^{29}Si NMR in Cement: a Theoretical Study on Calcium Silicate Hydrates

Pawel Rejmak, Jorge S. Dolado, Malcolm J. Stott, Andrés Ayuela.

Supporting Information

S1. Periodic models of the C-S-H gel based on the experimental structures of tobermorite 14Å and jennite. See also attached .cif file for structures.

Unit cell formula	Notation	Details
	TOBERMORITE	
$Ca_{20}Si_{24}O_{64}(OH)_8 \cdot (H_2O)_{28}$	T∞	Infinite silicate chains, initial structure from ref. 4
	Pentameric silicate chains	
$Ca_{22}Si_{20}O_{60}(OH)_4 \cdot (H_2O)_{28}$	T5-Ca1	Charge compensation with 2 interlayer Ca^{2+} ions
$Ca_{26}(OH)_4Si_{20}O_{64} \cdot (H2O)_{24}$	T5-Ca2	4 Si(OH) groups replaced with 4 $Ca(OH)^+$ ions
$Ca_{28}(OH)_8Si_{20}O_{64}\ (H_2O)_{20}$	T5-Ca3	Total charge compensation with 8 $Ca(OH)^+$ ions
$Ca_{20}Si_{20}O_{60}(OH)_4 \cdot (H_2O)_{28}$ (H charge compensation)	T5-H1	1 Q^1OH tetrahedron per each pentamer
	T5-H2	2 Q^1OH tetrahedra per pentamer in one layer and 2 Q^1 tetrahedra per pentamer in the next layer
	T5-H3	2 Q^1OH tetrahedra per each pentamer, Q^2b tetrahedra not protonated
	Dimeric silicate chains.	
$Ca_{24}Si_{16}O_{56} \cdot (H_2O)_{28}$	T2-Ca	Charge compensation with 4 interlayer Ca^{2+} ions
$Ca_{20}Si_{16}O_{48}(OH)_8 \cdot (H_2O)_{28}$ (H charge compensation)	T2-H1	1 Q^1OH tetrahedra per each dimer
	T2-H2	One layer fully protonated (Q^1OH) and the next one deprotonated (Q^1 only)
	JENNITE	
$Ca_{18}(OH)_{12}Si_{12}O_{36} \cdot (H_2O)_{16}$	J∞	Infinite silicate chains, initial structure from ref. 3
	Pentameric silicate chains	
$Ca_{18}(OH)_{12}Si_{10}O_{32}(H_2O)_{16}$	J5-1	2 nearest Q^2b tetrahedra removed from J∞
	J5-2	2 next nearest Q^2b tetrahedra removed from J∞
$Ca_{17}(OH)_{14}Si_{10}O_{32}(OH)_4 \cdot (H_2O)_{14}$	J5-H	4 Q^1OH tetrahedra replaced 1 Ca^{2+} ion and 2 protons from H_2O molecules in J5-2 model
	Dimeric silicate chains	
$Ca_{18}(OH)_{12}Si_8O_{28} \cdot (H_2O)_{16}$	J2	
$Ca_{17}(OH)_{14}Si_8O_{24}(OH)_4 \cdot (H_2O)_{14}$	J2-H	4 Q^1OH tetrahedra replaced 1 Ca^{2+} ion and 2 protons from H_2O molecules in J2 model

S2. Geometries of the cluster models of α-quartz and β-belite in xyz format. These models were cut from the periodic structures optimized by GULP and used for ADF calculations. The outermost Si-O bonds were saturated with H atoms, to avoid artificial open-shell systems. The isotropic NMR shieldings were calculated at the innermost Si atoms, printed in bold style.

S3a Quartz cluster model

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XYZ
H          6.115700000   18.207300000   26.531300000
H          8.345100000   14.882100000   26.312700000
H          8.533100000   22.395700000   21.184800000
H          9.491600000   20.748100000   24.530500000
H          3.697600000   22.395700000   21.184800000
H          4.656200000   20.748100000   24.530500000
O         11.700600000   19.228500000   22.587800000
O          9.282800000   18.460300000   21.967000000
Si        10.763900000   18.692900000   21.386300000
O          8.885300000   20.394600000   23.749500000
H         12.667900000   19.076600000   22.967100000
H         11.909500000   16.560000000   24.530500000
O          6.865100000   19.228500000   22.587800000
O          6.467800000   17.293800000   26.151800000
O          8.342100000   17.916400000   24.369600000
O          4.447300000   18.460300000   21.967000000
Si         5.928400000   18.692900000   21.386300000
O          4.049800000   20.394600000   23.749500000
Si         8.346200000   18.995900000   23.168500000
Si         7.400200000   16.750200000   24.950700000
O          2.029700000   19.228500000   22.587800000
O          3.506600000   17.916400000   24.369600000
O         -0.388100000   18.460300000   21.967000000
Si         1.093000000   18.692900000   21.386300000
Si         3.510700000   18.995900000   23.168500000
H          2.891500000   17.154800000   24.749100000
H         -0.997100000   18.808500000   22.748200000
O         14.118500000   15.040500000   22.587800000
O         11.700700000   14.272300000   21.967000000
Si        13.181800000   14.504900000   21.386300000
O         11.303200000   16.206600000   23.749500000
H         15.085700000   14.888600000   22.967100000
O          9.283000000   15.040500000   22.587800000
O         10.760000000   13.728400000   24.369600000
O          6.865200000   14.272300000   21.967000000
Si         8.346300000   14.504900000   21.386300000
O          6.467700000   16.206600000   23.749500000
Si        10.764100000   14.807900000   23.168500000
O          8.342300000   15.584000000   25.531800000
```

H	10.144900000	12.966700000	24.749100000
O	4.447500000	15.040500000	22.587800000
O	5.924500000	13.728400000	24.369600000
O	2.029700000	14.272300000	21.967000000
Si	3.510800000	14.504900000	21.386300000
Si	5.928600000	14.807900000	23.168500000
H	5.309400000	12.966700000	24.749100000
H	1.420700000	14.620500000	22.748200000
H	10.762700000	10.694500000	20.966200000
H	5.927200000	10.694500000	20.966200000
H	14.326900000	20.748500000	19.184000000
O	8.885100000	21.482200000	20.805300000
O	10.759500000	22.104800000	19.023000000
H	10.762100000	22.806700000	18.242100000
Si	9.817600000	20.938600000	19.604200000
O	4.049700000	21.482200000	20.805300000
O	5.924000000	22.104800000	19.023000000
H	5.926600000	22.806700000	18.242100000
Si	4.982100000	20.938600000	19.604200000
H	0.473500000	20.534000000	19.805800000
O	14.118100000	18.460700000	16.620400000
H	15.085400000	18.612700000	16.241200000
O	13.720600000	20.395100000	18.403000000
O	11.700400000	19.228900000	17.241300000
O	11.303000000	17.294200000	20.805300000
O	13.177300000	17.916800000	19.023000000
O	9.282600000	18.460700000	16.620400000
Si	10.763700000	18.693300000	16.039800000
O	8.885100000	20.395100000	18.403000000
Si	13.181400000	18.996300000	17.822000000
O	10.759700000	19.772400000	20.185200000
Si	12.235500000	16.750600000	19.604200000
O	6.864900000	19.228900000	17.241300000
O	6.467500000	17.294200000	20.805300000
O	8.341900000	17.916800000	19.023000000
O	4.447100000	18.460700000	16.620400000
Si	5.928200000	18.693300000	16.039800000
O	4.049600000	20.395100000	18.403000000
Si	8.346000000	18.996300000	17.822000000
O	5.924200000	19.772400000	20.185200000
Si	**7.400000000**	**16.750600000**	**19.604200000**
O	2.029400000	19.228900000	17.241300000
O	1.632100000	17.294200000	20.805300000
O	3.506400000	17.916800000	19.023000000
H	1.420400000	18.880700000	16.460000000
Si	3.510500000	18.996300000	17.822000000
O	1.088700000	19.772400000	20.185200000
Si	2.564500000	16.750600000	19.604200000
O	13.720900000	13.106200000	20.805300000
O	11.700500000	14.272700000	16.620400000

H	12.667800000	14.424600000	16.241200000
O	11.303000000	16.207000000	18.403000000
O	13.177600000	15.584400000	20.185200000
H	14.327200000	12.752700000	20.024300000
O	9.282800000	15.040900000	17.241300000
O	8.885400000	13.106200000	20.805300000
O	10.759700000	13.728800000	19.023000000
O	6.865000000	14.272700000	16.620400000
Si	8.346100000	14.505300000	16.039800000
O	6.467500000	16.207000000	18.403000000
Si	10.763800000	14.808300000	17.822000000
O	8.342100000	15.584400000	20.185200000
Si	9.817900000	12.562600000	19.604200000
O	4.447300000	15.040900000	17.241300000
O	4.049900000	13.106200000	20.805300000
O	5.924300000	13.728800000	19.023000000
O	2.029500000	14.272700000	16.620400000
Si	3.510600000	14.505300000	16.039800000
O	1.632000000	16.207000000	18.403000000
Si	5.928300000	14.808300000	17.822000000
O	3.506600000	15.584400000	20.185200000
Si	4.982400000	12.562600000	19.604200000
O	-0.388200000	15.040900000	17.241300000
O	1.088800000	13.728800000	19.023000000
H	-0.997200000	14.692700000	16.460000000
Si	1.092900000	14.808300000	17.822000000
H	0.473700000	12.967100000	19.402600000
O	8.885400000	12.019000000	18.403000000
O	10.759900000	11.396400000	20.185200000
O	4.049900000	12.019000000	18.403000000
H	8.533200000	11.105500000	18.023600000
O	5.924500000	11.396400000	20.185200000
H	3.697800000	11.105500000	18.023600000
H	10.144200000	20.534400000	14.459200000
H	5.308800000	20.534400000	14.459200000
O	11.302800000	17.294600000	15.458700000
O	10.759500000	19.772900000	14.838700000
H	11.909100000	16.941100000	14.677700000
O	6.467300000	17.294600000	15.458700000
O	8.341600000	17.917200000	13.676500000
H	8.344300000	18.619100000	12.895600000
O	5.924000000	19.772900000	14.838700000
Si	7.399800000	16.751000000	14.257600000
H	2.891100000	16.346400000	14.459200000
O	8.885200000	13.106600000	15.458700000
O	6.467300000	16.207400000	13.056500000
O	8.341900000	15.584800000	14.838700000
H	9.491500000	12.753100000	14.677700000
O	4.049700000	13.106600000	15.458700000
H	6.115100000	15.293900000	12.677000000

O	3.506400000	15.584800000	14.838700000
H	4.656000000	12.753100000	14.677700000

S2b Belite cluster model

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XYZ

H	4.563900000	6.990500000	5.953200000
H	4.563900000	11.601200000	5.953200000
Si	4.011300000	9.295800000	5.658800000
O	4.846200000	7.990600000	6.103500000
O	4.846200000	10.601100000	6.103500000
O	2.512600000	9.295800000	6.353200000
O	3.750900000	9.295800000	4.027900000
H	2.015800000	10.052800000	6.884900000
H	3.914800000	10.052700000	3.318900000
H	3.687800000	3.527100000	8.800700000
Si	4.240500000	5.832400000	9.095000000
O	3.405500000	4.527200000	8.650400000
O	5.739100000	5.832400000	8.400600000
O	4.500900000	5.832400000	10.726000000
O	3.405500000	7.137600000	8.650400000
H	4.337000000	5.075500000	11.435000000
H	6.437600000	3.527100000	3.636700000
Si	6.990200000	5.832400000	3.931100000
O	6.155300000	4.527200000	3.486400000
O	8.488900000	5.832400000	3.236700000
O	7.250600000	5.832400000	5.562000000
O	6.155300000	7.137600000	3.486400000
H	7.086700000	5.075500000	6.271000000
H	8.985700000	5.075500000	2.704900000
H	10.063500000	6.990500000	5.567500000
H	10.063500000	11.601200000	5.567500000
Si	9.510800000	9.295800000	5.273200000
O	10.345800000	7.990600000	5.717800000
O	10.345800000	10.601100000	5.717800000
O	8.012200000	9.295800000	5.967600000
O	9.250400000	9.295800000	3.642200000
H	9.414300000	8.538900000	2.933200000
H	9.187400000	3.527100000	8.415100000
Si	9.740000000	5.832400000	8.709400000
O	8.905100000	4.527200000	8.264800000
O	8.905100000	7.137600000	8.264800000
O	10.000400000	5.832400000	10.340300000
O	11.238700000	5.832400000	8.015000000
H	11.735500000	5.075500000	7.483200000
H	9.836500000	5.075500000	11.049300000
H	6.664600000	1.612100000	8.097200000
O	6.500700000	2.369000000	8.806200000

O	5.262400000	2.369000000	11.131500000
Si	6.761100000	2.369000000	10.437100000
O	7.596000000	3.674200000	10.881800000
O	7.596000000	1.063800000	10.881800000
H	7.058300000	0.199300000	10.624600000
H	4.765600000	1.612100000	11.663200000
H	0.938000000	3.527100000	13.964600000
Si	1.490700000	5.832400000	14.258900000
O	0.655800000	4.527200000	13.814300000
O	2.989400000	5.832400000	13.564600000
O	1.751100000	5.832400000	15.889900000
O	0.655800000	7.137600000	13.814300000
H	1.587200000	5.075500000	16.598900000
H	4.564000000	6.990500000	15.895400000
H	4.564000000	11.601200000	15.895400000
Si	4.011300000	9.295800000	15.601100000
O	4.846200000	7.990600000	16.045700000
O	4.846200000	10.601100000	16.045700000
O	2.512600000	9.295800000	16.295400000
O	3.750900000	9.295800000	13.970100000
H	2.015800000	8.538900000	16.827100000
H	1.814200000	6.990500000	11.117100000
O	1.001100000	9.295800000	9.191800000
Si	1.261500000	9.295800000	10.822800000
O	2.096500000	10.601100000	11.267400000
O	2.096500000	7.990600000	11.267400000
O	-0.237100000	9.295800000	11.517100000
H	-0.763200000	10.153100000	11.215500000
H	0.457500000	10.153200000	8.923700000
H	1.814200000	11.601200000	11.117100000
Ca	4.014700000	9.295800000	8.974700000
Si	4.240500000	12.759300000	9.095000000
O	3.405500000	11.454000000	8.650400000
O	5.739100000	12.759300000	8.400600000
O	4.500900000	12.759300000	10.726000000
H	3.687800000	15.064600000	8.800700000
O	3.405500000	14.064500000	8.650400000
H	4.337000000	13.516200000	11.435000000
Ca	6.764400000	9.295800000	3.810800000
Ca	9.514200000	9.295800000	8.589100000
Ca	6.875600000	7.564100000	7.184100000
Ca	6.875600000	11.027600000	7.184100000
Si	6.990200000	12.759300000	3.931100000
O	6.155300000	11.454000000	3.486400000
O	8.488900000	12.759300000	3.236700000
O	7.250600000	12.759300000	5.562000000
O	6.155300000	14.064500000	3.486400000
H	6.437600000	15.064600000	3.636700000
H	7.086700000	13.516200000	6.271000000
H	8.985700000	13.516200000	2.704900000

Si	9.740000000	12.759300000	8.709400000
O	8.905100000	11.454000000	8.264800000
O	10.000400000	12.759300000	10.340300000
O	8.905100000	14.064500000	8.264800000
O	11.238700000	12.759300000	8.015000000
H	9.187400000	15.064600000	8.415100000
H	11.735500000	13.516200000	7.483200000
H	9.836500000	13.516200000	11.049300000
O	6.500700000	9.295800000	8.806200000
O	5.262400000	9.295800000	11.131500000
Si	**6.761100000**	**9.295800000**	**10.437100000**
O	7.596000000	10.601100000	10.881800000
O	7.596000000	7.990600000	10.881800000
Ca	1.264900000	9.295800000	14.138700000
Ca	4.125900000	7.564100000	12.348000000
Ca	4.125900000	11.027600000	12.348000000
Si	1.490700000	12.759300000	14.258900000
O	0.655800000	11.454000000	13.814300000
O	2.989400000	12.759300000	13.564600000
O	1.751100000	12.759300000	15.889900000
H	0.938000000	15.064600000	13.964600000
O	0.655800000	14.064500000	13.814300000
H	1.587200000	13.516200000	16.598900000
H	4.765600000	16.979600000	11.663200000
O	5.262400000	16.222700000	11.131500000
O	6.500700000	16.222700000	8.806200000
H	7.313700000	18.528000000	10.731500000
Si	6.761100000	16.222700000	10.437100000
O	7.596000000	17.527900000	10.881800000
O	7.596000000	14.917500000	10.881800000
H	6.664600000	16.979600000	8.097200000
H	6.437600000	3.527100000	13.579000000
H	7.086700000	5.075500000	16.213300000
Si	6.990200000	5.832400000	13.873300000
O	6.155300000	4.527200000	13.428700000
O	8.488900000	5.832400000	13.179000000
O	7.250600000	5.832400000	15.504300000
O	6.155300000	7.137600000	13.428700000
H	10.063500000	6.990500000	15.509800000
H	10.063500000	11.601200000	15.509800000
H	7.515400000	10.052700000	16.441500000
Si	9.510800000	9.295800000	15.215500000
O	10.345800000	7.990600000	15.660100000
O	10.345800000	10.601100000	15.660100000
O	8.012200000	9.295800000	15.909800000
O	9.250400000	9.295800000	13.584500000
Ca	6.986900000	5.832400000	10.557400000
Ca	6.764400000	9.295800000	13.753000000
Ca	6.986900000	12.759300000	10.557400000
Ca	9.625400000	7.564100000	11.962400000

Ca	9.625400000	11.027600000	11.962400000
Si	6.990200000	12.759300000	13.873300000
O	6.155300000	11.454000000	13.428700000
O	8.488900000	12.759300000	13.179000000
O	7.250600000	12.759300000	15.504300000
H	6.437600000	15.064600000	13.579000000
H	7.086700000	13.516200000	16.213300000
O	6.155300000	14.064500000	13.428700000
O	10.761900000	9.295800000	10.745900000
O	12.000200000	9.295800000	8.420600000
Si	12.260600000	9.295800000	10.051500000
O	13.095500000	10.601100000	10.496200000
O	13.095500000	7.990600000	10.496200000
H	12.813200000	11.601200000	10.345900000
H	12.813200000	6.990500000	10.345900000
H	12.164100000	8.538900000	7.711600000
H	11.937100000	3.527100000	13.193400000
H	12.586300000	5.075500000	15.827700000
Si	12.489800000	5.832400000	13.487700000
O	11.654800000	4.527200000	13.043100000
O	13.988400000	5.832400000	12.793300000
O	12.750200000	5.832400000	15.118700000
O	11.654800000	7.137600000	13.043100000
H	14.485200000	5.075500000	12.261600000
Ca	12.264000000	9.295800000	13.367400000
Si	12.489800000	12.759300000	13.487700000
O	11.654800000	11.454000000	13.043100000
O	13.988400000	12.759300000	12.793300000
O	12.750200000	12.759300000	15.118700000
H	12.586300000	13.516200000	15.827700000
O	11.654800000	14.064500000	13.043100000
H	11.937100000	15.064600000	13.193400000
H	14.485200000	13.516200000	12.261600000

S3. GULP parametrization used in this work. The optimizations at constant pressure were performed with the options *molq* and *fix*. The covalent radius of Ca and Si were set to 0.0 Å, and the intramolecular Coulomb potential applied to water molecules has a substraction of 50%. The Hessian update switched to the Rational Function Optimizer when the gradient norm dropped to 0.001 eV/Å^{-1}.

Species				
	Core charge	Shell charge	Spring (eV/Å^{-2})	
Ca	2.00000			
Si				
H1	0.40000			Hydroxyl group
H2	0.40000			Water molecule
O1	0.90000	-2.30000	74.9200	Hydroxyl group
O2	1.2500	-2.0500	209.4496	Water molecule
O3	0.86902	-2.86902	74.9200	
Buckingham potential, intermolecular				
	A (eV)	ρ (Å^{-1})	C (eV/Å^6)	R_{cutoff} (Å)
Ca - O3	1090.400	0.34370	0.00000	10.0
Ca – O1, O2	777.270	0.34370	0.00000	10.0
Si - O3	1283.907	0.32052	10.66158	10.0
Si – O1, O2	983.557	0.32052	10.66158	10.0
H - O	311.970	0.25000	0.00000	10.0
O3 - O3	22764.000	0.14900	27.87900	12.0
O3 - O2	22764.000	0.14900	28.92000	12.0
O3 - O1	22764.000	0.14900	13.94000	12.0
O2 - O1	22764.000	0.14900	17.14000	12.0
O1 - O1	22764.000	0.14900	6.97000	12.0
Lennard – Jones potential, intermolecular				
	A (eV/Å^{12})		B(eV/Å^6)	R_{cutoff} (Å)
O2 - O2	39344.98		42.15	12.0
Morse potential, intramolecular				
	D (eV)	α (Å^{-1})	R_0 (Å)	R_{cutoff} (Å)
H1 - O1	7.05250	3.17490	0.94285	1.4
H2 - O2	6.203713	2.22030	0.92376	1.4

Three body harmonic potential			
	k (eV/rad^{-2})	Θ_0 (rad)	R_{cutoff} 1-2, 2-3 (Å) R_{cutoff} 1-3 (Å)
O3,O1 - Si - O3	2.09724	109.470000	1.9 3.5
H2 - O2 -H2 intramolecular	4.19978	108.693195	1.4 2.0

S4. Complete ref 19

Giannozzi, P.; Baroni, S.; Bonini, N.; Calandra, M.; Car, R.; Cavazzoni, C.; Ceresoli, D.; Chiarotti, G. L.; Cococcioni, M.; Dabo, I.; Corso, A. D.; de Gironcoli, S.; Fabris, S.; Fratesi, G.; Gebauer, R.; Gerstmann, U.; Gougoussis, C.; Kokalj, A.; Lazzeri, M.; Martin-Samos, L.; Marzari, N.; Mauri, F.; Mazzarello, R.; Paolini, S.; Pasquarello, A.; Paulatto, L.; Sbraccia, C.; Scandolo, S.; Sclauzero, G.; Seitsonen, A. P.; Smogunov, A.; Umari, P.; Wentzcovitch, R. M. *J. Phys.: Cond. Matter.* **2009**, *21*, 395502.

S5. The convergence of GIPAW isotropic magnetic shieldings (σ) with k-point sampling, energy cutoffs and SCF threshold. Mean values of of isotropic magnetic shieldings (‹σ›) are used for computing the chemical shifts (‹δ›), according to formula (1) (see Main text). The convergence threshold for the self-consistent-field procedure was 10^{-9} Ry.

	σ	
k-points/E cutoff/SCF convergence	α-quartz	β-belite
Γ point/80 Ry/10^{-6}	444.9	432.8
2×2×2/80 Ry/10^{-6}	434.2	411.3
2×2×2/80 Ry/10^{-9}	435.3	409.4
4×4×4/80 Ry/10^{-9}	434.7	408.9
4×4×4/120 Ry/10^{-10}	434.6	408.9

J5-2 model						
	Γ point			(2×2×2) points		
	σ	‹σ›	‹δ›	σ	‹σ›	‹δ›
Q^1	409.4 409.3 410.3 412.5	410.4	-72.7	408.7 409.5 410.3 412.2	410.2	-72.4
Q^2	418.2 416.9 416.5 419.5	417.8	-83.0	417.6 416.1 417.0 418.7	417.4	-82.5
Q^2b	413.9 411.1	412.5	-75.6	413.8 411.1	412.5	-75.6

T5-Ca1 model						
	(2×1×1) points			(4×2×1) points		
	σ	‹σ›	‹δ›	σ	‹σ›	‹δ›
Q^1	412.6	415.3	-80.3	412.6	415.3	-80.3
	413.6			413.7		
	413.8			414.0		
	414.3			414.3		
	416.5			416.4		
	416.8			416.8		
	417.1			417.1		
	417.3			417.3		
Q^2	415.7	417.6	-83.5	416.0	417.7	-83.5
	415.9			416.3		
	417.4			417.3		
	417.6			417.4		
	417.9			418.0		
	417.9			418.3		
	418.3			418.3		
	419.7			419.6		
Q^2b	410.8	414.7	-79.4	410.8	414.8	-79.6
	415.5			415.5		
	415.8			416.0		
	416.8			417.0		

S6. The comparison between GULP results and experimental data for selected silicate minerals: lattice constants (*a*, *b*, *c*), lattice angles (α, β, γ), cell volume (V_{cell}), density (ρ), and elastic properties (K - bulk modulus, G - shear modulus, Y - Young's modulus).

	a *b*[1] (Å) *c*		α β (°) γ		V_{cell}[1] (Å^3) ρ (g/cm^3)		K[2] G[2] (GPa) Y[3]	
	GULP	Exp.	GULP	Exp.	GULP	Exp.	GULP	Exp.
β-Belite	5.51 6.93 9.94	5.50 6.76 9.94	90.0 94.0 90.0	90.0 94.2 90.0	378.7 3.02	346.3 3.30	114.0 53.1 137.9	- - 130-140
Tobermorite 14Å (T∞ model)	6.77 14.65 28.48	6.74 14.86 27.99	90.0 94.8 123.2	90.0 90.0 123.3	2350.0 2.22	2340.8 2.23	38.6 22.5 56.5	- - -
Jennite (J∞ model)	10.66 14.70 10.71	10.58 14.53 10.93	104.1 95.3 110.0	101.3 97.0 109.7	1501.2 2.35	1518.9 2.33	32.0 19.9 49.5	- - -
α-Quartz	4.84 4.84 5.35	4.92 4.92 5.41	90.0 90.0 120.0	90.0 90.0 120.0	108.3 2.76	113.1 2.65	46.5 41.8 96.5	38.0 - -

[1] Values given for supercell doubled along the *b* direction. [2] Hill definition. [3] Calculated from the formula Y = (9G)/(3+(G/K)).

S7 The simulated spectra of (a) tobermorite 14Å (T∞ model) and (b) jennite (J∞ model). The spectra show the chemical shifts of individual Si atoms (dotted line) and the average on the site types of Table 2 (solid line). The peaks are broaden with gaussians with a half-maximum width of 2.0 ppm.