

Supplementary Information for the manuscript

Natrolite is not a “soda-stone” anymore:

Structural study of alkali (Li^+), alkaline-earth (Ca^{2+} , Sr^{2+} , Ba^{2+}) and heavy metal (Cd^{2+} , Pb^{2+} , Ag^+) cation-exchanged natrolites

Yongjae Lee,* Donghoon Seoung, Yongmoon Lee

Department of Earth System Sciences, Yonsei University, Seoul 120-749, Korea

(office) +82-2-2123-5667, (e-mail) YongjaeLee@yonsei.ac.kr

389 Supplementary Table1. EDS chemical composition of the fully cation-exchanged natrolites.^a

Elements		Atomic percent (%) ^c					Composition
		1	2	3	4	5	
Ca-NAT	Ca	5.63	5.93	4.45	5.69	6.64	Ca _{7.7(1)} Al ₁₆ Si ₂₄ O ₈₀ ·xH ₂ O
	K	0.00	0.00	0.00	0.00	0.00	
	Al	11.94	12.01	11.13	12.43	12.43	
Sr-NAT	Sr	5.93	5.62	6.36	5.96	5.76	Sr _{7.6(1)} Al ₁₆ Si ₂₄ O ₈₀ ·xH ₂ O
	K	0.00	0.00	0.00	0.00	0.00	
	Al	12.41	12.49	12.95	11.96	11.37	
Ba-NAT	Ba	5.74	6.26	6.17	6.48	6.74	Ba _{7.9(1)} Al ₁₆ Si ₂₄ O ₈₀ ·xH ₂ O
	K	0.00	0.00	0.00	0.00	0.00	
	Al	12.59	13.08	13.3	12.87	12.07	
Ag-NAT	Ag	12.78	12.31	12.32	12.32	13.05	Ag _{16.4(1)} Al ₁₆ Si ₂₄ O ₈₀ ·xH ₂ O
	K	0.00	0.00	0.00	0.00	0.00	
	Al	12.20	12.24	12.3	12.64	11.7	
Cd-NAT	Cd	6.39	6.19	6.38	6.3	6.52	Cd _{8.2(1)} Al ₁₆ Si ₂₄ O ₈₀ ·xH ₂ O
	K	0.00	0.00	0.00	0.08	0.14	
	Al	12.07	12.9	12.42	11.99	12.72	
Pb-NAT	Pb	6.63	6.11	6.58	6.28	6.24	Pb _{8.2(1)} Al ₁₆ Si ₂₄ O ₈₀ ·xH ₂ O
	K	0.00	0.00	0.00	0.00	0.01	
	Al	12.74	11.47	12.54	12.85	12.24	
Li-NAT	K	0.00	0.07	0.00	0.00	0.07	Li _{15.97(1)} K _{0.03(1)} Al ₁₆ Si ₂₄ O ₈₀ ·xH ₂ O ^b
	Al	13.57	14.39	13.41	12.97	12.54	

^aValues are normalized based on 16 aluminum atoms per unit cell.
^bLithium contents are estimated based on the measured potassium contents.
^cInstrumental detection limit down to the third decimal point.

419
420

Supplementary Table2. Strcutural comparison between the Ca-NAT model and that of natural scolecite by Comodi et al. (2002).

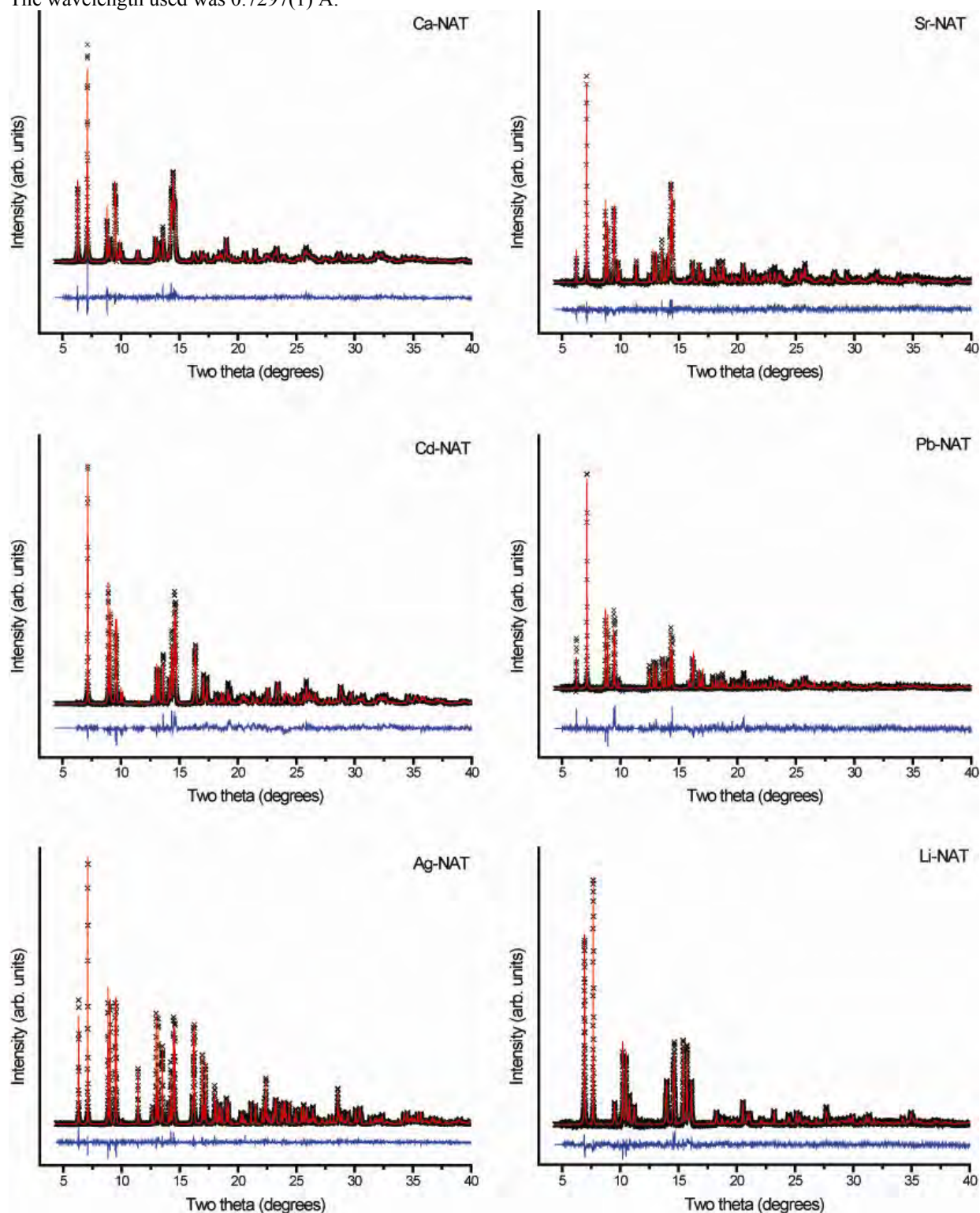
Scolecite			
a	6.533(2)		
b	19.030(3)		
c	9.830(3)		
alpha	90		
beta	109.95(3)		
gamma	90		
volume	1148.76(4)		
	x	y	z
Ca	0.1613(1)	0.14323(2)	0.0521(1)
Si1	0.5	0.37057(3)	0
Si2	0.2298(1)	0.33184(3)	0.2001(1)
Si3	0.5400(1)	0.08257(3)	0.3312(1)
Al1	0.9344(1)	0.46209(3)	0.0998(1)
Al2	0.3555(1)	0.21662(3)	0.4338(1)
O1	0.5420(3)	0.0315(1)	0.4608(2)
O2	0.4472(3)	0.0460(1)	0.1744(2)
O3	0.3836(3)	0.1513(1)	0.3156(2)
O4	0.1154(3)	0.1998(1)	0.4681(2)
O5	0.3535(3)	0.2994(1)	0.3852(2)
O6	0.0868(3)	0.2712(1)	0.0905(2)
O7	0.4143(3)	0.3587(1)	0.1345(2)
O8	0.0767(3)	0.3956(1)	0.2148(2)
O9	0.7894(3)	0.1101(1)	0.3565(2)
O10	0.6602(3)	0.4369(1)	0.0342(2)
Wat1	0.8908(4)	0.0803(1)	0.1083(3)
Wat2	0.9085(5)	0.326(1)	0.4404(4)
Wat3	0.5782(4)	0.4454(1)	0.3742(2)

Ca-NAT			
a	6.5242(3)		
b	18.9885(7)		
c	9.8466(5)		
alpha	90		
beta	109.70(1)		
gamma	90		
volume	1148.4(1)		
	x	y	z
Ca	0.157(1)	0.1412(3)	0.0562(7)
Si1	0.503(1)	0.3690(4)	0.0001(8)
Si2	0.229(1)	0.3335(3)	0.1997(7)
Si3	0.549(1)	0.0826(4)	0.3337(7)
Al1	0.933(1)	0.4628(4)	0.0989(7)
Al2	0.356(1)	0.2186(4)	0.4391(7)
O1	0.544(2)	0.0276(5)	0.459(1)
O2	0.447(2)	0.0453(5)	0.1776(9)
O3	0.401(2)	0.1520(5)	0.330(1)
O4	0.116(2)	0.2038(5)	0.477(1)
O5	0.342(2)	0.3008(5)	0.3595(9)
O6	0.085(1)	0.2730(5)	0.0937(9)
O7	0.425(2)	0.3573(6)	0.139(1)
O8	0.086(2)	0.3995(5)	0.221(1)
O9	0.797(1)	0.1104(6)	0.357(1)
O10	0.658(1)	0.4385(5)	0.037(1)
Wat1	0.874(3)	0.0755(8)	0.086(2)
Wat2	0.903(3)	0.3256(8)	0.429(2)
Wat3	0.568(3)	0.4537(7)	0.354(2)

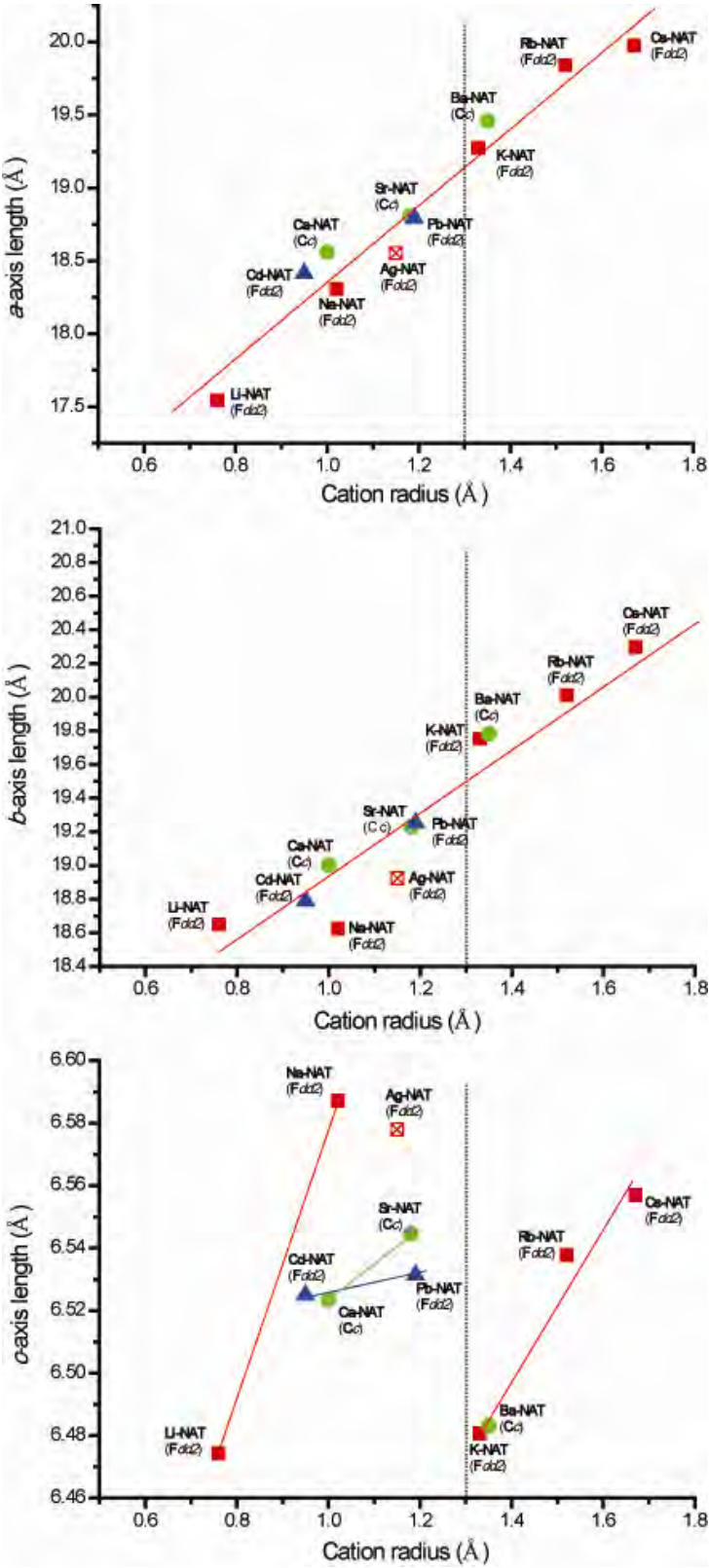
Difference			
a	0.0088(10)		
b	0.0415(15)		
c	-0.0166(15)		
alpha	0		
beta	0.25(2)		
gamma	0		
volume	0.36(5)		
	x	y	z
Ca	0.0043(1)	0.00203(2)	-0.0041(4)
Si1	-0.0032(5)	0.00157(20)	-0.0001(4)
Si2	0.0008(5)	-0.00166(15)	0.0004(4)
Si3	-0.009(5)	0.00003(2)	-0.0025(4)
Al1	0.0014(5)	-0.00071(2)	0.0009(4)
Al2	-0.0005(5)	-0.002(1)	-0.0053(4)
O1	-0.0020(10)	0.0039(3)	0.0018(5)
O2	0.0002(10)	0.0007(3)	-0.0032(5)
O3	-0.0174(10)	-0.0007(3)	-0.0144(5)
O4	-0.0006(10)	-0.0040(3)	-0.0089(5)
O5	0.0115(10)	-0.0014(3)	0.0257(5)
O6	0.0018(5)	-0.0018(3)	-0.0032(5)
O7	-0.0107(10)	0.0014(3)	-0.0045(5)
O8	-0.0093(10)	-0.0039(3)	-0.0062(5)
O9	-0.0076(5)	-0.0003(3)	-0.0005(5)
O10	0.0022(2)	-0.0016(3)	-0.0028(5)
Wat1	0.0168(15)	0.0048(4)	0.0223(10)
Wat2	0.0055(15)	0.0004(4)	0.0114(10)
Wat3	0.0102(15)	-0.0083(4)	0.0202(10)

421

Supplementary Figure 1. The Rietveld refinement fits of the structural models of Ca-NAT, Sr-NAT, Cd-NAT, Pb-NAT, Ag-NAT, and Li-NAT to the synchrotron X-ray powder diffraction data measured at room temperature. Backgrounds were subtracted from the data. Points represent observed data. The continuous lines are the calculated profiles. The lower curves represent the differences between the observed and calculated profiles ($I_{\text{obs}} - I_{\text{calc}}$) plotted on the same scale as the observed data. The wavelength used was 0.7297(1) Å.



Supplementary Figure 2. The refined unit cell lengths of the caion-exchanged natrolites, normalized to the orthorhombic unit cell. Data for Na-NAT are from the work of Baur et al. (1990), and K-, Rb-, and Cs-NAT from Lee et al. (2010). The lines are guides to the eyes. The dotted line indicates a possible threshold for the order-disorder transition.



433 Supplementary Figure 3. Changes in the degree of orthorhombicity, defined as $2(b-a)/(b+a)$, of the caion-exchanged natrolites
 434 plotted as a function of the cation radius.
 435

