Supporting Information: Figures

Uptake of CO₂ in Layered P2-Na₀.₆₇Mn₀.₅Fe₀.₅O₂: Insertion of Carbonate Anions

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Figure S1. TGA curve and lattice parameters of air-exposed Na₂/₃[Mn₁/₂Fe₁/₂]O₂ powder measured from thermo-diffraction data collected under a helium atmosphere. The arrows show the sequence in the heating and cooling processes. The colored markers correspond to the cell parameters reported for Na₂/₃[Mn₁/₂Fe₁/₂]O₂ in the literature (refs 11, 15, 17 in main text).
Figure S2. a) XRD pattern b) SEM images of O3-\(\text{Na}[\text{Fe}_{1/2}\text{Mn}_{1/2}]\text{O}_2\) exposed to air for months showing the presence of a crystalline sodium carbonate phase.
Figure S3. Rietveld refinement of the neutron t.o.f. pattern of the air exposed Na$_{2/3}$Fe$_{1/2}$Mn$_{1/2}$O$_2$ sample a) without carbonate and b) with carbonate.
Figure S4. Reactivity of Na$_{2/3}$[Fe$_{1/2}$Mn$_{1/2}$]O$_2$ studied by TGA under different atmospheres. For each experiment, the sample was primarily annealed under dry argon at 600 °C in order to obtain a reference sample free from any air contamination. After cooling down to room temperature, the atmosphere was switched to the gas of interest and the reactivity of the material with the flowing gas was inspected at room temperature for 12 hours followed by annealing to 600 °C.
**Figure S5.** Structural parameters and Rietveld refinement of the t.o.f. NPD pattern of Na$_{2/3}$Fe$_{1/2}$Mn$_{1/2}$O$_2$ equilibrated under O$_2$ at 600 °C. The Na$_{0.66}$Fe$_{0.49}$Mn$_{0.49}$O$_2$ formula was derived from the TGA experiment under O$_2$. In this case no additional nuclear density is found in the tetrahedral site of the transition metal layer, but the vacancy configuration implies a shift along the $z$ direction of about 15 % of the transition metal atoms. This new position, noted Fe/Mn’ induces a small perturbation of the oxygen framework.
Figure S6. High temperature x-ray diffraction under helium of Na$_{2/3}$[Fe$_{1/2}$Mn$_{1/2}$]O$_2$ exposed to air for months. Scan 1 was collected as the temperature was reached and scan 2 was collected after 50 minutes at isothermal condition.