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Ab Initio Simulations of Cathode Materials — Efficient Configurational Screening to Discover Novel and Stable Compositions

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Introduction to the GOAC code Configurational Optimization site occupations **INPUT** configurational result **Global Optimum** Cif File; Ion Charges disorder Configurations be can **GOAC Code GOAC Package** Heuristic Solver evaluated electrostatics ionic External Calculation of α and β matrices crystals T=1 by Coulomb MPS Global optimization Replica Interface to Atomistic Configurations by Exchange Formulation of general existing Optimization Problem (MINLP) Heuristics Coulomb (GOAC) finds Replica Exchange La End **Coulomb Energy Ground-State** lowest energy configuration Disorder Monte Carlo Optimizers Optimization by different Time Utilizes meta Heuristics (GD, MC, SA, REMC, GA, Hybrid) advanced **Gradient Descent (GD), Metropolis Monte** optimization algorithms **Energy Calculation:** Carlo (MC), Simulated Annealing MC + $\sum_{i} \sum_{i} \beta_{i,j} \times x_i \times x_j$ based on **Monte Carlo** and $\sum_i \alpha_i \times x_i$ OUTPUT **Genetic Algorithms** (SA), Replica Exchange MC (REMC), \longrightarrow

Cif Files & Energies of n best

atomistic configurations

Particle-Size Dependence of Single-Phase – Two-Phase Charging Characteristics in LFP

 Lithium-Iron Phosphate (LFP) is known to show a two-phase charging mechanism

Open-Source Code

npj Comput. Mater., 11, 202 (2025)

- Optimizations with GOAC in various supercell sizes yield a single-phase up to particle-sizes of approx. 40 nm
- **Experimentally** it is also shown that LFP has a single-phase charging mechanism at the nanoparticle regime
- Moreover, electrostatic optimizations GOAC match the reported surface orientation (010) of the two-phase interface

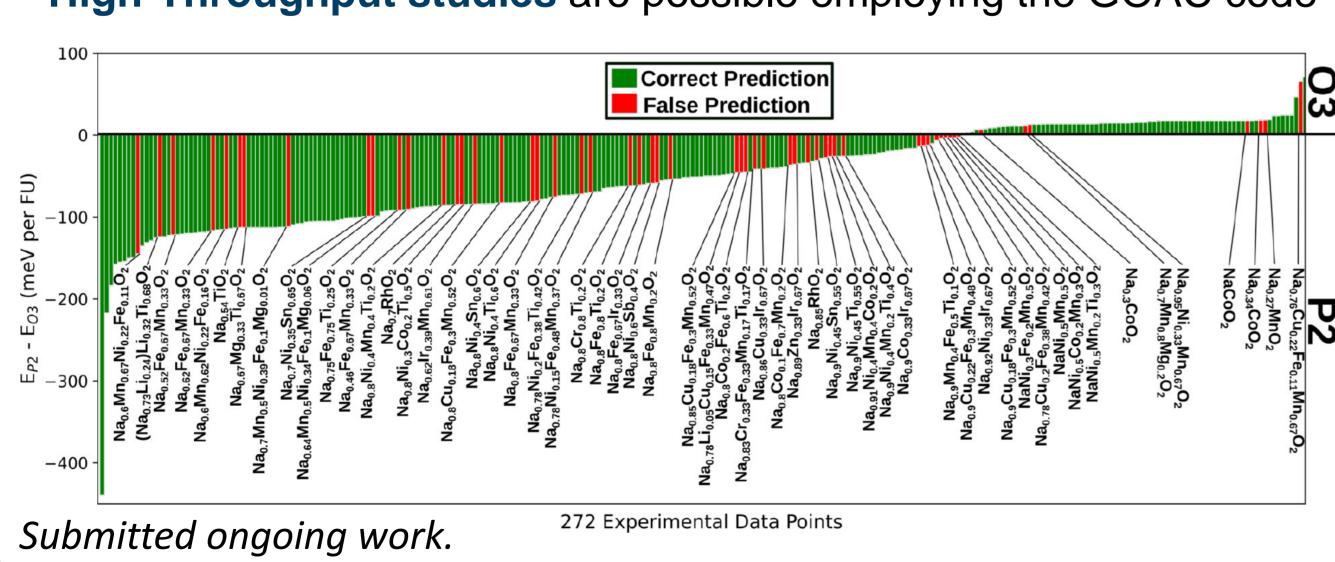
20 -Odd n in exact heuristic Under review in Phys. Rev. B, https://arxiv.org/pdf/2503.10581 Cell Length / nm

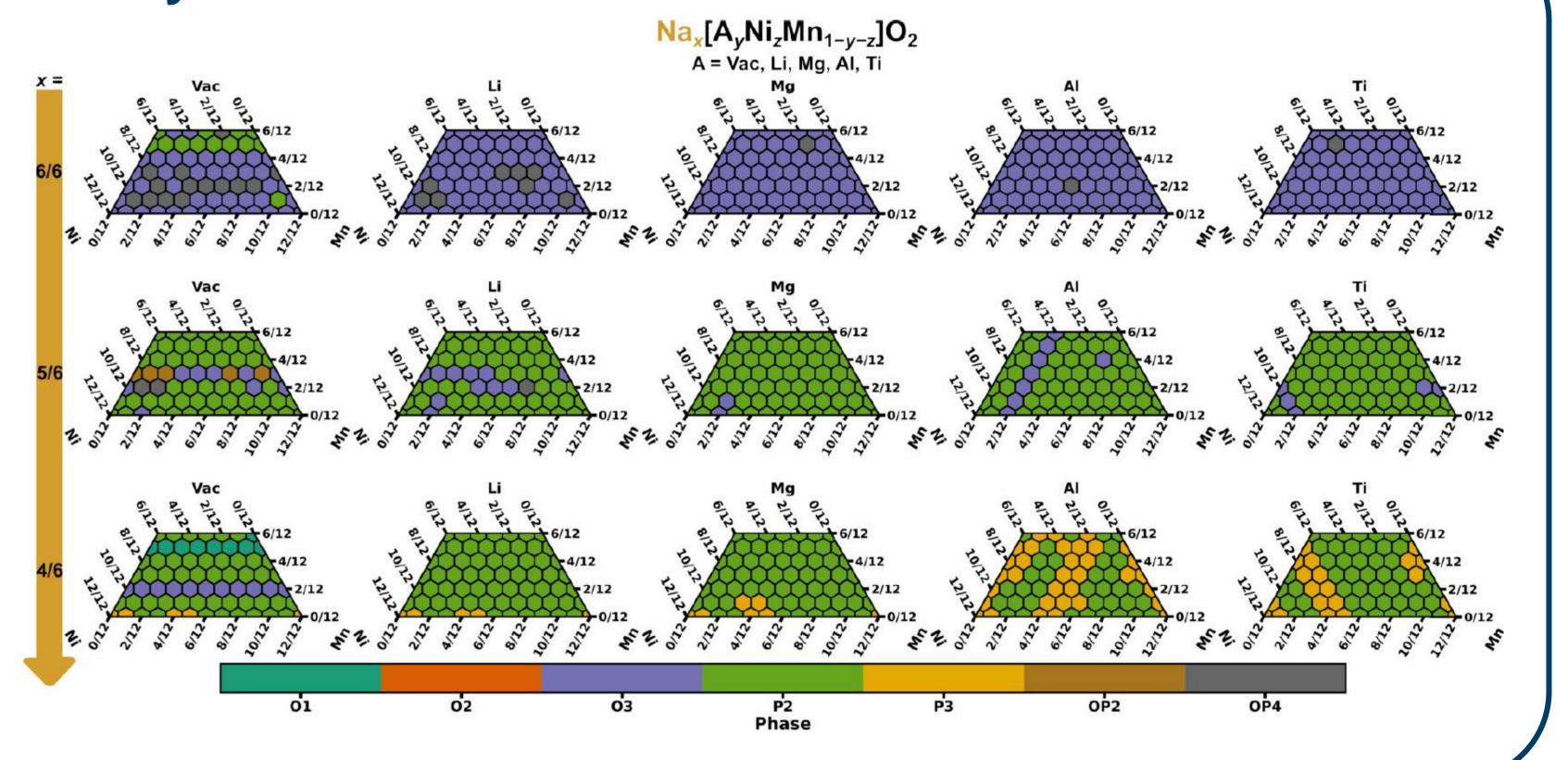
exact heuristic phase-separated **-10** - $\Delta \mathbf{E}_{\mathsf{coul}}$ -20 1x1xn (homogeneously distributed) -30 bulk LiFePO₄ / bulk FePO₄ average 15 20 25 30 35 40 45 50 55 60 65 70 75 80 b/nm

Genetic Algorithm (GA)

High-Throughput Predictions of Phase-Stabilities of Layered Oxide Sodium-Ion Cathodes

- Quantitative, ab initio prediction of most stable phases (O1, O2, O3, P2, P3, OP2, OP4) in layered oxide sodium-ion cathodes
- Energy evaluation of phases at lowest energy configurations
- Predictions are in 80% of cases in agreement to experiment
- High-Throughput studies are possible employing the GOAC code





Acknowledgements

DFG, German Research Foundation, project No. 501562980 JARA-HPC, JURECA, Forschungszentrum Jülich, Grant No. jiek12.



Conclusions and Outlook

- GOAC is a tool to optimize configurational disorders
- Configurational optimization of battery materials can give insights into charging mechanisms and phase stabilities
- GOAC offers fast optimizations in huge configurational spaces (10³⁰⁰) configurations)

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