sustainable energy technologies requires efficient, affordable, and durable electrocatalysts. Ni-based layered double hydroxides (LDHs) doped with Fe and V are promising candidates for the oxygen evolution reaction in anion exchange membrane water electrolyzers (AEMWE) due to their tunable structure and exceptional redox properties. However, the mechanisms by which dopants influence catalytic and structural properties are not fully understood, mainly due to the challenges posed by their humongous configurational space. In this study, we efficiently explore the thermally accessible configurations within the complex configurational space of γ-phase Ni LDHs doped with Fe and V. To maximize information gain, we sample the composition space using Design of Experiments (DoE), leverage machine learning interatomic potentials (MLIPs) to sample these configurations, and refine the results with Density Functional Theory (DFT) calculations for first-principles quality predictions. By combining these tools, we develop an optimal protocol to elucidate how the dopants influence material properties. This integrated methodology reveals pathways for optimizing NiFeV LDH compositions, enhancing energy conversion efficiencies while ensuring long-term stability.

MM 22.3 Wed 16:15 H23

Benchmarking exchange-correlation functionals for accurate computational predictions of metal organic frameworks — •Julia Santana Andreo, Joshua Edzards, Holger-Dietrich Sassnick, and Caterina Cocchi — Carl von Ossietzky Universität Oldenburg, Institute of Physics

Metal-organic frameworks (MOFs) have attracted significant attention due to their structural and chemical flexibility which makes them relevant for a wide range of technological applications, from gas storage to photocatalysis. The substitution of metal nodes and the functionalization of organic linkers can significantly expand the range of available MOFs, enabling the design of tailored properties for specific applications. While atomistic ab initio simulations can significantly contribute to explore this vast configurational space, it is mandatory to assess whether and how common approximations of density-functional-theory calculations affect the description of the structural, electronic, and vibrational properties of computationally predicted compounds. With the example of MOF-5 and its recently predicted functionalized variants [1] , we find that R2SCAN offers the optimal trade-off between accuracy and computational costs to describe electronic and vibrational properties in these materials.

[1] J. Edzards, H.-D. Saßnick, J. S. Andreo and C. Cocchi, J. Chem. Phys. 160, 184706 (2024)

MM 22.4 Wed 16:30 H23

Assessing the Bulk Stability of Oxygen Evolution Reaction Catalysts using Crystal Field Theory and Orbital Analysis — •Katarina Kretschmer[1], Michael Eikerling[1,2], and Tobias Binninger[1] — [1]Theory and Computation of Energy Materials (IET-3), Institute of Energy Technologies, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany — [2]Chair of Theory and Computation of Energy Materials, Faculty of Georesources and Materials Engineering, RWTH Aachen University, 52072 Aachen, Germany

Proton exchange membrane water electrolysis (PEMWE) is an effective method for producing green, i.e. carbon-neutral hydrogen. At the PEMWE anode catalyst, water molecules are split to form oxygen molecules in the so-called oxygen evolution reaction (OER). Iridium oxide (IrO2) and ruthenium oxide (RuO2) are recognized as highly effective catalysts for the OER.

In this talk, I will discuss the electronic properties governing the bulk stability if IrO2 and RuO2. Utilizing density functional theory (DFT) calculations, the stability of both oxide materials is assessed from the perspective of crystal field theory and bond orbital analysis. The orbital overlap between the metal (iridium or ruthenium) and oxygen atoms in the lattice and the population of the emerging bonding or antibonding orbitals is analyzed, which can either contribute to or counteract the stability of the IrO2 and RuO2 lattices. This analysis will lead to the definition of electronic descriptors for predicting the bulk stability of transition metal oxide catalysts.