PAUL SCHERRER INSTITUT

PSI

Guoqing Geng
LES Laboratory, PSI

Structure-Property Correlation of Cementitious Phases at Molecular Scale

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limestone + clay

↓

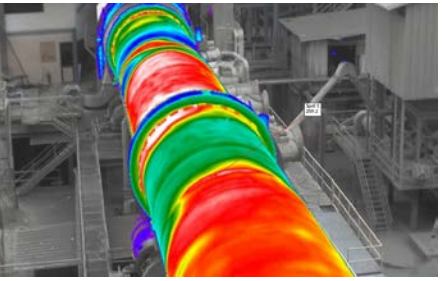
cement + water + rocks → concrete

+ steel fiber/rebar → reinforced concrete

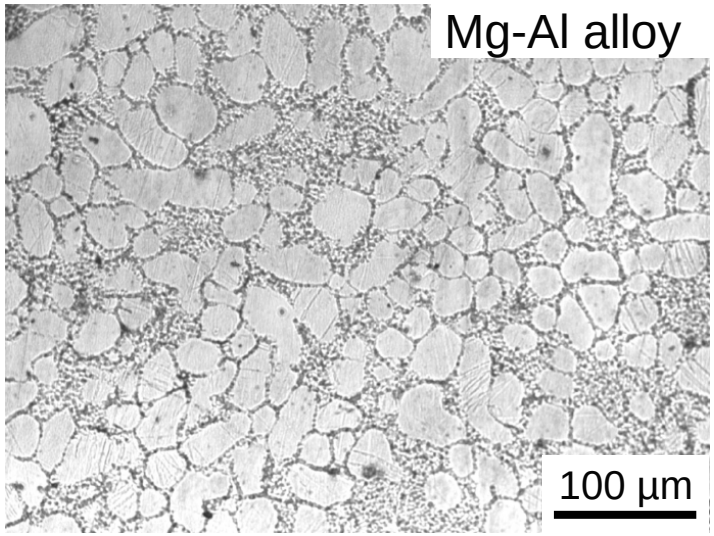
+ industrial waste → green concrete

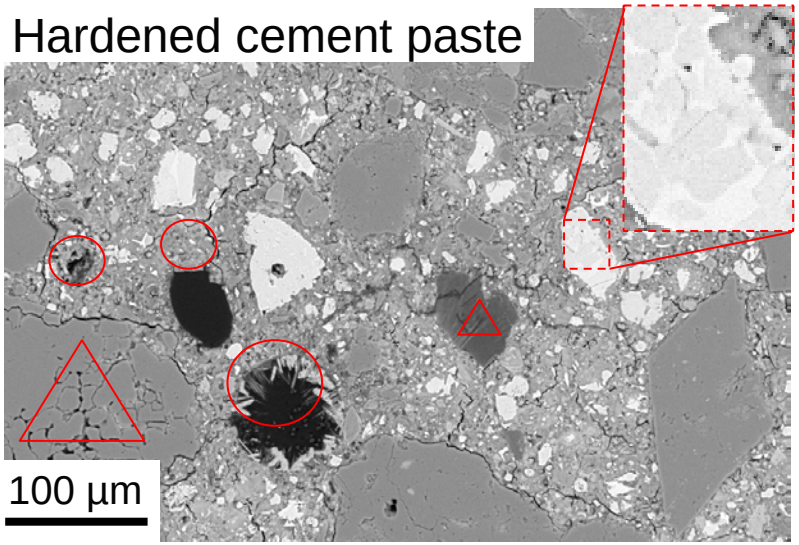
+ plasticizer → high rheology concrete

+ … → … concrete



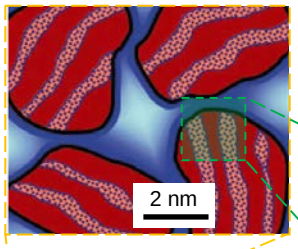

Challenge in from-bottom-up modeling of concrete

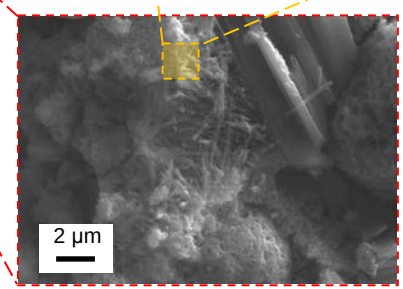




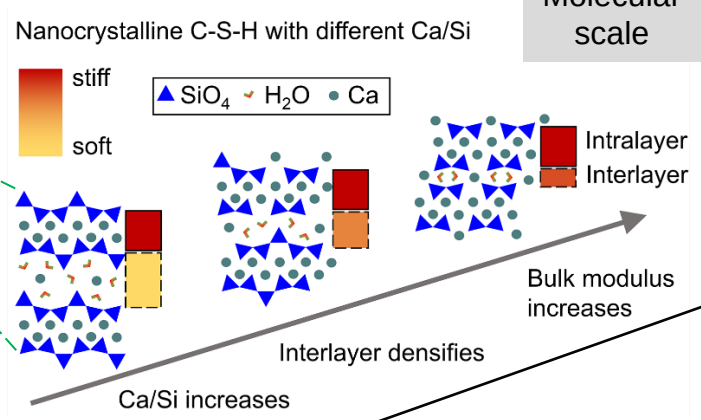
Compositions	A, B, A_mB_n.	aggregates, clinker (C_mS, C_nA, C_4AF), hydrates (C(A)SH, CH, AFt, AFm, CsH, C(A)cH…), admixtures (crystals and glasses in FA, slag etc)…
Spatial scale	atomic, micro, macro	Atomic (<2 nm): chemistry, stiffness Meso (2-10 nm): creep, shrinkage Micro: transport property, shrinkage Macro: performance
Time scale	Constant during service life	Constantly evolving

An overview of the research interest

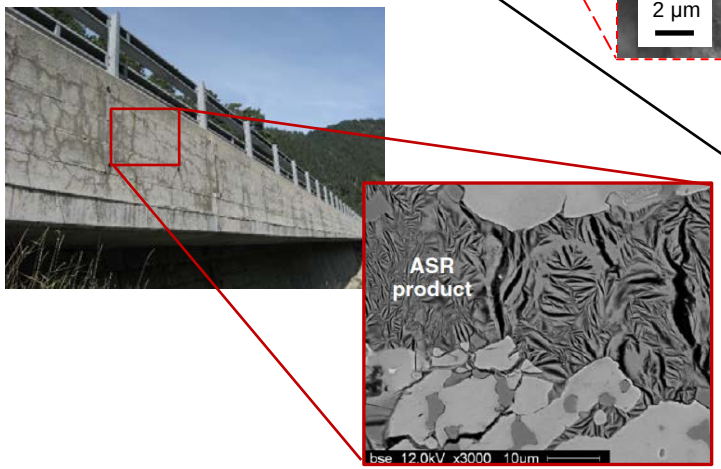




Molecular scale

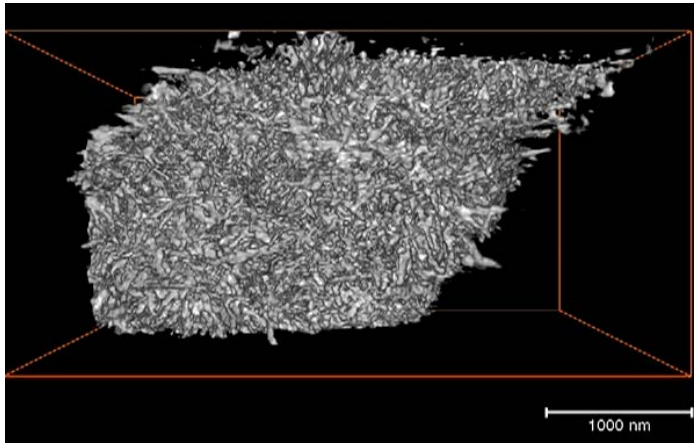


Long-term durability



Multiscale Structure vs. Properties

Mesoscale and microscale

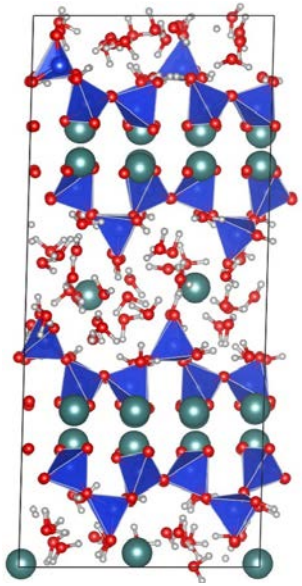


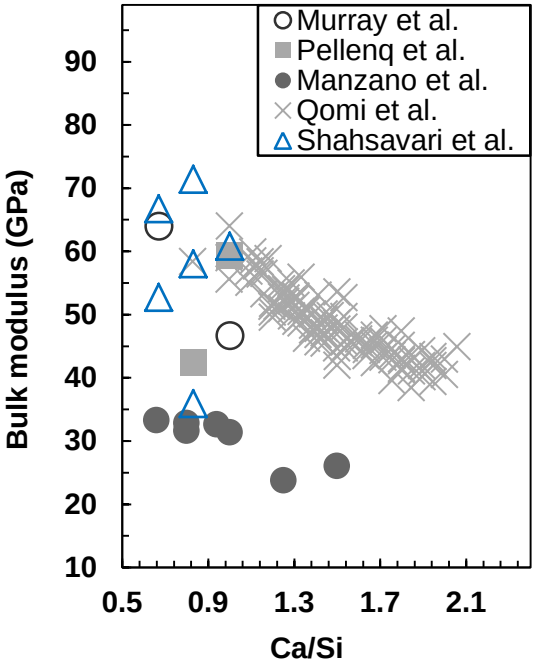
- XANES and EXAFS at Ca, Na, K *K*-edge
- Pair distribution function
- Powder diffraction
- High pressure XRD
- …

- Transport property
- Mechanical property
- …

Discrepancy in reported work

Molecular Calculations *vs* Nanoindentation



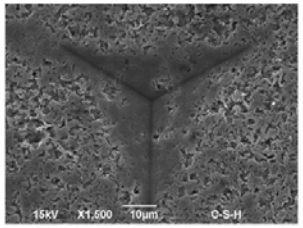


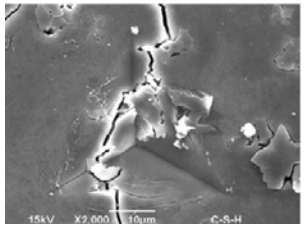
Ca/Si = 0.7 (Figure 4)

32 mN		512 mN	
E (GPa)	*H* (GPa)	*E* (GPa)	*H* (GPa)
29.6	1.77	25.0	0.90
27.5	0.98	23.4	0.98
–	–	–	–
–	–	–	–
25.7	1.07	21.4	0.89
24.7	1.09	26.7	0.91

Ca/Si = 2.1 (Figure 5)

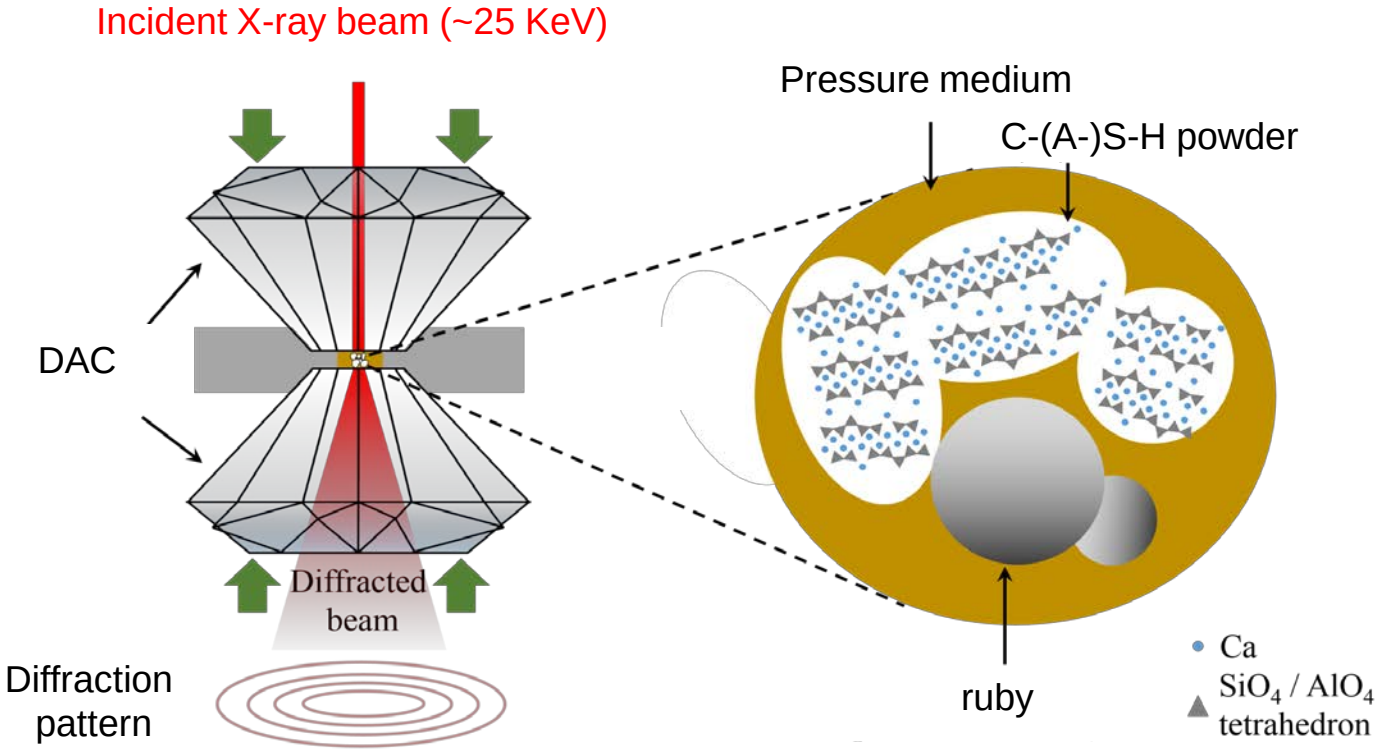
32 mN		512 mN	
E (GPa)	*H* (GPa)	*E* (GPa)	*H* (GPa)
17.4	0.35	16.8	0.44
18.2	0.38	18.1	0.48
24.3	0.72	22.4	0.51
23.7	0.51	25.1	0.56
16.2	0.38	12.8	0.28
–	–	–	–

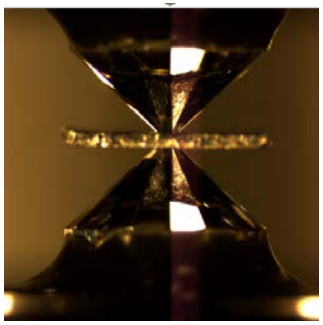


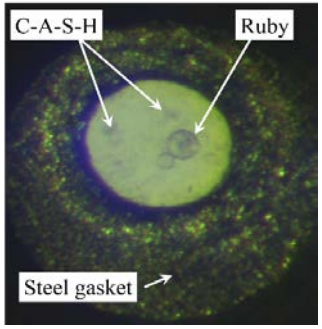


Pelisser, F., Phys. Chem. C. 2012.
Geng, G., et al. Sci. Rep. 2017.

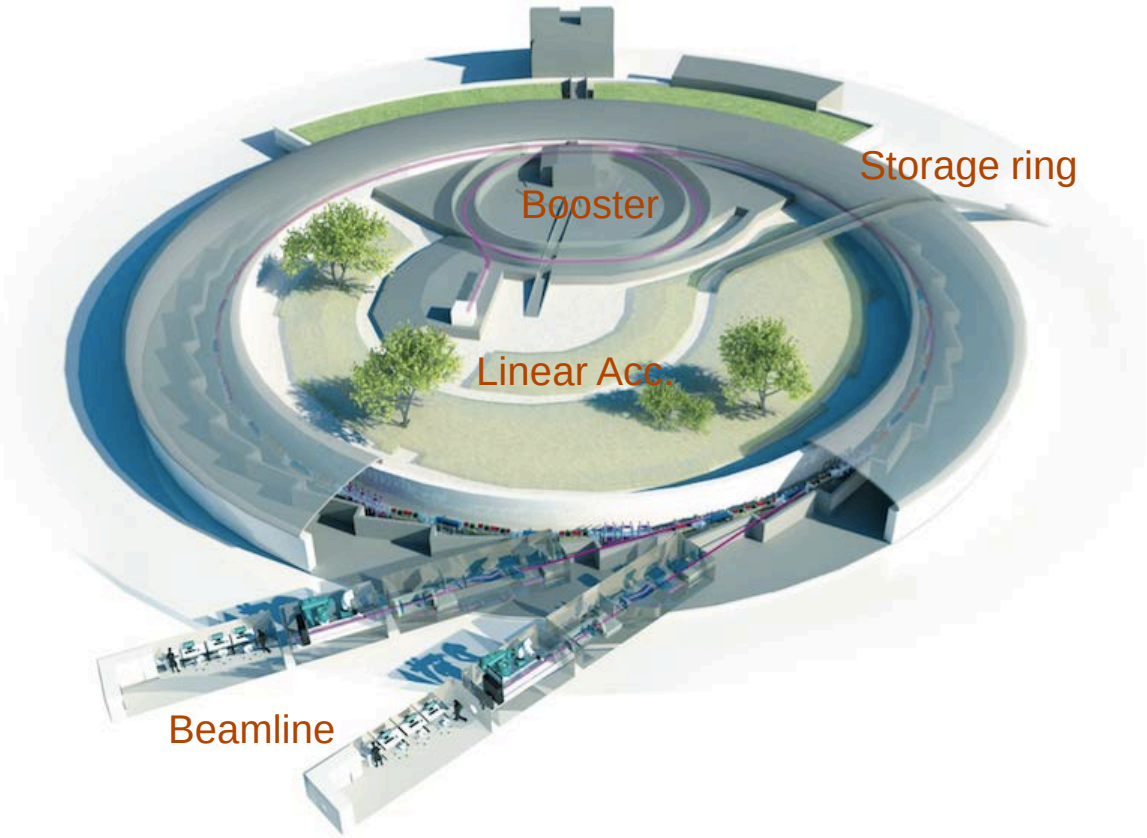
High pressure XRD

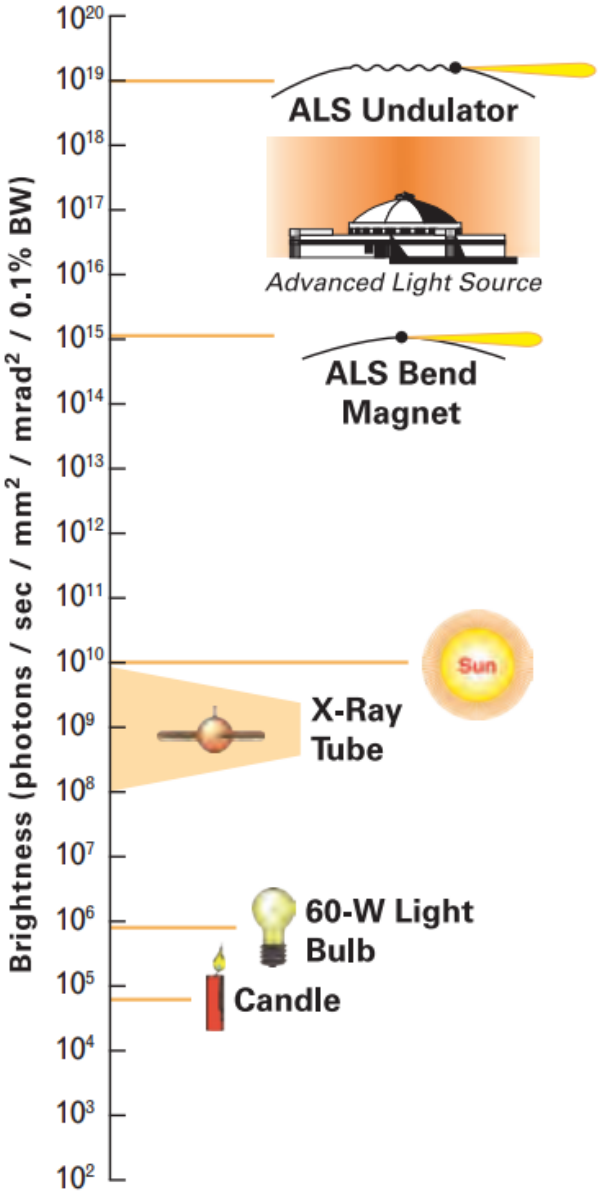






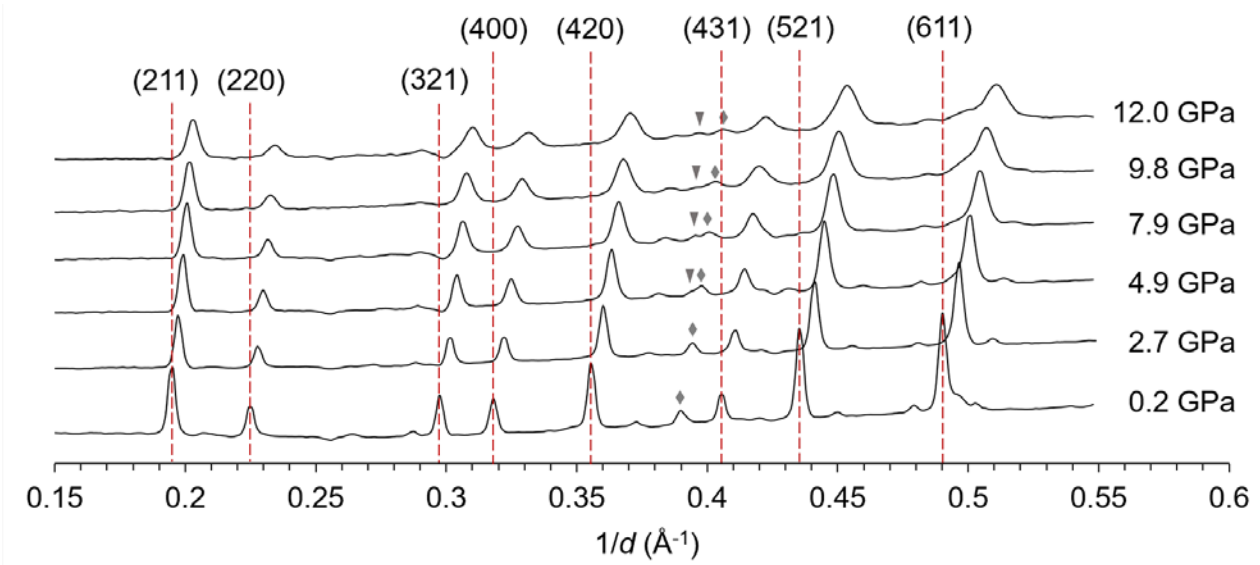
Synchrotron, hooray!





Diffractogram as a function of the applied pressure

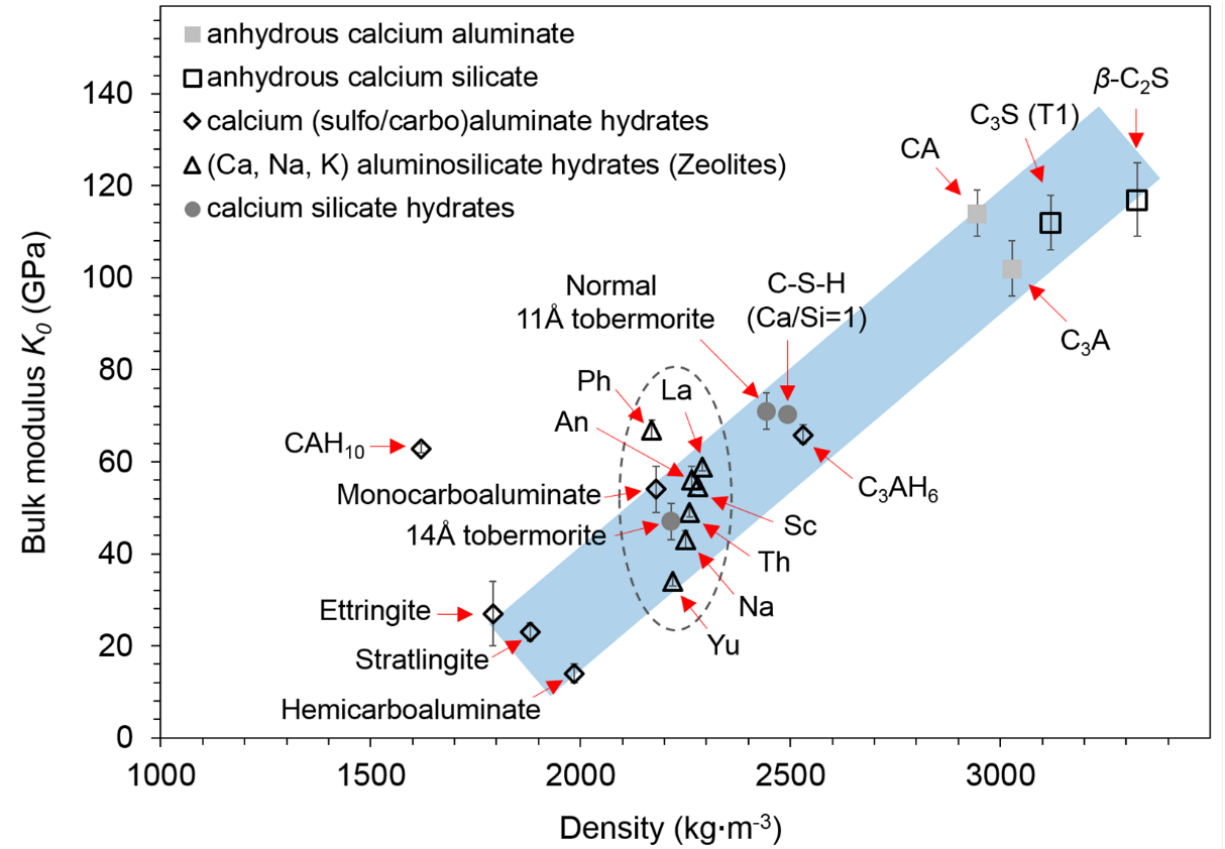
C_3AH_6 (Katoite)



P (GPa)	Interplanar d-spacings (Å)								Refined lattice parameter (Å)		V (Å^3)	V_{error} (Å^3)
	(211)	(220)	(321)	(400)	(420)	(431)	(521)	(611)	a	a_{error}		
0.2	3.978	4.594	6.077	6.497	7.266	8.282	8.899	10.024	12.58	0.01	1988.6	4.3
2.7	4.028	4.656	6.159	6.583	7.361	8.393	9.016	10.148	12.41	0.01	1912.5	4.2
4.9	4.065	4.695	6.213	6.633	7.42	8.467	9.094	10.233	12.31	0.02	1864.2	6.8
7.9	4.098	4.731	6.260	6.69	7.474	8.533	9.164	10.312	12.22	0.01	1824.8	6.1
9.8	4.116	4.746	6.286	6.721	7.515	8.576	9.206	10.366	12.16	0.01	1798	5.4
12	4.144	4.786	6.332	6.773	7.574	8.627	9.280	10.443	12.07	0.01	1757.7	2.2

Geng, G., et al. *Cem Concr Res.* 2018.

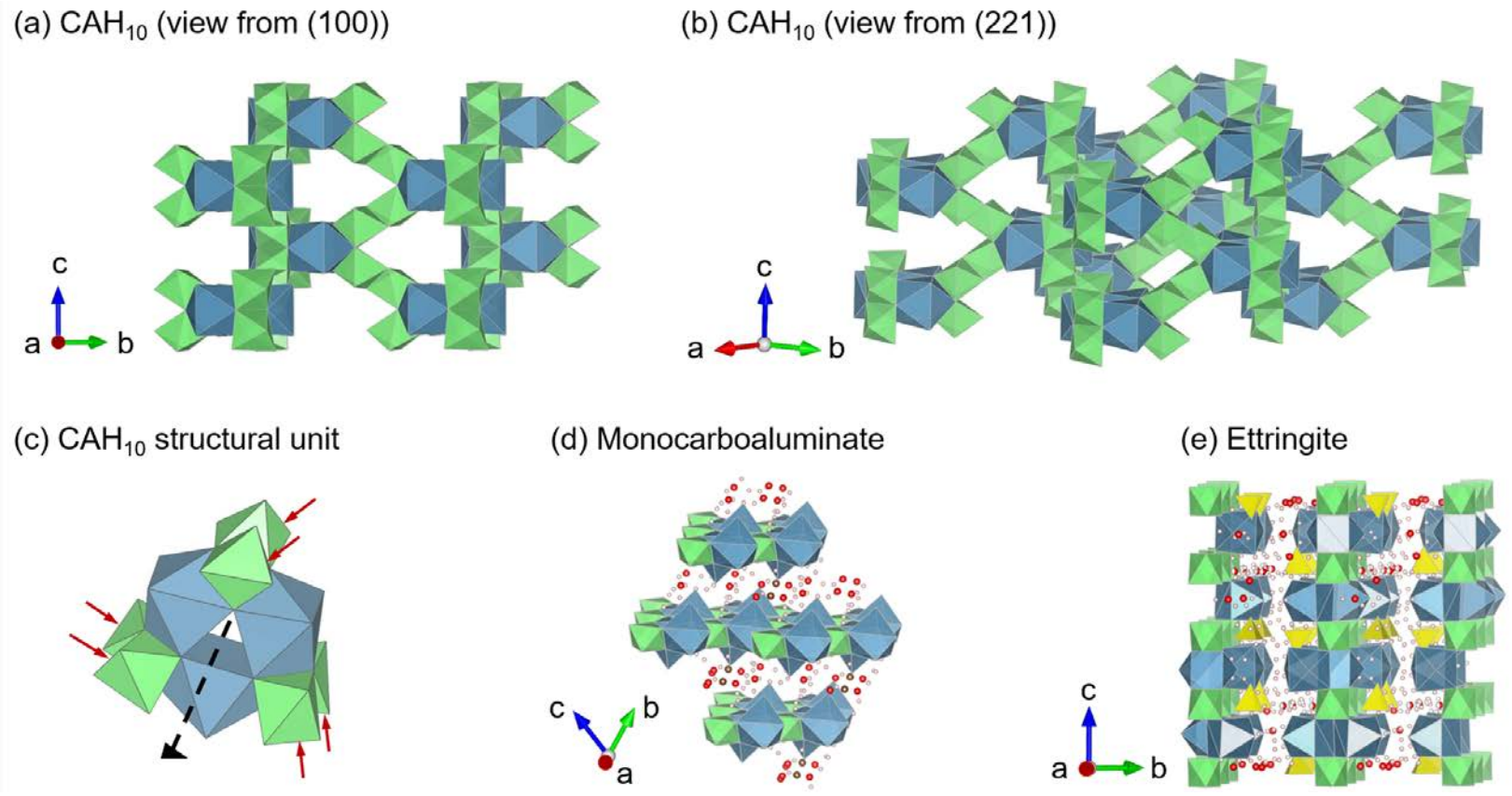
Densification-driven stiffening



The bulk moduli K_0 of several cement-based minerals and zeolites as a function of their densities

Geng, G., et al. *Cem Concr Res.* 2018.

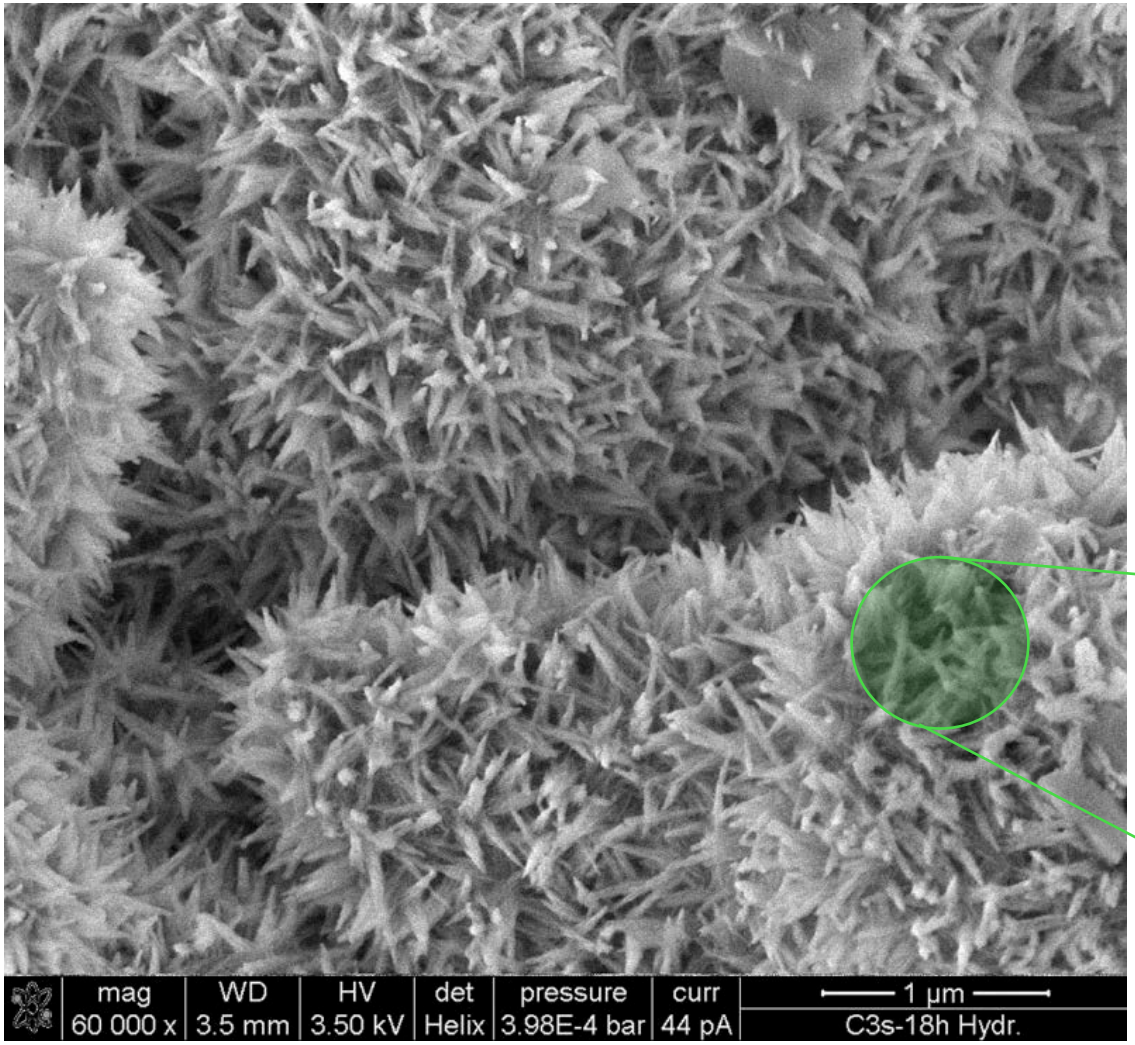
Densification-driven stiffening: exceptions

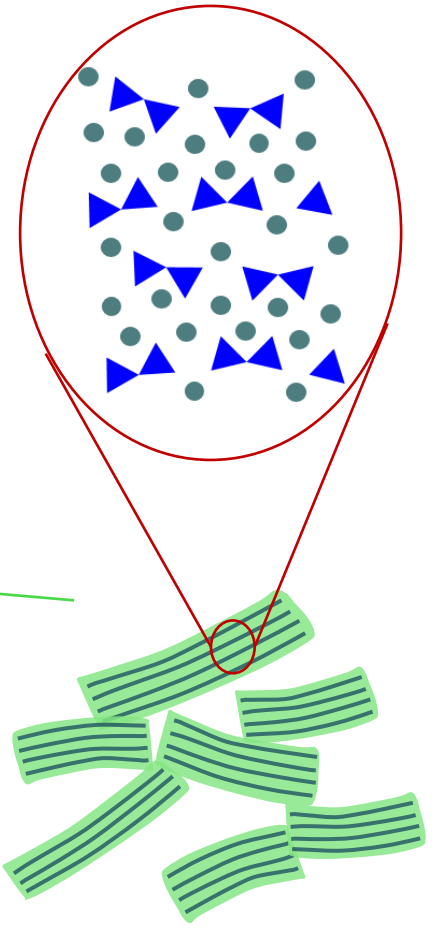


The topological structure of CaO_8 polyhedra (blue), AlO_6 octahedra (green) and SO_4 tetrahedra (yellow) in (a)–(c) CAH_{10}, (d) monocarboaluminate and (e) ettringite. The red, brown and pink spheres are oxygen, carbon and hydrogen atoms, respectively. For viewing convenience, the water and hydroxyl groups in CAH_{10} are not displayed.

Geng, G., et al. *Cem Concr Res.* 2018.

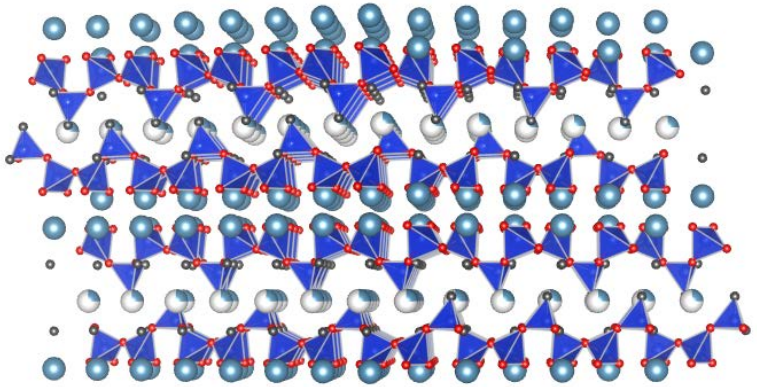
C-(A-)S-H (nanocrystalline? Not a problem!)

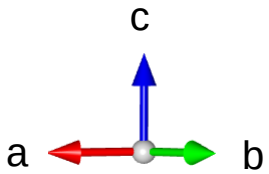


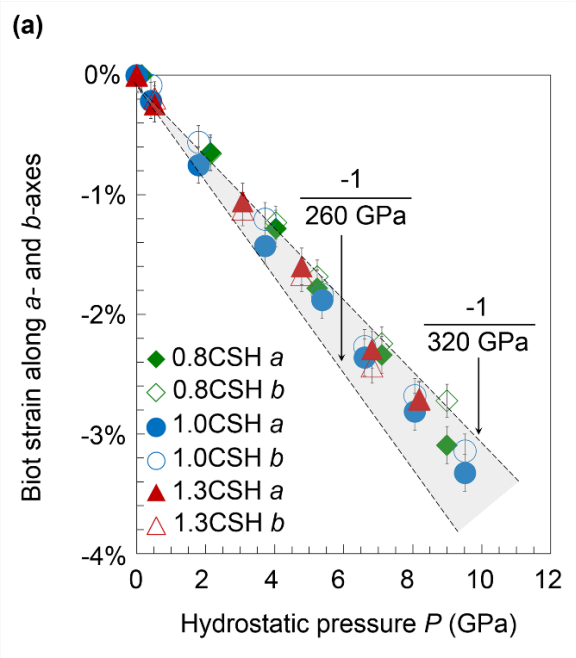


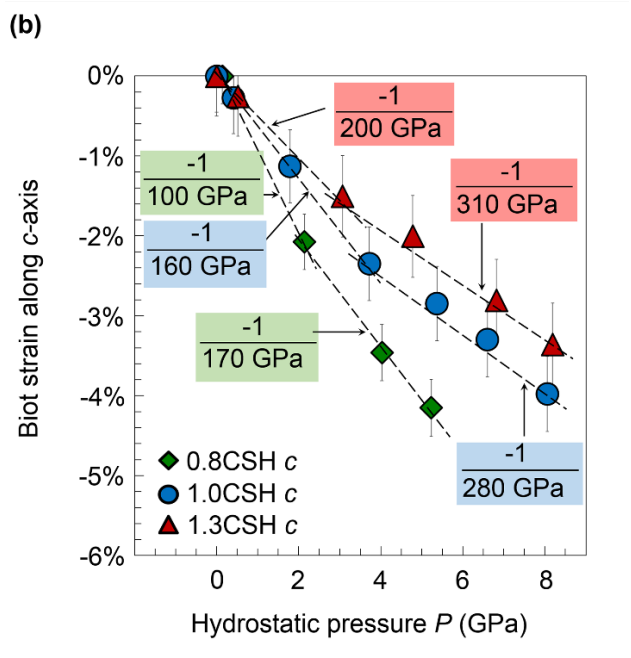
Anisotropic deformation of C-S-H:

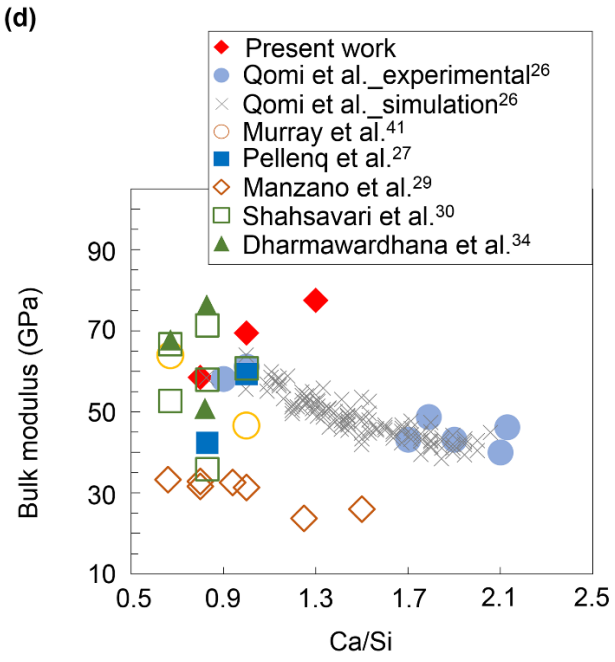
Al/Si=0, Ca/Si=0.8, 1.0, 1.3, 20°C



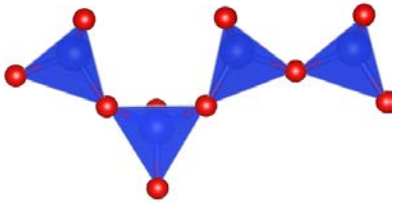


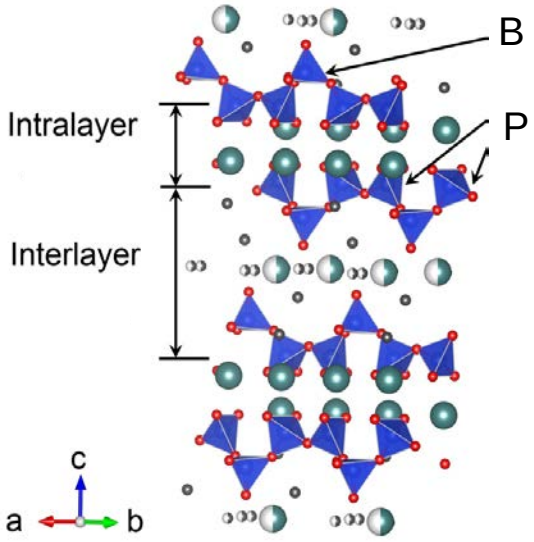


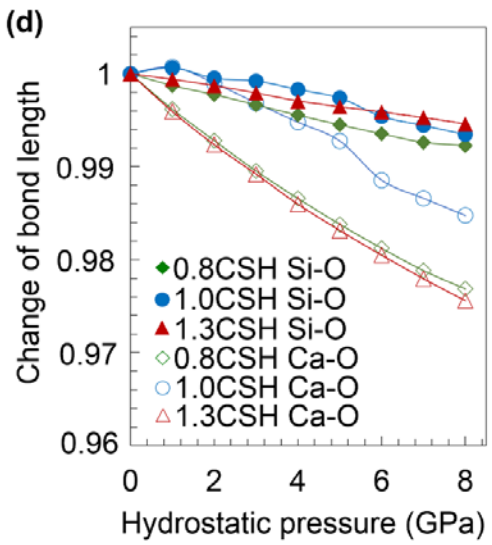


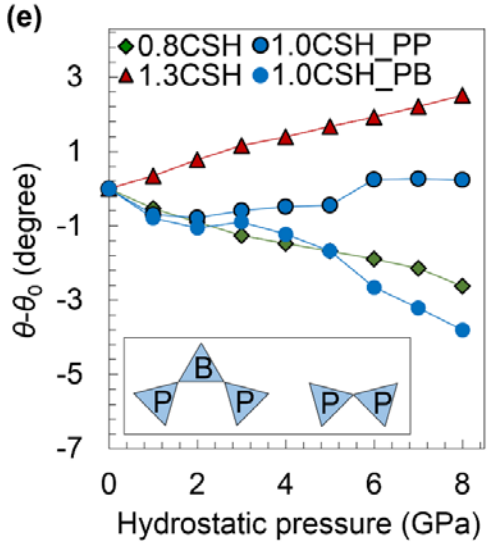


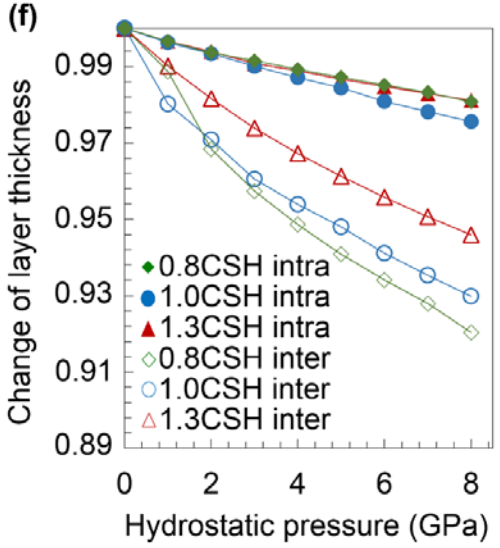
Molecular modeling of C-S-H



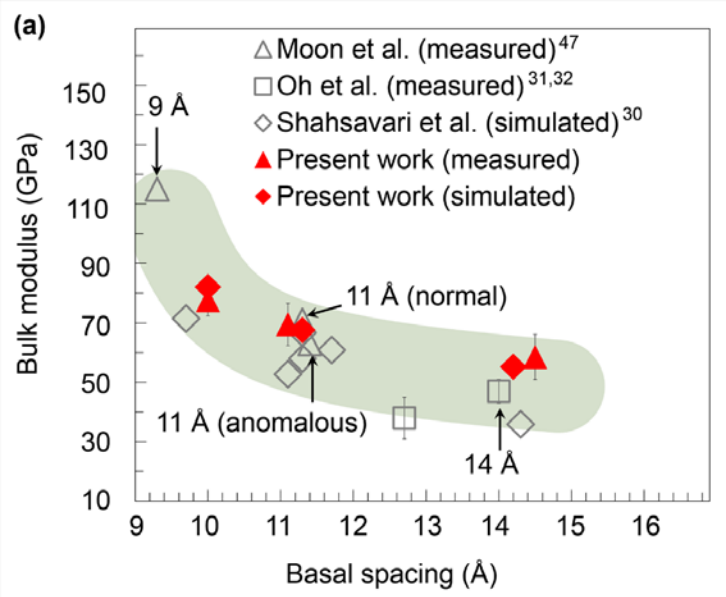


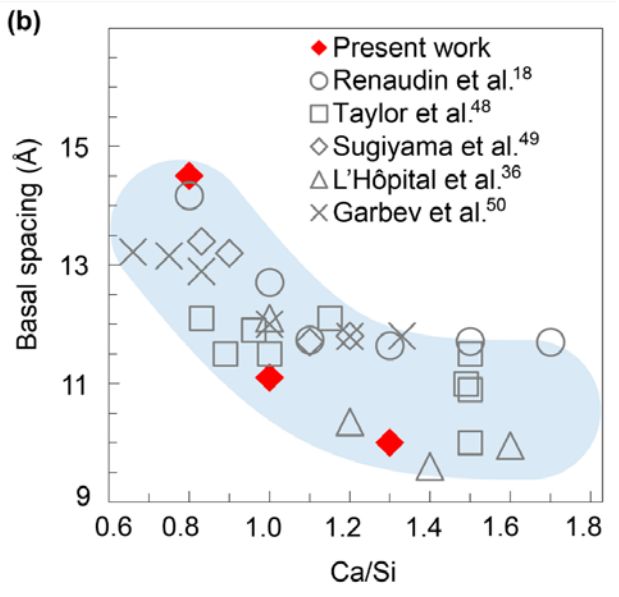


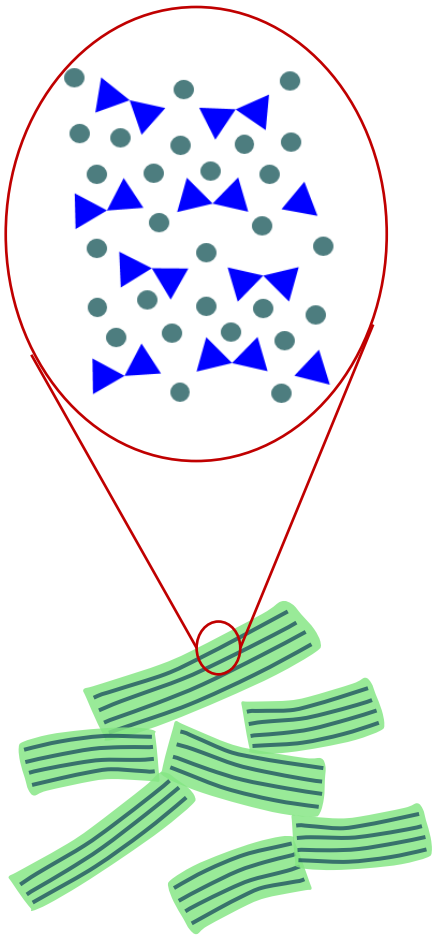




Densification of the interlayer controls the bulk modulus







Quick summary

- Molecular scale “mechanical testing” is possible via HP-XRD.
- There is no direction link between chemistry and elastic constant; Structure plays significant role.
- Humidity changes the mechanical property of concrete matrix
- Existing molecular scale modeling tools are reliable.

Acknowledgement

- European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 701647.
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Rainer Daehn
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Ravi A. Patel

B. Lothenbach
Yiru Yan
Zhangli Hu
Zhenguo Shi

Paulo J. Monteiro
Hans R. Wenk
Jiaqi Li