Study of the transition metal ordering in layered $Na_xNi_{x/2}Mn_{1-x/2}O_2$ ($2/3 \leq x \leq 1$) and consequences of Na/Li exchange

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Supporting Information

Figure S1: ^{23}Na MAS NMR spectra of all phase pure $Na_xNi_{x/2}Mn_{1-x/2}O_2$ prepared in this study, acquired at a spinning speed of 38 kHz, normalized to the amount of sample in the rotor. The "*" indicates the spinning sidebands for the main centerband. The peaks marked with "+" correspond to the isotropic resonance for a diamagnetic impurity, most likely Na_2CO_3, at around 0 ppm, and its spinning sidebands.

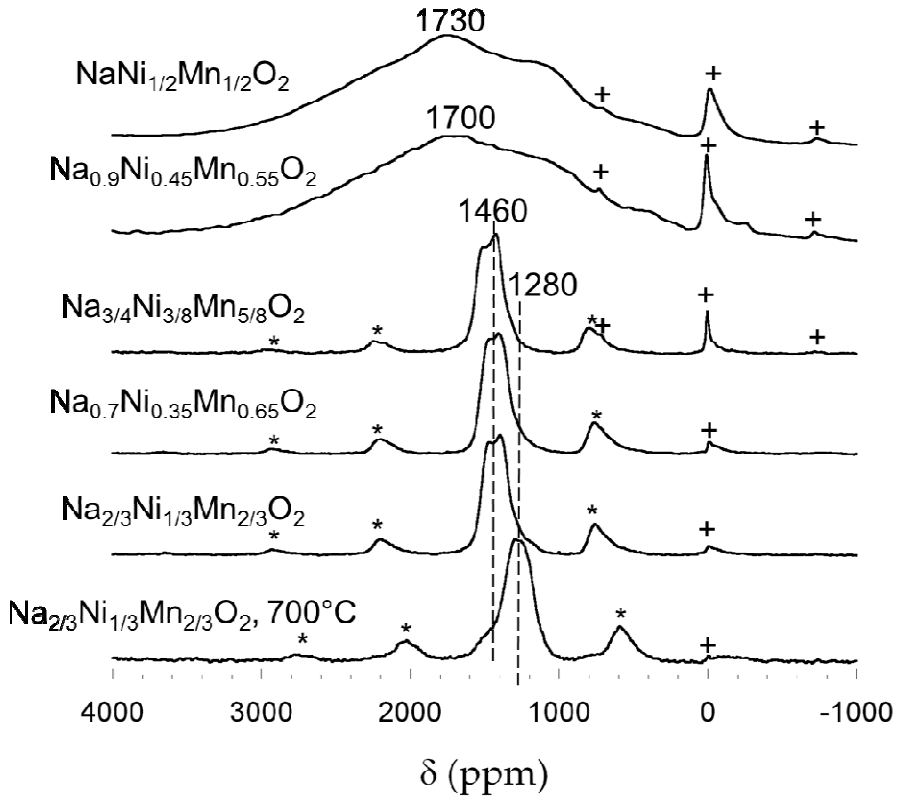
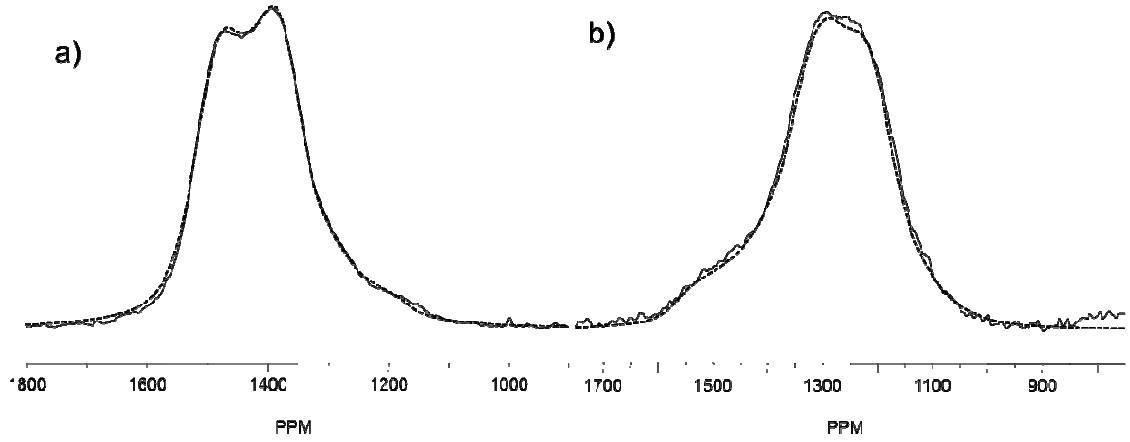


Figure S2: Simulation of the quadrupolar broadening of the central transition in the ^{23}Na MAS NMR spectrum of a) P2- and b) P3-$Na_{2/3}Ni_{1/3}Mn_{2/3}O_2$ using the minimum number of resonances.

Modeling of the ^{23}Na NMR spectra was performed using the program WinSolids1 by K. Eichele.[1] Values for quadrupolar coupling constant (χQC), the asymmetry parameter (η), and the isotropic shift (δiso) were refined. The best fits were obtained with 100% Lorentzian and 50%-50% Lorentzian-Gaussian functions for the P2 and P3 polytypes, respectively.



1. Eichele, K. *WSolids1*, Universität Tübingen (Germany), 2001. http://anorganik.uni-tuebingen.de/klaus/soft/index.php?p=wsolids1/wsolids1.

Table S1. Results of the fits of the central transition in the ^{23}Na MAS NMR spectra of P2- and P3-$Na_{2/3}Ni_{1/3}Mn_{2/3}O_2$, using quadrupolar broadening and the minimum number of peaks. See Figure S2 for visualization of the fits. These results are only meant to illustrate that the complexity of the signals requires more than one peak in the fits.

Polytype	Environment	Rel. Intensity	d_{iso} (ppm)	χ_{QC} (MHz)	h
P2	A	1	1543	3.5	0.3
	B	0.5	1560	3.9	1.0
P3	A	1	1370	3.5	0.4
	B	0.2	1400	3.9	0.9
	C	0.6	1575	4.4	0.7