

## REVISION 1

Table A1 Experimental conditions; this table shows additional data to those in Table 1.

Experiment No.	1	2	3	4	5	6	7
Pressure at the outlet of precipitation tube [MPa] <sup>1</sup>	30.9 (0.3)	31.8 (0.5)	31.4 (0.5)	31.0 (0.8)	30.9 (1.6)	30.5 (0.5)	30.5 (1.0)
Duration [hours]	113	76	83	80	91	87	76
Flow rate [g/min] <sup>2</sup>	1.9 (0.2)	2.1 (0.1)	2.1 (0.1)	2.0 (0.1)	1.8 (0.2)	1.8 (0.1)	1.6 (0.3)
Residence time [min] <sup>3</sup>	4.7 (0.4)	4.3 (0.1)	4.0 (0.2)	4.1 (0.1)	4.4 (0.4)	4.5 (0.3)	5.1 (0.9)
Total weight of precipitation [mg]	1491	1536	2725	2658	4573	4619	4645
Weight change [mg/h] <sup>4</sup>	13	20	33	33	50	53	61
Temperature in the vessel for the dissolution of albite [°C]	-	160	182	210	200	330	240
<b>Solution<sup>5</sup></b>							
Sampling number	11	3	3	3	3	3	3
pH <sup>6</sup> in <sup>7</sup>	5.5 (0.6)	5.9 (0.0)	6.7 (0.1)	6.6 (0.1)	6.6 (0)	7.9 (0.1)	7.2 (0.3)
out <sup>7</sup>	6.0 (0.7)	6.0 (0.1)	6.6 (0.2)	6.5 (0.2)	6.7 (0.3)	8.2 (0.3)	7.1 (0.4)
Concentrations [ppm]							
Si in	301 (28)	268 (11)	375 (5)	331 (8)	316 (26)	318 (18)	355 (15)
out	224 (30)	181 (4)	209 (15)	162 (16)	118 (14)	104 (3)	98 (7)
Ω <sup>8</sup> AmSil in	1.3	1.2	1.7	1.5	1.4	1.4	1.6
out	1.0	0.8	1.0	0.7	0.5	0.5	0.4
Qtz in	3.8	3.5	4.9	4.3	4.1	4.1	4.6
out	3.0	2.4	2.7	2.1	1.5	1.3	1.3
Al in	0.0 (-)	0.4 (0)	1.4 (0.2)	2.3 (0.1)	3.0 (0.1)	5.6 (0.9)	6.7 (0.6)
out	0.0 (-)	0.0 (0)	0.1 (0)	0.0 (0)	0.1 (0.1)	0.0 (0)	0.1 (0)
Na in	0.0 (-)	0.7 (0)	1.5 (0.2)	2.5 (0.3)	3.5 (0.1)	8.2 (1.1)	7.6 (0.7)
out	0.0 (-)	0.3 (0)	0.3 (0.1)	0.5 (0.1)	1.0 (0.1)	3.4 (0.1)	1.4 (0.4)

<sup>1</sup>Average pressures recorded by a datalogger during multiple runs (standard deviation).

<sup>2</sup>Average flow rates calculated based on the sampling time and weight of the solution (standard deviation).

<sup>3</sup>Average residence time in the precipitation vessel.

<sup>4</sup>Total weight of precipitate divided by the duration.

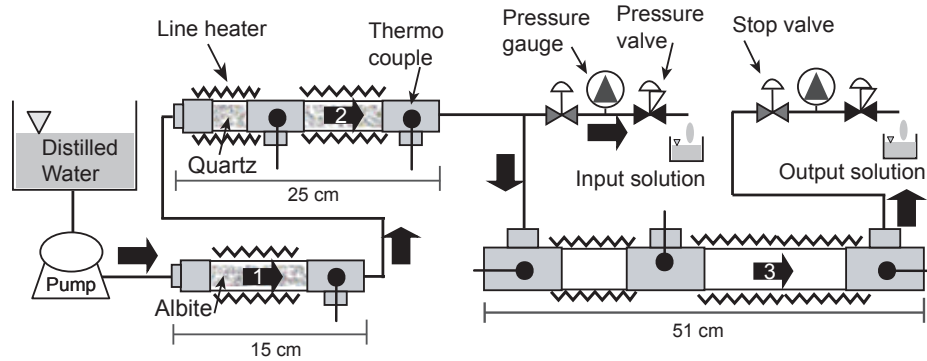
<sup>5</sup>Average value (standard deviation).

<sup>6</sup>at room temperature.

<sup>7</sup>in = input solution, out = output solution (solution before and after passing through the precipitation vessel, respectively).

<sup>8</sup>Saturation index of the input and output solution for amorphous silica (AmSil) and quartz (Qtz).

Continued -- more appendix material



Caption for Supplementary Figure

Fig. A1 Schematic illustration of the apparatus used for hydrothermal flow-through experiments. The apparatus consists of three reaction vessels made of stainless steel (SUS316) to create a horizontal flow path. The fluid pressure is maintained at  $31 \pm 1$  MPa using a back-pressure valve (Maximum pressure of the pump is 50 MPa). The flow rate is controlled at constant value  $2.0 \pm 0.5 \text{ g min}^{-1}$  by the pump. The input solution is prepared by the dissolution of albite sand (grain size, 1–2 mm; weight, 17 g) in the first vessel (length 15 cm) and by the dissolution of quartz sand (grain size, 1–2 mm; weight, 30 g) in the second vessel (length 25 cm). The concentrations of Al and Na (0–7 ppm) are controlled by varying temperature of the first vessel from 160 to 330 °C. The temperature of the second vessel is kept at 360 °C. The chemistry of the input solution is not in equilibrium with albite and quartz, but is probably kinetically controlled. After passing through the first and second vessels, silica minerals are precipitated at 430 °C in the third vessel (length 51 cm). This  $P$ - $T$  condition (430 °C and 31 MPa) is the same as that of our previous work (Okamoto et al. 2010, GCA). No rock/mineral substrate was used in the third vessel. Although we did not control the temperatures of the thin lines (internal diameter, 1.59 mm) connecting the three vessels, no silica precipitation occurred in these lines.

Table A2 Experimental conditions (data of Exp. 1 at different time points)

Sample No.	Time [hour]	Concentration of Si [ppm]		pH <sup>1</sup>	
		in <sup>2</sup>	out <sup>2</sup>	in	out
1	0.0	246	162	6.8	7.4
2	2.4	260	203	n.m. <sup>3</sup>	6.9
3	6.0	285	221	5.7	5.7
4	24.0	310	250	5.3	6.0
5	44.2	341	197	4.8	5.4
6	48.7	311	246	n.m.	n.m.
7	74.1	284	204	5.4	5.5
8	95.2	309	248	n.m.	n.m.
9	117.9	323	256	5.4	5.7
10	143.9	321	268	n.m.	n.m.
11	165.6	323	214	5.2	5.5
Average <sup>4</sup>		301 (28)	224 (30)	5.5 (0.6)	6.0 (0.7)

<sup>1</sup>at room temperature.

<sup>2</sup>in = input solution, out = output solution (solution before and after passing through the precipitation vessel, respectively).

<sup>3</sup>not measured.

<sup>4</sup>Average value (standard deviation).

Table A3 Experimental conditions (data of Exp. 2-7 at different time points)

Exp.	2		3		4		5		6		7	
Sample No.	start time of sampling from beginning of the run [hours]											
1	22.8		23.6		14.9		15.8		22.9		20.4	
2	46.1		49.0		41.8		39.2		53.6		48.5	
3	n.s. <sup>1</sup>		79.2		60.4		64.6		71.7		72.3	
concentration of components in the solution [ppm]												
Sample No.	in <sup>2</sup>	out <sup>2</sup>	in	out	in	out	in	out	in	out	in	out
<b>Si</b>	1	257	177	223	333	181	282	116	324	100	358	104
	2	278	185	215	340	142	320	102	293	108	335	101
	3	n.s. <sup>1</sup>	n.s.	188	319	165	345	136	337	103	371	88
Average <sup>3</sup>	1	268	184	209	338	162	326	118	318	104	355	98 (7)
	2	0.4	0.0	0.1	2.1	0.1	3.0	0.2	6.5	0.1	6.7	0.1
	3	0.4	0.0	0.1	2.4	0.0	3.1	0.1	4.4	0.0	6.0	0.1
	Average	n.s.	n.s.	0.0	2.4	0.0	3.1	0.0	5.8	0.0	7.4	0.1
<b>Na</b>	1	0.4 (0)	0.0	0.1 (0)	0.7	0.0	0.7	0.1	0.9	0.0 (0)	0.6	0.1 (0)
	2	0.7	0.2	0.3	2.2	0.4	3.4	1.2	9.1	3.2	7.4	0.9
	3	0.6	0.3	0.5	2.6	0.6	3.5	1.0	6.7	3.6	6.9	1.4
	Average	n.s.	n.s.	0.2	2.6	0.6	3.6	0.9	8.9	3.5	8.5	1.9
	Average	0.7 (0)	0.3	0.3	2.5	0.5	3.5	1.0	8.2	3.4	7.6	1.4
<b>pH<sup>4</sup></b>	1	5.9	6.1	6.3	6.6	6.2	6.6	6.3	8.0	7.9	6.9	6.6
	2	5.9	6.0	6.6	6.6	6.8	6.6	6.9	7.8	8.4	7.7	7.6
	3	n.s.	n.s.	6.8	6.8	6.6	6.6	6.9	7.9	8.4	7.0	7.1
	Average	5.9 (0)	6.0	6.5	6.6	6.6	6.6	6.9	7.9	8.4	7.3	7.1

<sup>1</sup>no sample collected.

<sup>2</sup>in = input solution, out = output solution (solution before and after passing through the precipitation vessel, respectively).

<sup>3</sup>Average value (standard deviation).

<sup>4</sup>at room temperature.