P02: Transport properties of C-S-H

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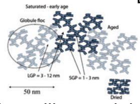
Project description

Moisture transport in cementitious materials is instrumental to understand their durability. Calcium silicate hydrate phases are the reaction product between cement and water.

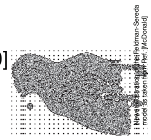
- C-S-H contains the smallest pores being found in cement
- **There is no general agreement on the C-S-H structure**
- C-S-H may govern the moisture transport in cementitious materials if all other pores/cracks depercolate

Literature:

- Two major structural models are currently being discussed:
  - Colloidal models [Jennings2008]



  - Sheet-like models [FeldmanSereda1970]



- The material has a characteristic length scales of a few nano metres. Thus, the transport laws in such pores may deviate significantly from continuum values.
- Due to its amorphous character scattering data is hard to interpret
- There is no information about transport properties in just C-S-H.

Approach

- It is perceived that C-S-H's structure affects many of its properties in an ambiguous way. Thus, the connection of one structure with a given property is hard.
- By generating different model structures and calculting their properties it is hoped to generate a clearer picture than just considering individual properties.

Project results – are perturbed sphere packings a good transport model for C-S-H?

Idea:

Particle models have been used to describe C-S-H. [Powers1958,Jennings2000,Ulm2012] Thus, the simplest imaginable particle model is used for C-S-H: monodisperse particles.

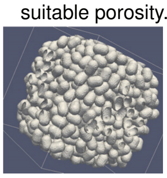
- Scattering results indicate a characteristic size of 4-5 nm for C-S-H particles [Skinner2010, Popova2002].
- NMR-results (McDonald) suggest porosities ε(Gel) of 31 % and ε(Sheet) of 24.5 %.

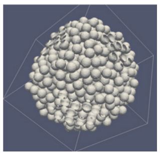
Hypothesis:

- Monodisperse particles suffice
- Continuum theory suffices.
- Gel pores are described as interparticle space
- Sheet pores are assumed to be contained within particles [Jennings2008] and are not modelled.

Approach

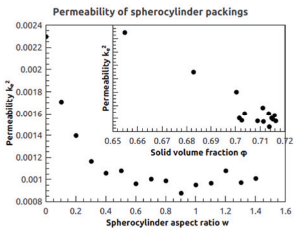
- Generate suitable packings with a suitable porosity.



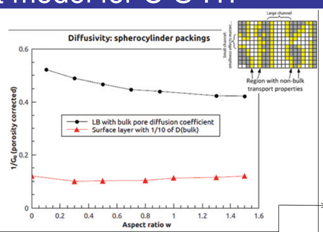


- Characterise their properties:
  - LB-Permeability: $\boldsymbol{v_0} = -\frac{k}{\mu}(\nabla p - \boldsymbol{g})$
  - LB-Diffusivity: $\frac{D}{D_0} = \frac{\varepsilon\delta}{\tau^2} = \frac{\epsilon}{G}$
  - Compute the scattering pattern to compare against data available for C-S-H [Debye].

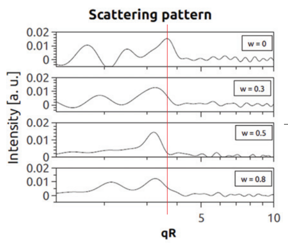
$$I(q) \propto \int_0^\infty \chi(r)\, r^2 \frac{\sin(qr)}{qr}\, dr$$



- Permeability for ε(Gel) = 31 %. Minimum permeability for this model: k = 4.5×10^{-21} m²



- Diffusion:
  D(bulk) = 3×10^{-10} m²/s
  D(surf. L.) = 7.4×10^{-11} m²/s



(a) The permeability is likely to be „an upper limit“ for a spherocylinder-based C-S-H model, because irregular fluid behaviour due to the small pore sizes may reduce the permeability further [Karniadakis2005]. (b) The same argument applies for diffusion. However, the introduction of immobile surface layers might be able to compensate for it [Churakov2011]

Transport data for "pure" C-S-H ***does not exist.*** However, this data may be approximated by data obtained for very mature and crack-free paste with *de-percolated pore systems.* Thus, the lower limit of the literature data may be a base for comparison.

a) Permeability: the lowest values in the literature go down to 10^{-23}m²/s. [Cui2001]

b) Diffusivity: the lowest values being found in the literature are 9.81×10^{-12} – 6.3×10^{-13}m²/s [Pigeon1998]

c) The scattering curve obtained shows distinct peaks, in contrast to typical SANS results [Allen2007]

Conclusion

Monodisperse packings of spherical particles with continuum transport laws *do not match the transport properties of C-S-H as estimated from experimental data.*

The next step is to try different base units, e. g. (polydisperse) sheets (see below).

In principle, this approach can be applied to any perceivable structure.

Future plans

Investigation of other "simple model structures"

The idea is to use a random sequential addition (RSA) sequence to fill space with various geometric forms:

1. Determine insertion position and orientation.
2. Check whether insertion is possible w/o overlap

If no further insertion is possible, shrink inserted structure by some factor and return to 1. Repeat until space is filled to the desired level. This could be done with:

- Discs and similar platelets
- Spheres

Compute the resulting properties

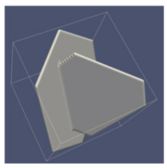
Study of 3D sheet growth structures

Synthetic C-S-H looks like clusters of sheets formed by homogenous nucleation. For C-S-H in cement paste, however, a heterogeneous nucleation growth mechanism is conceivable.

It is planed to investigate the formation and the properties of structures formed by interacting sheets.

- Straight sheets
- Crumpled sheets
- Sheets with "hard" interactions as well as attractive interactions

Key problem: how to represent a growing sheet in space?



Trials have been conducted for an approach based on a 3D cellular automaton with 2D oriented update sequence.

However, bent and randomly oriented sheets have not been realised

A **finite element** based vectorial description (triangles in space) has been considered. Different approaches are being currently evaluated.

Outstanding questions

- ***<u>Experimental data of the transport properties in C-S-H?</u>*** -> direct measurements on C-S-H would allow better conclusions.
- How to generate Feldman-Sereda type sheet structures?
- Modelling of mechanical properties of the structures?
- Obtaining a structure morel consistent with all observations.

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