

Supplementary Information AM-11-047 for

**Structural studies of NH₄-exchanged natrolites at ambient conditions
and high-temperature**

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Supplementary Table 1. EDS chemical composition of the fully and partially NH₄-exchanged natrolites.^a

Fully NH4-exchanged natrolite					
Elements		N	K	Al	Si
Atomic percent (%)	1	9.77	0.00	9.47	12.88
	2	9.72	0.00	9.33	13.05
	3	9.12	0.00	10.30	14.10
	4	9.46	0.00	9.50	13.21
	5	10.00	0.00	9.73	13.57
	6	10.62	0.00	9.50	13.02
Unit cell composition		(NH ₄) _{16.24} Al ₁₆ Si ₂₄ O ₈₀ · xH ₂ O			
Partially NH4-exchanged natrolite					
Elements		N	K	Al	Si
Atomic percent (%)	1	4.30	6.76	10.84	15.90
	2	5.17	6.17	10.34	15.60
	3	2.04	6.64	11.41	18.12
	4	2.46	6.73	11.04	17.89
	5	3.06	6.59	10.42	17.79
	6	3.44	7.40	10.94	16.50
	7	2.33	6.95	11.16	17.85
Unit cell composition		(NH ₄) _{4.78} K _{9.93} Al ₁₆ Si ₂₄ O ₈₀ · xH ₂ O			
Atomic ratio of N ⁺ : K ⁺		0.325 : 0.675			

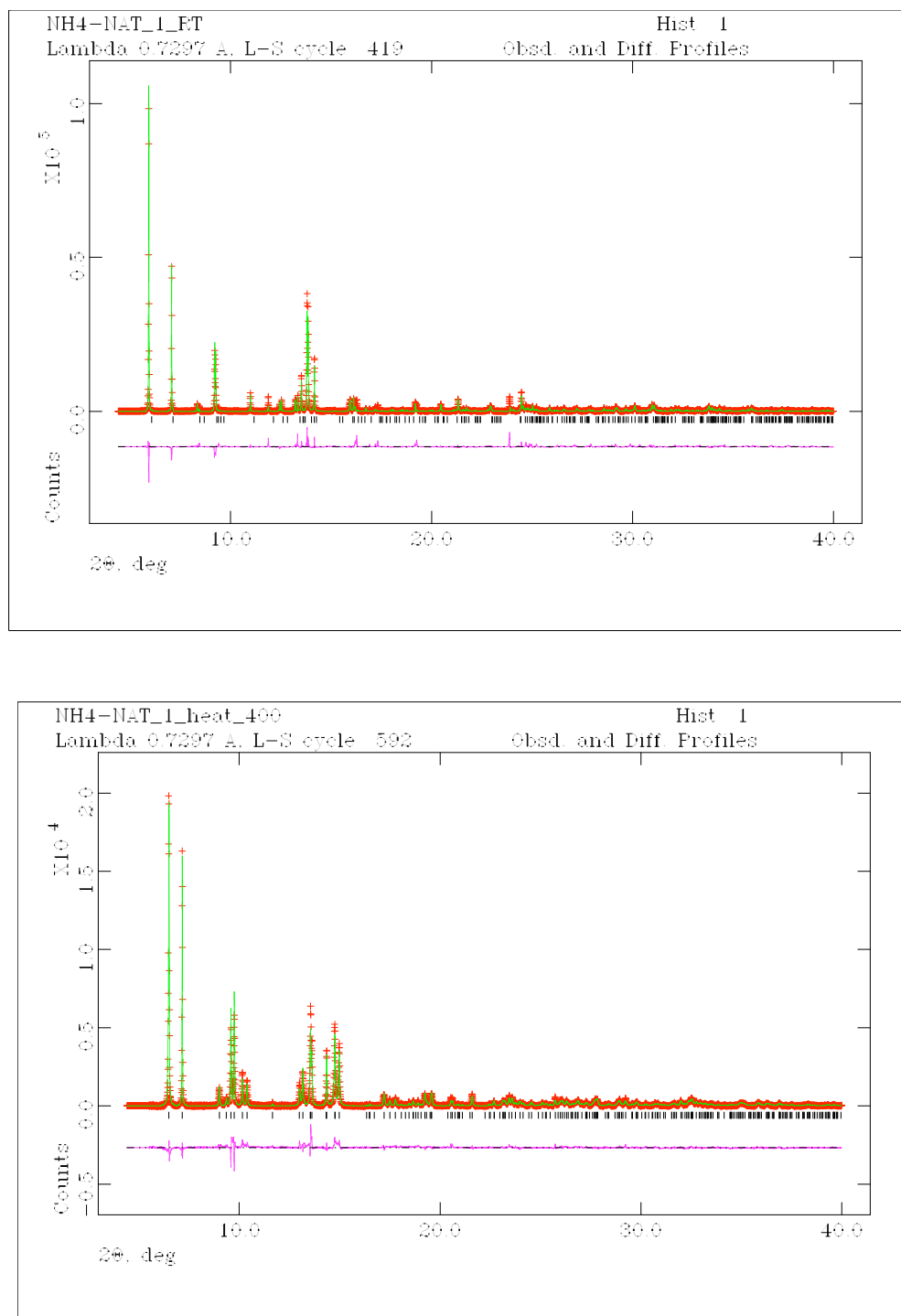
^aValues are normalized based on 16 aluminum atoms per unit cell.

Supplementary Table 2. Unit cell parameters of NH₄- and partially NH₄-exchanged natrolites at different temperatures.^a

Temperature (°C)	Fully NH ₄ -exchanged natrolite				Partially NH ₄ -exchanged natrolite			
	a	b	c	volume	a	b	c	volume
25	19.8368(2)	20.0574(2)	6.5165(1)	2592.74(4)	19.5511(2)	19.9342(2)	6.4923(1)	2530.27(4)
50	19.8424(1)	20.0516(1)	6.5168(1)	2592.85(3)	19.5535(2)	19.9269(2)	6.4925(1)	2529.75(4)
75	19.8465(2)	20.0404(2)	6.5161(1)	2591.65(4)	19.5437(2)	19.9050(2)	6.4906(1)	2524.96(4)
100	19.8482(2)	20.0308(2)	6.5146(1)	2590.04(5)	19.5258(2)	19.8799(2)	6.4876(1)	2518.29(5)
125	19.8464(2)	20.0289(2)	6.5140(1)	2589.31(5)	19.5203(2)	19.8674(2)	6.4867(1)	2515.66(5)
150	17.8927(4)	18.4934(5)	6.5466(1)	2166.2(1)	19.5185(2)	19.8600(1)	6.4871(1)	2514.64(5)
175	17.8881(5)	18.5015(5)	6.5464(1)	2166.6(1)	19.4938(3) ^h 17.657(5) ^d	19.8367(2) ^h 18.527(5) ^d	6.4808(1) ^h 6.523(1) ^d	2506.08(6) ^h 2134.0(7) ^d
200	17.8845(5)	18.5090(5)	6.5463(1)	2167.0(1)	19.484(1) ^h 17.628(1) ^d 16.969(7) ^k	19.827(1) ^h 18.533(1) ^d 18.155(8) ^k	6.4774(2) ^h 6.5252(4) ^d 6.486(2) ^k	2502.3(2) ^h 2131.8(2) ^d 1998.1(8) ^k
225	17.8842(4)	18.5194(5)	6.5479(1)	2168.7(1)	17.5869(9) ^d 16.943(3) ^k	18.513(1) ^d 18.121(5) ^k	6.5227(3) ^d 6.489(2) ^k	2123.7(1) ^d 1992.3(6) ^k
250	17.8826(5)	18.5278(5)	6.5485(1)	2169.7(1)	17.5741(7) ^d 16.9860(9) ^k	18.5348(8) ^d 18.131(1) ^k	6.5321(2) ^d 6.4889(5) ^k	2127.7(2) ^d 1998.5(2) ^k
275	17.8806(5)	18.5377(5)	6.5494(1)	2170.9(1)	17.5585(8) ^d 16.9996(8) ^k	18.5354(6) ^d 18.1538(9) ^k	6.5361(2) ^d 6.4899(6) ^k	2127.2(2) ^d 2002.8(2) ^k
300	17.8779(5)	18.5441(5)	6.5493(1)	2171.3(1)	17.534(1) ^d 17.008(1) ^k	18.524(1) ^d 18.164(1) ^k	6.5346(3) ^d 6.4928(5) ^k	2122.4(2) ^d 2005.8(2) ^k
325	17.8792(5)	18.5528(5)	6.5504(1)	2172.8(1)	17.5129(8) ^d 17.029(1) ^k	18.5233(6) ^d 18.173(1) ^k	6.5373(2) ^d 6.4928(7) ^k	2120.7(2) ^d 2009.3(2) ^k
350	17.8797(5)	18.5620(5)	6.5512(1)	2174.2(1)	17.4947(8) ^d 17.071(2) ^k	18.5247(6) ^d 18.199(3) ^k	6.5365(3) ^d 6.49(1) ^k	2118.4(2) ^d 2016.2(5) ^k
375	17.8806(5)	18.5698(5)	6.5519(1)	2175.5(1)	17.4702(7) ^d 17.076(2) ^k	18.5141(6) ^d 18.207(2) ^k	6.5388(2) ^d 6.491(1) ^k	2114.9(2) ^d 2018.2(4) ^k
400	17.8820(5)	18.5781(5)	6.5527(1)	2176.9(1)	17.4722(5) ^d 17.095(2) ^k	18.5303(4) ^d 18.2219(2) ^k	6.5430(1) ^d 6.484(1) ^k	2118.4(1) ^d 2019.8(4) ^k
425	17.8770(5)	18.5896(5)	6.5530(1)	2177.7(1)	17.4481(7) ^d 17.149(2) ^k	18.5104(6) ^d 18.269(2) ^k	6.5428(2) ^d 6.478(1) ^k	2113.1(2) ^d 2029.5(4) ^k
450					17.4335(5) ^d 17.263(2) ^k	18.5056(4) ^d 18.306(3) ^k	6.5468(2) ^d 6.502(1) ^k	2112.1(1) ^d 2054.9(5) ^k
475					17.4196(4) ^d 17.344(2) ^k	18.5118(4) ^d 18.455(2) ^k	6.5420(1) ^d 6.5137(9) ^k	2109.6(1) ^d 2085.0(1) ^k
Cooled down to RT					16.9152(6) ^k	18.0634(7) ^k	6.4703(2) ^k	1977.0(1) ^k

^aEsd's are in parentheses. In the case of the partially NH₄-exchanged natrolite, three or two different phases coexist from 175°C; ^hthe original hydrated phase, ^dthe partially NH₄-exchanged natrolite in the dehydrated state, and ^kthe K-rich phase separated from the partially NH₄-exchanged natrolite.

Supplementary Figure 1. Rietveld refinement fits of the structural models of the fully NH₄-exchanged natrolite at RT (upper) and at 400 °C (lower) using synchrotron X-ray powder diffraction data. Points shown represent the observed data. The continuous lines through the sets of points are the calculated profiles from the structure refinements summarized in Tables 1-2. The sets of tic marks below the data indicate the positions of the allowed reflections. The lower curves represent the differences between observed and calculated profiles ($I_{\text{obs}} - I_{\text{calc}}$) plotted on the same scale as the observed data.



Supplementary Figure 2. Rietveld refinement fits of the structural models of the partially NH₄-exchanged natrolite at RT (upper), at 400°C (middle), and after one week of exposure to atmospheric conditions (lower) using synchrotron X-ray powder diffraction data. Points shown represent the observed data. The continuous lines through the sets of points are the calculated profiles from the structure refinements summarized in Tables 1-2. The sets of tic marks below the data indicate the positions of the allowed reflections. In the case of the fit to the 400°C data, the K-rich phase is included as a minor secondary phase. The lower curves represent the differences between observed and calculated profiles ($I_{\text{obs}} - I_{\text{calc}}$) plotted on the same scale as the observed data.

