Advancing Sodium-Ion Batteries through Electrode Doping and Optimized Electrolytes

<u>Dr. Zeynep Erdöl</u> ^a, Prof. Philipp Adelhelm ^{a,b}
a Institut für Chemie, Humboldt-Universität zu Berlin;
b Joint Research Group Operando Battery Analysis (CE-GOBA), Helmholtz-Zentrum Berlin,
Email: <u>zeynep.erdoel@hu-berlin.de</u>, <u>philipp.adelhelm@hu-berlin.de</u>

P2-type layered oxides (Na_xTMO₂) are promising cathode materials for sodium-ion batteries due to their cost-effectiveness, structural flexibility, and promising energy density. However, cycling to high voltages (>4.2 V) induces structural degradation, irreversible oxygen loss, and interfacial instability. These issues stem from irreversible anionic redox reactions, phase transitions, and Jahn–Teller distortion, all of which compromise capacity retention and safety [1].

To address these challenges, we employed a dual modification strategy: (1) Al doping at transition metal sites and (2) use of a low-concentration NaPF₆-based electrolyte. Al substitution, though electrochemically inactive, enhances structural integrity by suppressing Mn³+-induced distortions and reducing TM migration, thereby stabilizing the oxygen redox process. Concurrently, low-concentration electrolytes limit interfacial side reactions and phase instability.

The optimized system demonstrated high initial capacities of 184/178 mAh·g⁻¹ (discharge/charge) at 10 mA·g⁻¹ between 2.0–4.6 V, with an electrode loading of 7 mg·cm⁻² and 84% capacity retention after 100 cycles. These results highlight the synergistic role of bulk and interfacial modifications in enabling high-voltage, long-life sodium-ion batteries [2].

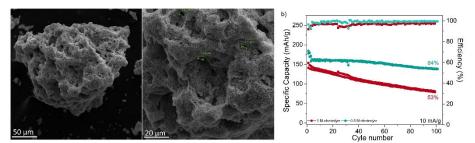


Figure 1: a) SEM images of synthesized material, b) cycling performances of P2-Na_{0.67}Mn_{0.56}Ni_{0.11}Fe_{0.25}Al_{0.08}O₂ with standard and low-concentration electrolytes at 10 mA·g⁻¹ within the voltage range between 2.0-4.6 V (vs. Na⁺/Na) [2].

Funding for this project is provided by the BMBF (Project Transition Transfer 03XP0533C)

References

[1] a) C. Zhao, Z. Yao, Q. Wang, H. Li, J. Wang, M. Liu, S. Ganapathy, Y. Lu, J. Cabana, B. Li, X. Bai, A. Aspuru-Guzik, M. Wagemaker, L. Chen, Y.-S. Hu, *J Am Chem Soc* **2020**, *142*, 5742. b) J. Geisler, L. Pfeiffer, G. A. Ferrero, P. Axmann, P. Adelhelm, *Batteries & Supercaps* 2024, 7, e202400006. c) M. Ghosh, N. Yadav, P. Adelhelm, Batteries & Supercaps 2025, e202400744. [2] Z. Erdöl et al., 2025, manuscript in preparation.