

Tables:

Table 1: Total Si and Na concentrations in solutions at the end of syntheses and minerals identified for series of experiments at $\text{pH}_i = 12$ and $\text{pH}_i = 13.3$. pH_i = initial pH of the starting solutions at 25 °C. pH_f = final pH at the end of the synthesis (cooling fluids at 25 °C).

	Time (day)	Sample	pH_i	pH_f	[Si] $\text{mol}\cdot\text{L}^{-1}$	[Na] $\text{mol}\cdot\text{L}^{-1}$	Minerals
$\text{pH}_i = 12$	1	NT42	11.93	11.10	8.16×10^{-3}	2.68×10^{-2}	nontronite
	6	NT0	12.03	11.44	1.47×10^{-2}	1.47×10^{-2}	nontronite
	15	NT38	12.01	11.66	1.02×10^{-2}	3.13×10^{-2}	nontronite
	31	NT28	11.97	11.64	1.17×10^{-2}	3.29×10^{-2}	nontronite
	62	NT10	12.03	11.36	1.37×10^{-2}	3.49×10^{-2}	nontronite
	183	NT33	11.99	11.57	1.46×10^{-2}	3.87×10^{-2}	nontronite + ϵ aegirine
$\text{pH}_i = 13.3$	1	NT44	13.33	13.13	3.14×10^{-2}	2.37×10^{-1}	nontronite
	6	NT7	13.30	13.22	4.31×10^{-2}	3.04×10^{-1}	nontronite
	15	NT41	13.35	13.15	4.20×10^{-2}	2.51×10^{-1}	nontronite + ϵ aegirine
	31	NT32	13.29	13.23	3.35×10^{-2}	2.61×10^{-1}	nontronite + aegirine + hematite
	62	NT17	13.30	13.22	1.90×10^{-2}	2.59×10^{-1}	aegirine + hematite
	183	NT35	13.31	13.33	7.63×10^{-3}	3.01×10^{-1}	aegirine + hematite

Table 2: Thermodynamic activities of aqueous species of H^+ , Na^+ , and H_4SiO_4 at 150 °C for the series of experiments at $\text{pH}_i = 12$ and $\text{pH}_i = 13.3$.

	Time	Sample	a(H^+)	a(Na^+)	a(H_4SiO_4)
$\text{pH}_i = 12$	1	NT42	1.55E-10	2.16E-02	6.81E-04
	6	NT0	1.78E-10	2.46E-02	1.38E-03
	15	NT38	1.35E-10	2.48E-02	7.31E-04
	31	NT28	1.41E-10	2.60E-02	8.73E-04
	62	NT10	1.41E-10	2.74E-02	1.02E-03
	183	NT33	1.26E-10	3.01E-02	9.60E-04
$\text{pH}_i = 13.3$	1	NT44	2.00E-11	1.52E-01	1.71E-04
	6	NT7	2.00E-11	1.66E-01	2.29E-04
	15	NT41	2.09E-11	1.61E-01	2.41E-04
	31	NT32	1.82E-11	1.66E-01	1.56E-04
	62	NT17	1.70E-11	1.64E-01	8.11E-05
	183	NT35	1.38E-11	1.89E-01	2.30E-05

Table 3: Thermodynamic data of aegirine, hematite, ferripyrophyllite, and nontronites $[Si_{4-x}Fe(III)_x]Fe(III)_2O_{10}(OH)_2Na_x$ with $x = 0.5, 0.75, 1.15$, and 1.35 . ΔG°_f = standard Gibbs free energy of formation; ΔH°_f = standard enthalpy of formation; S° = standard entropy; C_p = heat capacity; K_{eq} $150\text{ }^\circ\text{C}$ = equilibrium constant of dissolution at $150\text{ }^\circ\text{C}$

	ΔG°_f (kJ·mol ⁻¹)	ΔH°_f (kJ·mol ⁻¹)	S° (J·K ⁻¹ ·mol ⁻¹)	C_p (J·K ⁻¹ ·mol ⁻¹)	$\log_{10}(K_{eq})$ $150\text{ }^\circ\text{C}$
Aegirine	-2417.20	-2584.50	170.57	169.88	-1.81
Hematite	-744.25	-826.23	87.4	103.88	-6.82
amorphous silica	-850.36	-903.15	46.90	59.89	-2.00
ferripyrophyllite	-4382.44	-4739.82	292.73	328.72	-13.70
Fe(III)- nontronite ($x = 0.5$)	-4324.36	-4678.20	334.51	374.67	-13.23
Fe(III)- nontronite ($x = 0.75$)	-4292.30	-4645.54	351.43	397.65	-12.56
Fe(III)- nontronite ($x = 1.15$)	-4257.94	-4611.66	364.76	420.62	-11.55
Fe(III)- nontronite ($x = 1.35$)	-4220.80	-4573.38	386.54	448.19	-10.93

Table 4: Tetrahedral iron content ($[^{[4]}\text{Fe} \text{ (III)}]$) of nontronites synthesized at $\text{pH}_i = 12$ and 13, estimated from the position ($\pm 0.5 \text{ cm}^{-1}$) of the $\nu \text{ Si-O}$ band (Baron et al., 2016). The Full Width at Half Maximum (FWHM) and the intensity of the $\nu \text{ Fe}^{3+}_2 - \text{OH}$ band were obtained from the decomposition of IR spectra (normalized to the $\nu \text{ Si-O}$ band) using one pseudo-Voigt component.

	Time (day)	Sample	$\nu \text{ Si-O}$ (cm$^{-1}$)	$[^{[4]}\text{Fe(III)}$	$\nu \text{ Fe}^{3+}_2 - \text{OH}$	Intensity (a.u.)
$\text{pH}_i = 12$	1	NT42	1012	0.46	72	0.131
	6	NT0	1009	0.51	64	0.163
	15	NT38	1004	0.61	59	0.159
	31	NT28	1002	0.64	56	0.167
	62	NT10	997	0.73	51	0.177
	183	NT33	997	0.73	52	0.185
$\text{pH}_i = 13.3$	1	NT44	974	1.16		
	6	NT7	965	1.32		
	15	NT41	962	1.38		