

Supporting Information for:

Thermodynamic properties of sodium aluminosilicate hydrate (N-A-S-H)

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Appendix A - Quantification of the ^{29}Si MAS and ^1H - ^{29}Si CPMAS NMR data

Table S1: Relative integral area^a for $\text{Q}^4(m\text{Al})$ environments within N-A-S-H in each sample extracted from the deconvoluted ^{29}Si MAS NMR spectra.

Sample ID	<i>N-A-S-H sites</i>					<i>Precursor sites</i>					N-A-S-H total	Precursor total	Si/Al ^b
	Q ⁴ (4Al)	Q ⁴ (3Al)	Q ⁴ (2Al)	Q ⁴ (1Al)	Q ⁴ (0Al)	Q ⁴ (4Al)	Q ⁴ (3Al)	Q ⁴ (2Al)	Q ⁴ (1Al)	Q ⁴ (0Al)			
<i>As cured</i>													
NASH-1	24	12	4	0	0	15	22	15	6	1	40	60	1.15
NASH-1.5	50	26	9	0	0	1	12	3	0	0	85	15	1.15
NASH-2	28	13	8	3	1	1	12	13	7	14	54	46	1.25
GP-1	58	25	9	0	0	0	0	1	2	5	92	8	1.13
GP-1.5	35	33	18	5	0	0	0	1	3	6	90	10	1.30
<i>Dissolution</i>													
NASH-1	27	14	5	0	0	13	20	14	4	1	47	53	1.15
NASH-1.5	34	17	6	0	0	2	17	15	9	0	58	42	1.15
NASH-2	28	13	8	3	1	3	10	13	8	12	54	46	1.25
GP-1	50	22	8	0	0	0	10	5	2	4	79	21	1.13
GP-1.5	36	33	18	5	0	0	0	1	2	5	92	8	1.30
<i>Precursor difference</i>													
NASH-1	-	-	-	-	-	-1	-2	-2	-2	0	-1	-7	-
NASH-1.5	-	-	-	-	-	1	5	12	9	0	-	27	-
NASH-2	-	-	-	-	-	2	-2	0	1	-1	-	0	-
GP-1	-	-	-	-	-	0	9	4	0	-1	-	13	-
GP-1.5	-	-	-	-	-	0	0	0	-1	-1	-	-2	-

^a The relative integrated intensity for each resonance is normalised to the sum of all sites within the reaction product and is obtained by simulating the ^{29}Si MAS NMR spectra. Estimated error in the relative integral area is 1%. ^b The Si/Al of the N-A-S-H gel is calculated using Engelhardt's formula³⁹ and the relative integral areas of the $\text{Q}^4(m\text{Al})$ resonances.

Appendix B – Thermodynamic data for the sodium alkali aluminosilicate gels

Table S2: The ionic coefficients, ionic activities and total ionic strength of ionic species calculated according to Eq. 1 to Eq. 3, using the experimentally measured concentration shown in Figure 8 as input. The only ionic species allowed to form in these systems are: Na⁺, AlO₂⁻, SiO₂@, OH⁻, including trace of K⁺.

Sample ID	Ionic coefficient (γ)					Ionic activity mmol/L					Total ionic strength (mol/kg-H ₂ O)
	Na ⁺	AlO ₂ ⁻	SiO ₂ @	OH ⁻	K ⁺	Na ⁺	AlO ₂ ⁻	SiO ₂ @	OH ⁻	K ⁺	
NASH-1-1d	0.9362	0.9362	1.0008	0.9362	0.9362	5.1584	1.2075	0.3431	0.3243	0.1434	0.0036
NASH-1-3d	0.9344	0.9344	1.0009	0.9344	0.9344	5.3791	1.4533	0.4550	0.3117	0.1236	0.0039
NASH-1-7d	0.9306	0.9306	1.0010	0.9306	0.9306	6.1049	1.7556	0.5429	0.2649	0.0750	0.0044
NASH-1-14d	0.9295	0.9295	1.0010	0.9295	0.9295	6.2236	1.9314	0.5602	0.2587	0.0908	0.0046
NASH-1-28d	0.9274	0.9274	1.0011	0.9274	0.9274	6.7255	2.0554	0.5219	0.1940	0.0795	0.0049
NASH-1-90d	0.9294	0.9294	1.0010	0.9294	0.9294	6.9922	1.4157	0.1744	0.1073	0.0149	0.0046
NASH-1.5-1d	0.9174	0.9174	1.0015	0.9174	0.9174	8.7836	0.1777	2.0526	2.9280	0.1616	0.0066
NASH-1.5-3d	0.9177	0.9177	1.0015	0.9177	0.9177	9.5351	0.2011	3.2873	2.0909	0.1133	0.0065
NASH-1.5-7d	0.9147	0.9147	1.0016	0.9147	0.9147	10.6564	0.2433	4.4546	1.8297	0.2119	0.0071
NASH-1.5-14d	0.9161	0.9161	1.0015	0.9161	0.9161	10.6613	0.1567	4.8502	1.5249	0.1322	0.0068
NASH-1.5-28d	0.9144	0.9144	1.0016	0.9144	0.9144	11.2633	0.1279	5.1476	1.5575	0.0841	0.0071
NASH-1.5-90d	0.9134	0.9134	1.0017	0.9134	0.9134	10.8329	0.0522	4.9622	2.4819	0.0253	0.0073
NASH-2-1d	0.9240	0.9240	1.0012	0.9240	0.9240	8.9269	0.0236	3.6069	0.7055	0.3566	0.0054
NASH-2-3d	0.9229	0.9229	1.0013	0.9229	0.9229	9.3946	0.0162	5.0221	0.6382	0.3026	0.0056
NASH-2-7d	0.9218	0.9218	1.0013	0.9218	0.9218	9.7251	0.0220	5.6285	0.4996	0.4308	0.0058
NASH-2-14d	0.9215	0.9215	1.0013	0.9215	0.9215	10.0977	0.0181	5.8264	0.4068	0.2423	0.0058
NASH-2-28d	0.9193	0.9193	1.0014	0.9193	0.9193	10.6641	0.0137	5.5795	0.3258	0.4447	0.0062
NASH-2-90d	0.9210	0.9210	1.0013	0.9210	0.9210	10.2176	0.0068	5.3936	0.6369	0.0514	0.0059
GP1-1	0.9641	0.9641	1.0002	0.9641	0.9641	1.2864	0.0753	0.0666	0.6273	0.0243	0.0010
GP1-3	0.9460	0.9460	1.0006	0.9460	0.9460	3.4510	0.6872	0.9642	0.6108	0.0316	0.0025
GP1-7	0.9462	0.9462	1.0006	0.9462	0.9462	3.7992	0.6537	0.8503	0.2149	0.0637	0.0025
GP1-14	0.9397	0.9397	1.0007	0.9397	0.9397	4.0980	0.9055	1.1176	0.9812	0.0643	0.0032
GP1-28	0.9421	0.9421	1.0007	0.9421	0.9421	3.9687	1.0498	1.1386	0.5067	0.0247	0.0029
GP1-90	0.9376	0.9376	1.0008	0.9376	0.9376	5.1315	0.8359	0.5911	0.5044	0.0361	0.0035
GP1.5-1	0.9480	0.9480	1.0005	0.9480	0.9480	3.0672	0.3230	0.7256	1.0050	0.0156	0.0023
GP1.5-3	0.9426	0.9426	1.0006	0.9426	0.9426	3.7730	0.4414	1.0174	1.1914	0.0322	0.0029
GP1.5-7	0.9414	0.9414	1.0007	0.9414	0.9414	4.0240	0.5152	1.2016	1.0773	0.0727	0.0030
GP1.5-14	0.9411	0.9411	1.0007	0.9411	0.9411	4.0459	0.6732	1.4291	0.9826	0.0546	0.0031
GP1.5-28	0.9448	0.9448	1.0006	0.9448	0.9448	4.4356	0.5127	0.9061	0.0165	0.0326	0.0026
GP1.5-90	0.9390	0.9390	1.0007	0.9390	0.9390	5.6380	0.5152	0.8076	0.0164	0.0280	0.0033