3D imaging of moisture distribution and transport in early-age cementitious materials

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Concrete & Construction Chemistry

hydration thermodynamics sustainability

3D-microstructure

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Introduction - Empa

- Empa, Swiss Federal Laboratories for Materials Science and Technology is an interdisciplinary research institute for material sciences and technology development within the ETH domain in Switzerland.

- About 30 research laboratories
- ~1000 employees
- Concrete / Construction Chemistry Laboratory, about 25 people

- P. Lura: Head CCC Lab (since 2008) Professor ETH Zurich EiC of Materials & Structures (RILEM)

NEST at Empa
http://nest.empa.ch/en/
Introduction – Concrete / Construction Chemistry Lab at Empa

- **Fundamental research and application-oriented research (R&D) on cement-based materials:**
  - Cement hydration, blended and alternative cements, admixtures
  - Durability (AAR, carbonation, sulfate attack, chlorides, permeability and porosity)
  - Early age concrete (plastic, autogenous and drying shrinkage, microstructure)
  - 3D microscopy and modelling
  - Support of the qualified lab staff and infrastructure in research and service

http://empa.ch/
Outline

- Introduction to internal curing

- Neutron tomography and modelling of internal curing with superabsorbent polymers

- Combined neutron and X-ray tomography to study internal curing with lightweight aggregates

- Even faster: neutron tomography of fresh mortars while drying (plastic shrinkage)

- Multi-contrast X-ray tomography: an alternative to neutrons?
High Performance Concrete for High-Performance Applications

Low w/c \rightarrow Admixtures

\rightarrow Dense concrete

\rightarrow Good durability \leftrightarrow High strength

Burj Khalifa, 828 m, Dubai, UAR, 2010
Autogenous shrinkage in HPC

**Autogenous strain:** bulk strain of a closed, isothermal, cementitious material system not subjected to external forces

*Jensen and Hansen CCR 2001*

- Less water (*low w/c*)
- More cement
  * (*more cement paste*)
- Silica fume
  * (*high chemical shrinkage, fine pores*)
- Dense aggregate
  * (*with low water absorption*)

Great Belt Link, Denmark, 1998
Span: 1624 m
Menisci, RH and pressure

Curing keeps RH high and \( p_{\text{cap}} \) low, hydration goes further
Water curing of concrete surface

Labor intensive, expensive, sometimes “forgotten” or delayed
Tight microstructure of HPC (depercolation of capillary pores) limits water penetration from surface
Concept of internal curing

w/c 0.30 + 0.05

Saturated lightweight aggregate

Pumice

Expanded shale

Lura et al. 2003-2004, Jensen and Lura MS 2006

Jensen and Hansen CCR 2001
Efficiency of internal curing

- **Sufficient amount of internal curing water**
  - Compensate for chemical shrinkage
    *Bentz & Snyder CCR 1999*
    *ACI 2013: “…hydration of cement continues because of the availability of internal water that is not part of the mixing water.”*

- **Availability of water**
  - *Thermodynamic availability*
    - Controlled by the curing reservoirs
    - Water activity $\approx 1$ (equilibrium RH~100%)
    - For LWA – water in large pores
  - *Kinetic availability*
    - Controlled by the microstructure of cement paste and the curing reservoirs
    - Fast and uniform distribution of water from reservoirs to the cement paste

*Trtik, Lura et al. RILEM 2010*
Water transport from SAP

Optimization of chemistry and particle size distribution of SAP:

- How much and how fast do SAP absorb?
- When do SAP release the water?
- How far does water reach in the hardening cement paste?
Why neutron tomography?

- Inconclusive / unclear results from X-ray radiography and tomography

- Cold-neutrons (e.g. ICON at Paul Scherrer Institute, CH) have high sensitivity for water and good spatial resolution (25-50 μm voxel size)

- Sample of cement paste with large SAP, max. transport distance ~3 mm, reinforced Teflon holder

~1 mm

Source: NIST 2003
Water release from SAP and transport to paste

![Graph showing water release from SAP and chemical shrinkage over time.]

- Water released by SAP
- Chemical shrinkage

Time (hours)

Volume (mm^3)

Portland cement paste w/c 0.25, curing 28°C

Trtik, Lura et al. RILEM 2010

Neutron tomography @ SINQ (PSI), cold neutrons ICON beamline

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Water transport during IC: meso-level

Poromechanics

Accumulation terms

\[ n \left( \rho^w - \rho^w \right) \frac{\partial S_w}{\partial t} + \frac{\partial p^e}{\partial t} - \left( \beta_s \rho^w (1 - n) (1 - S_w) + \left[ (1 - n) \beta_s + n \beta_w \right] \rho^w S_w \right) \frac{\partial T}{\partial t} + (1 - S_w) n \left( \frac{\partial \rho^w}{\partial T} \frac{\partial T}{\partial t} \right) + \]

\[ + \left[ \rho^w (1 - S_w) + \rho^w S_w \right] \frac{\partial \mathbf{u}}{\partial t} - \nabla \cdot \left[ \rho^w \frac{M_g M_w}{M_w} D^g \nabla \left( \frac{p^g}{p^w} \right) \right] + \nabla \left[ \rho^w \frac{\mathbf{K}^{g \varepsilon}}{\mu^g} \left( - \nabla p^g + \nabla p^c \right) \right] + \]

\[ + \nabla \left[ \rho^w \frac{k^{g \varepsilon}}{\mu^w} \left( - \nabla p^g + \nabla p^c \right) \right] \]

\[ = \frac{\rho^w}{\rho^c} \left( 1 - S_w \right) \dot{m}_{\text{hydr}} + \frac{\rho^w}{\rho^c} S_w \dot{m}_{\text{hydr}} - \dot{m}_{\text{hydr}} \]

Flux terms

Source terms

\[ \text{SAP 2.4 mm (0.094 in.), w/c_e 0.050, max dist. 2.2 mm (0.087 in.)} \]

\[ \text{SAP 0.8 mm (0.031 in.), w/c_e 0.050, max dist. 0.74 mm (0.029 in.)} \]

Wyrzykowski, Lura, Pesavento, Gavin, JMCE 2012
Modelling IC: macro-level

Water conservation equation with additional mass source term

\[
 n \left( \rho^w - \rho^{gw} \right) \frac{\partial S_w}{\partial p^c} \frac{\partial p^c}{\partial t} = \frac{\rho^{gw}}{\rho^s} \left( 1 - S_w \right) \dot{m}_{\text{hydr}} + \frac{\rho^w}{\rho^s} S_w \dot{m}_{\text{hydr}} - \dot{m}_{\text{hydr}} + \dot{m}_{\text{IC}}
\]

Sorption isotherm for SAP (LWA)

\[
 \dot{m}_{\text{IC}} \left( p^c \right) = \frac{\eta}{1 - \eta} \rho^w \frac{\partial S_{\text{IC}}}{\partial p^c} \frac{\partial p^c}{\partial t}
\]

Capillary suction

Mortars with SAP w/c 0.3 + 0.04

Possible application to LWA

Wyrzykowski, Lura, Pesavento, Gawin, CCR 2011
Bio-LWA for internal curing

LWA from biomass-derived waste (sugar cane bagasse fly ash)

- Pelletization and sintering (1100°C), crushing
- Density 1.7 g/cm$^3$
- Porosity 30-40%
- Water absorption 5-15%

Lura, Wyrzykowski, Tang, Lehmann CCR 59, 2014
Neutron tomography of mortars with LWA

- w/c 0.3 cement paste with LWA aggregates
- Tomographies run at 1.2h, 9.7h and 15h from mixing (scan lasted about 1.25 h)
- Setting around 6-7h
- Voxel size for NT 27 µm

Neutron tomography @ SINQ (PSI), cold neutrons ICON beamline

Lura, Wyrzykowski, Tang, Lehmann CCR 59, 2014
Neutron and X-ray microtomography (segmenting of LWA)

- Slices from the reconstructed 3-D data

Lura, Wyrzykowski, Tang, Lehmann CCR 59, 2014
Neutron tomography of mortars with LWA (4)

- Subtraction images to find the changes in water content

1.2 h

LWA#30

15.0 h

LWA#30

Lura, Wyrzykowski, Tang, Lehmann CCR 59, 2014
Internal curing with expanded shale LWA (Liapor)

- Combination of NT and X-ray $\mu$CT to detect LWA boundaries
- Upper LWA dry, absorbs water from paste, then releases
- Lower LWA saturated, only release


Measurements at ICON
Plastic shrinkage settlement and cracking

Cracks go through concrete slab, water seeps

Photos by A. Leemann

Evaporation of bleed water

Constant rate period

Falling rate period

Lura, Weiss et al. ACIMJ 2007
Water release from SAP in fresh pastes

Cement paste w/c 0.3
Temperature: 24 ± 0.5°C
Relative humidity: 25 ± 3 % RH

Wyrzykowski, Lura et al. 2012, unpublished
Combined neutron and X-ray tomography of mortar during early-age drying (plastic shrinkage phase)

Mortar samples (w/c 0.50, 45% vol. aggregates 0.25-1mm)
PTFE containers, φ 18 mm, height 19 mm (internal), wall thickness 1 mm
Wind speed ~1 m/s, Temperature ~20°C, RH ~40%
Measurements at ICON, PSI: voxel ~50 μm, acquisition time ~33 s

Ghouchian, Griffa, Wyrzykowski, Münch, Lura et al., unpublished 2016
Reconstruction from golden ratio sequence

- Observe the sample at oblique angles
- The golden ratio $\phi$ gives the next angle $\theta$

\[ \phi = \frac{1 + \sqrt{5}}{2} \]

\[ \theta_i = \text{mod}(i \cdot \phi \cdot \pi, \pi) \]

- Allows to find optimal compromise between spatial and temporal resolution

Kaestner et al. Opt Eng 2011

Reconstructed image (horizontal slice) with different number of projections

Different reconstruction methods:
- Filtered back-projection Kaestner 2011
- Penalized likelihood Ahn et al. 2006
- Spatio-temporal regularization Kazantsev et al. 2013
- SIRT (Simultaneous Iterative Reconstruction Technique)
Animation (vertical mid-section) for different numbers of projections used in reconstruction

- 16 projections / 3-D dataset
  time span – 9 min / 3-D dataset
- 32 projections / 3-D dataset
  time span – 18 min / 3-D dataset
- 64 projections / 3-D dataset
  time span – 36 min / 3-D dataset

Overall time span – 7 h

Reference mortar
Animation (vertical mid-section) for different numbers of projections used in reconstruction

16 projections / 3-D dataset
time span – 9 min / 3-D dataset

32 projections / 3-D dataset
time span – 18 min / 3-D dataset

64 projections / 3-D dataset
time span – 36 min / 3-D dataset

Overall time span – 6 h

SAP mortar
X-ray – in processing

Projections

Time 0 h

Time 6 h
Drying of fresh concrete (plastic shrinkage)

Neutron tomography study of water transport – SRA, LWA

Measurements at NEUTRA

Water loss [kg/m²] vs. Time [h]

- Initial evaporation before the scans
- Deionized water

Wyrzykowski, Trtik, Münch, Weiss, Vontobel, Lura
CCR 2015
X-ray phase-contrast imaging

Talbot-(Lau) interferometer

source grating, G0

ideal for laboratory-scale setup

sketch of X-ray grating interferometer
adapted from Zanette 2011
Dark-field contrast X-ray imaging of water capillary uptake in mortars

Talbot-Lau interferometer with conventional macro-focused tube

Setup at TU Munich, Germany

Samples of w/c 0.5 mortar 10x20x2mm³
All dried in an oven at 50°C for 48 h
Left sample pre-conditioned at 200°C for 1 h
Middle sample pre-conditioned at 120°C for 3 h

wetting front profiles extracted from dark-field radiographs

Yang, Prade, Griffa, Jerjen, Di Bella, Herzen, Sarapata, Pfeiffer, Lura Appl. Phys. Lett. 2014
Dynamic dark-field contrast tomography of internal curing in cement paste

Grating-based X-ray microtomography @ Empa

Voxel size 58.6 μm
Temporal resolution=2.71h/tomography

W/c=0.3 cement paste
Curing agent: pre-saturated LWA

Yang, Griffa, Lura et al. 2016, unpublished
Conclusions

- Neutron tomography and modelling of internal curing with SAP and LWA:
  - curing in sealed systems is released after setting
  - release follows closely chemical shrinkage of paste
  - no saturation gradients up to a few mm from internal curing agents

  ⇒ Simplification of modelling

- Combining neutron and X-ray tomography (different contrast):
  - useful to segment internal features, e.g. LWA boundaries
  - first attempts with simultaneous measurements

- Neutron / X-ray tomography of plastic mortars while drying:
  - golden ratio acquisition scheme
  - reconstruction from few projections, time/space resolution compromise

- Multi-contrast X-ray tomography
  - dark-field contrast sensitive to emptying of small pores
  - useful for capillary suction (2D), drying and internal curing (3D)
  - how to quantify the moisture loss/gain?
Collaborators / references

- B. Münch, S. Ghourchian, R. Kaufmann – *Empa*, Switzerland
- J. Weiss – *Oregon State University*, USA
- P. Trtik, A. Kaestner, P. Vontobel, E. Lehmann – *Paul Scherrer Institute*, Switzerland
- F. Pesavento – *University of Padua / D. Gawin – Lodz University of Technology*

Industrial partners:

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- Gregor Herth, Stefan Friedrich, Alexander Assmann – *BASF*, Germany
- Guillaume Jeanson – *SNF Floerger*, France

Main published results


