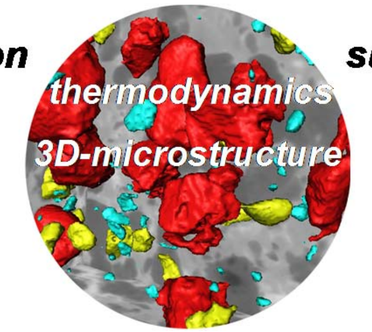
Empa
Materials Science and Technology

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

Pietro Lura, Mateusz Wyrzykowski, Michele Griffa, Fei Yang

Concrete & Construction Chemistry

hydration *sustainability*

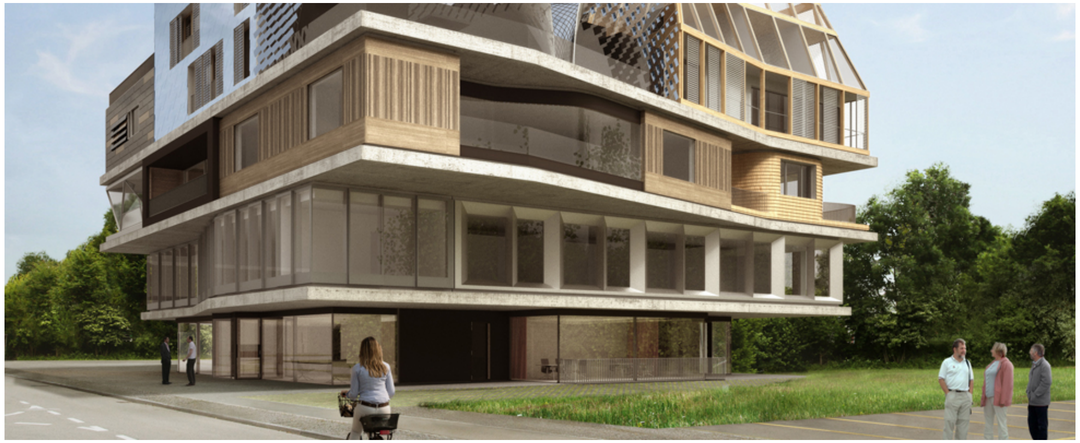


Imaging of Construction Materials and Geomaterials, 7-8 July, 2016, École des Ponts ParisTech, Champs-sur-Marne, France

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

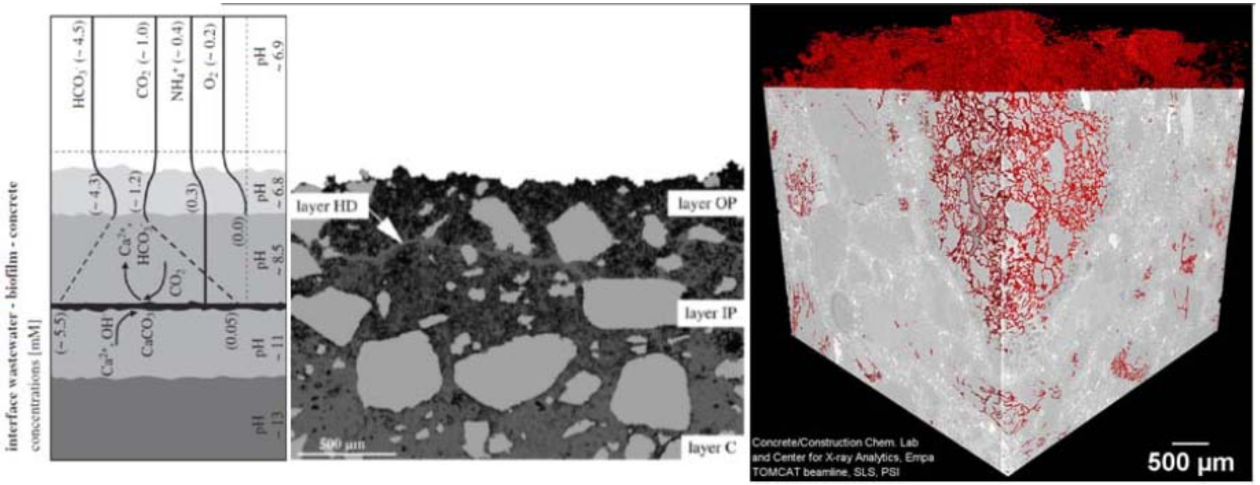


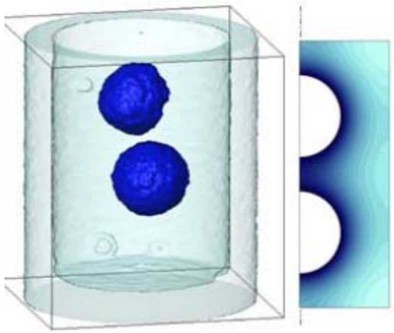
NEST at Empa
http://nest.empa.ch/en/

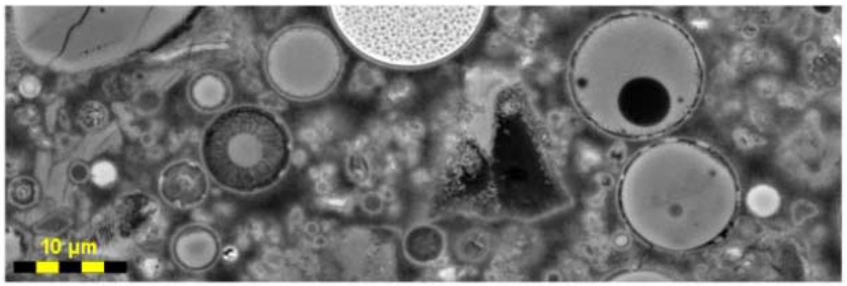
- **P. Lura**: Head CCC Lab (since 2008)
Professor ETH Zurich
EiC of Materials & Structures (RILEM)

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*







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

Admixtures

Dense concrete

Good durability

High strength

Burj Khalifa, 828 m, Dubai, UAR, 2010

Autogenous shrinkage in HPC

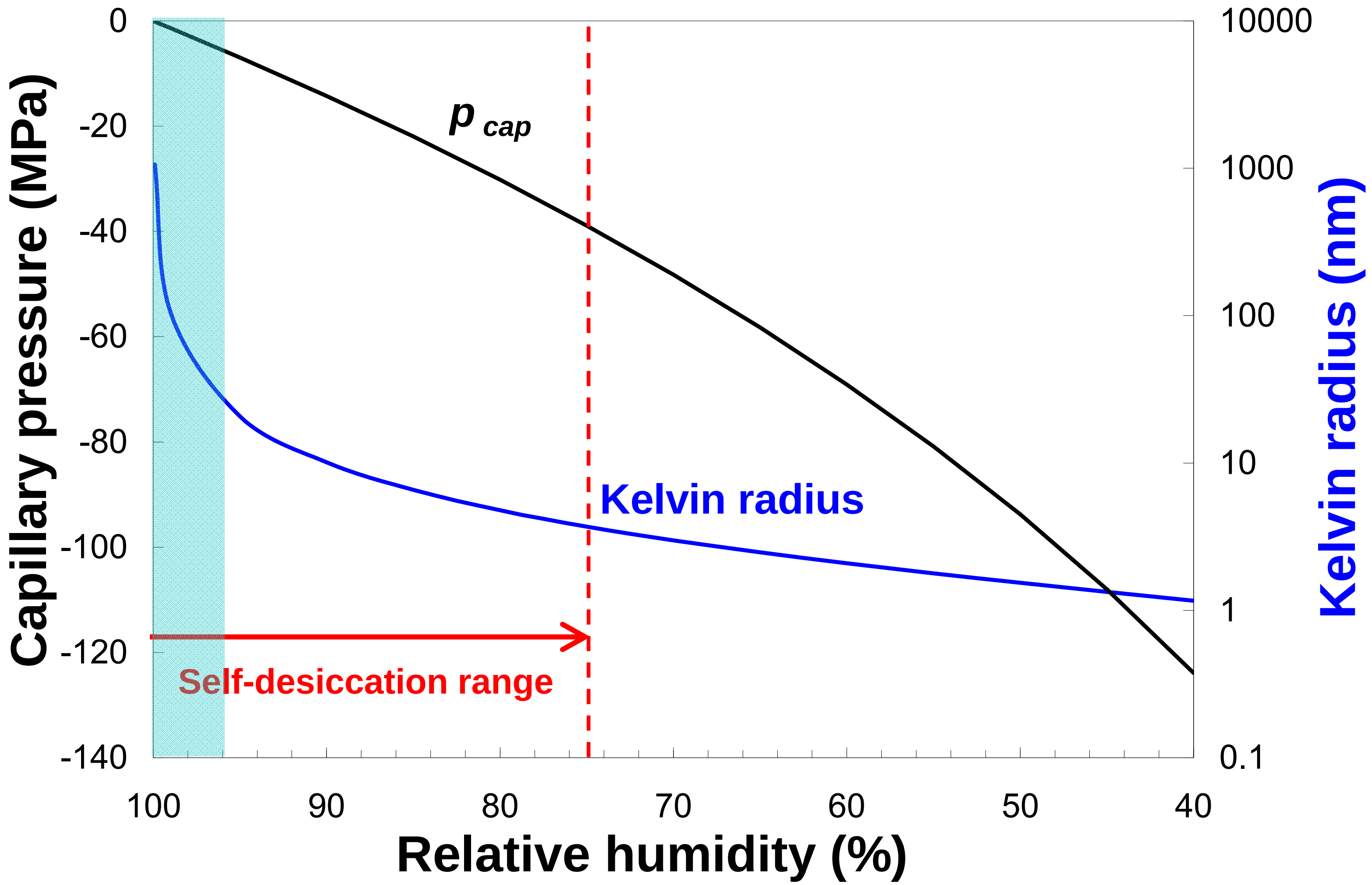
Autogenous strain: bulk strain of a closed, isothermal, cementitious material system not subjected to external forces

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)

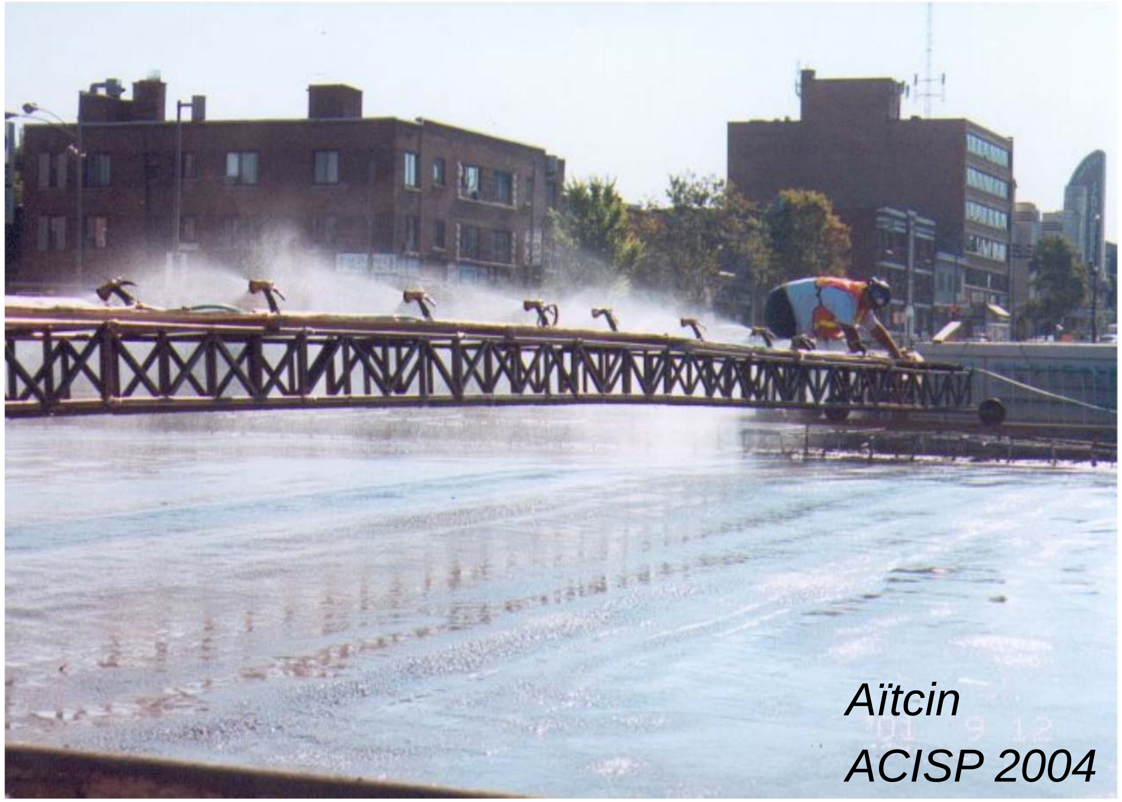
Great Belt Link, Denmark, 1998
Span: 1624 m

Menisci, RH and pressure



Curing keeps RH high and p_{cap} low, hydration goes further

Water curing of concrete surface



Aïtcin
ACISP 2004

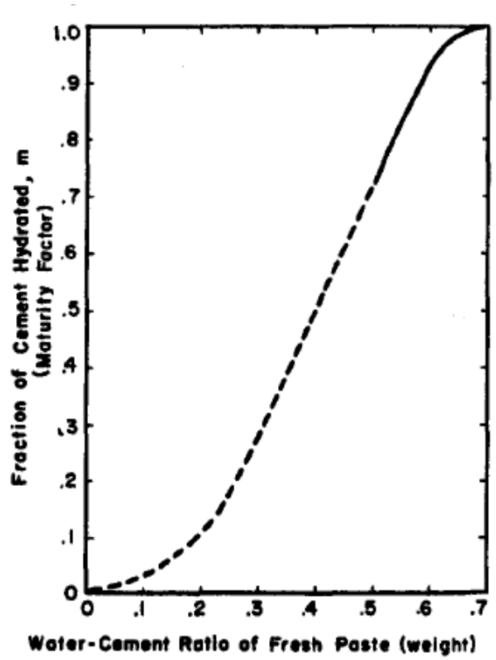


Fig. 3 — Estimated Relationship between Water-Cement Ratio of Fresh Paste and Maturity of Hardened Paste at which Capillary Continuity is Lost.

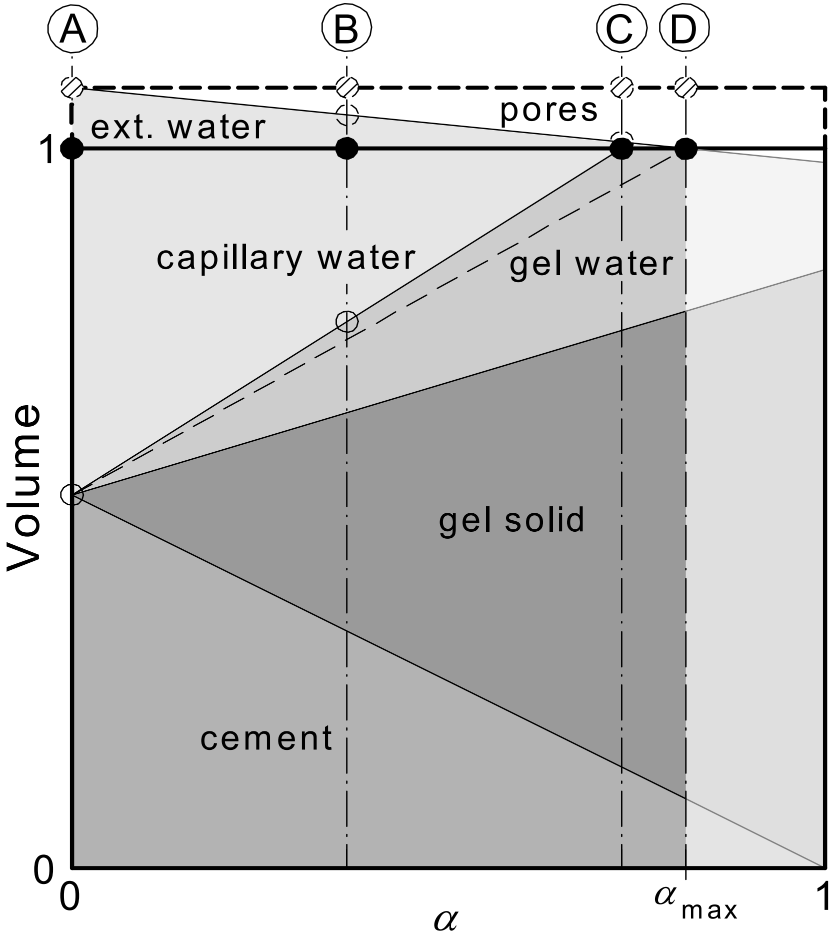
Powers et al. PCA Bull. 1959

Labor intensive, expensive, sometimes “forgotten” or delayed

Tight microstructure of HPC (depercolation of capillary pores) limits water penetration from surface

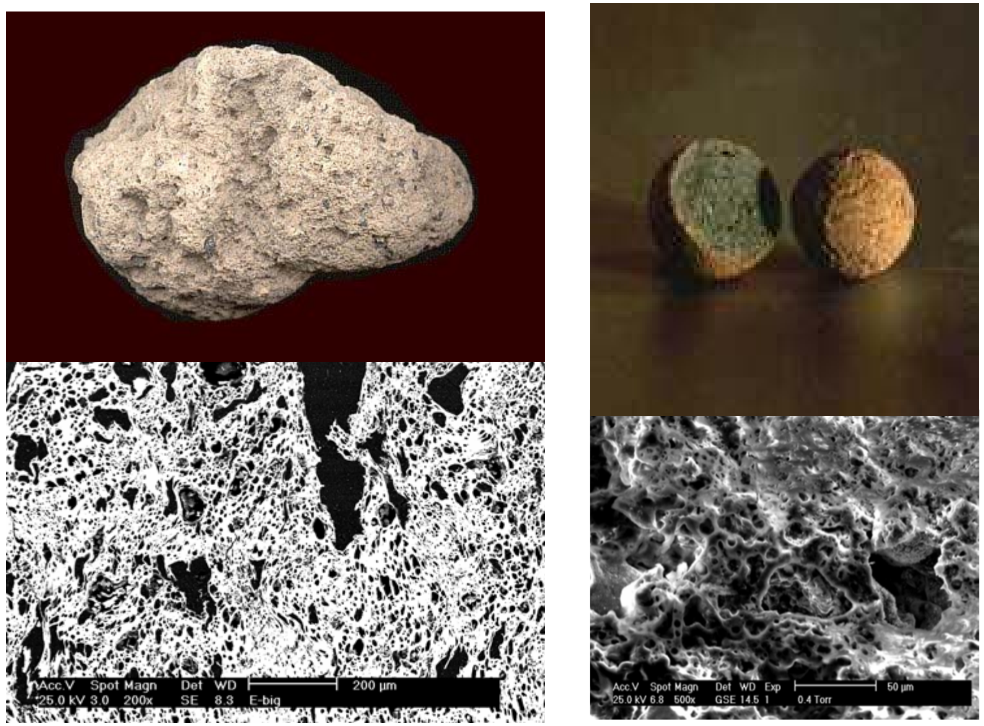
Concept of internal curing

w/c 0.30 + 0.05



Jensen and Hansen CCR 2001

Saturated lightweight aggregate



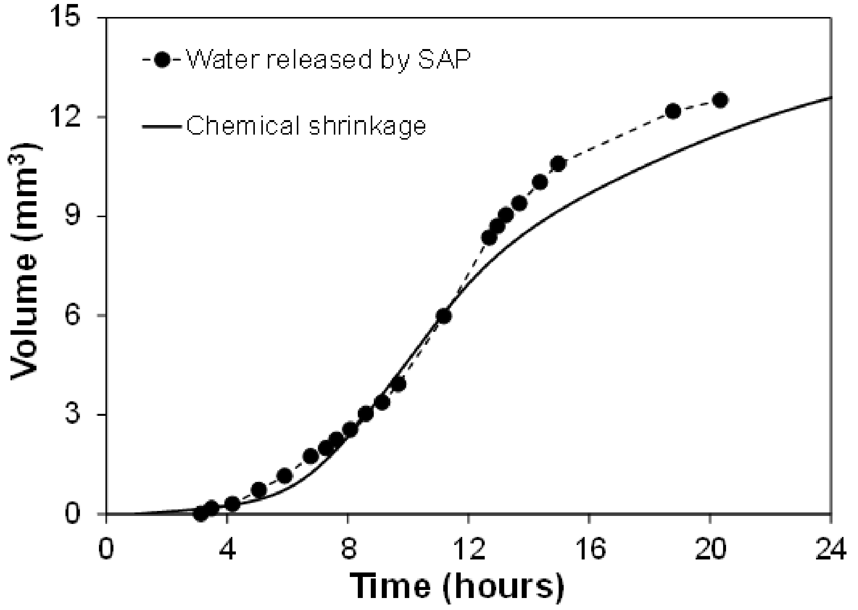
Pumice Expanded shale

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

Efficiency of internal curing

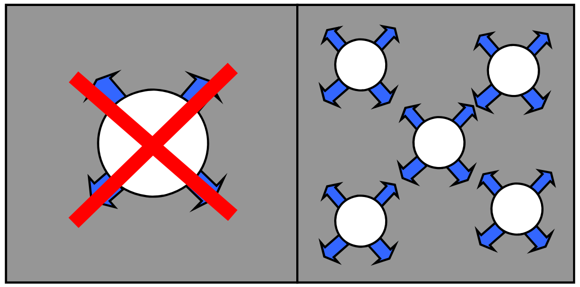
- **Sufficient amount of internal curing water**
  - Compensate for chemical shrinkage
    Bentz & Snyder CCR 1999

  ACI 2013: "…hydration of cement continues because of the availability of internal water that is not part of the mixing water."



Trtik, Lura et al. RILEM 2010

- **Availability of water**
  - *Thermodynamic availability*
    - Controlled by the curing reservoirs
    - Water activity ≈ 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

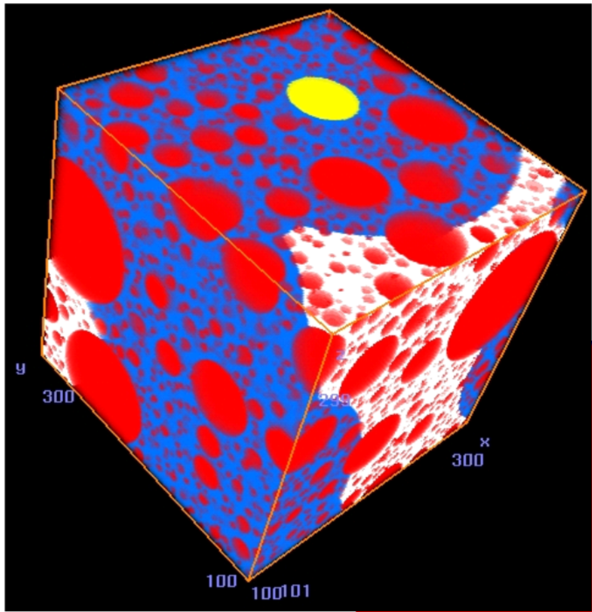


Jensen and Lura MS 2006

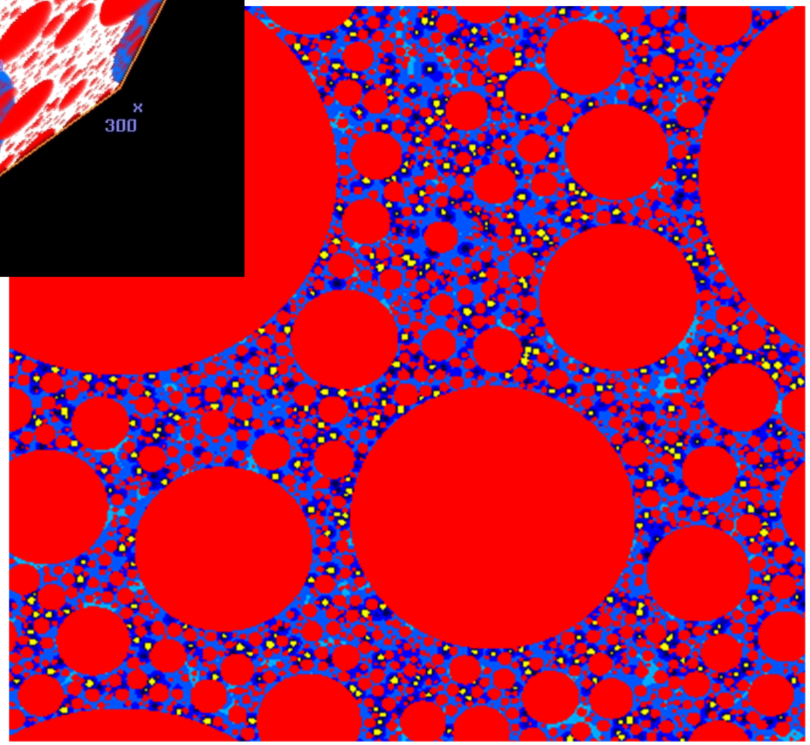
Water transport from SAP

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?



LWA
Bentz, Lura and Roberts CI 2005



SAP *Mönnig and Lura RILEM 2007*

Why neutron tomography?

- Inconclusive / unclear results from X-ray radiography and tomography
  Bentz et al. RILEM 2006, Lura et al. MS 2006, Henkesiefken et al. Strain 2011

- 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

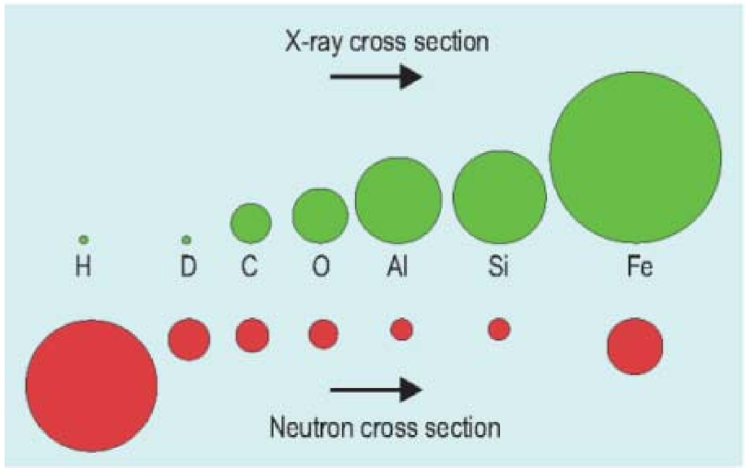
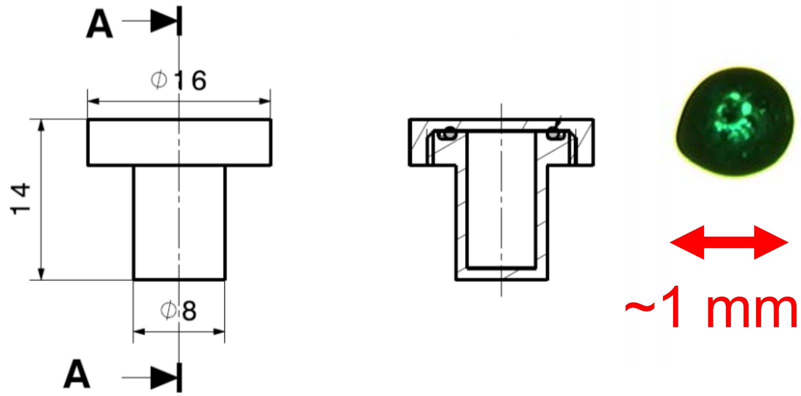
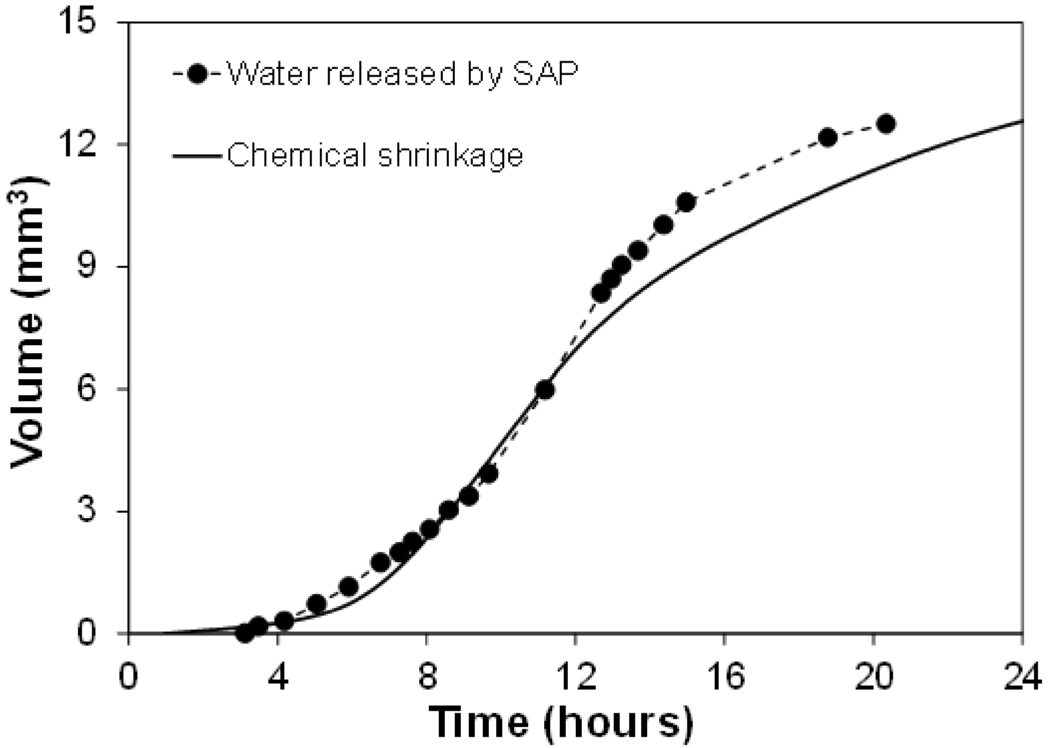


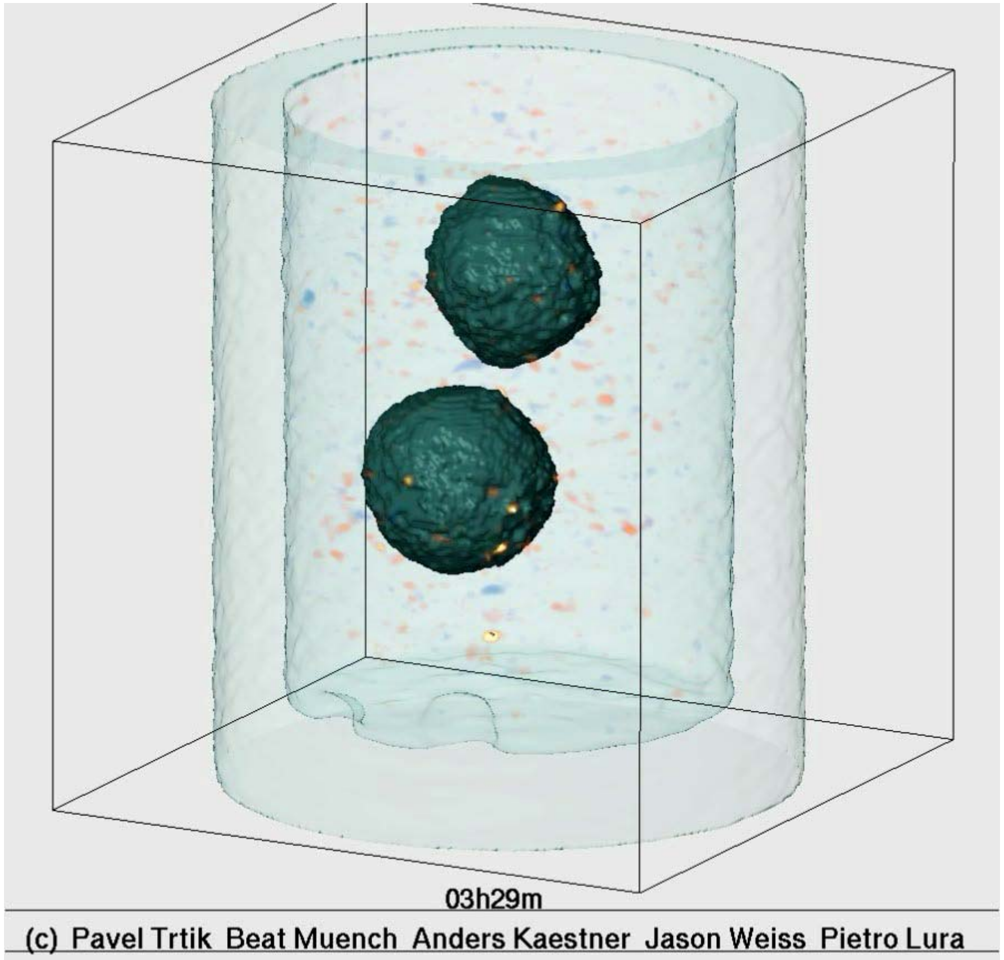
Fig. 2. Neutron and x-ray scattering cross-sections compared. Note that neutrons penetrate through Al much better than x rays do, yet are strongly scattered by hydrogen.

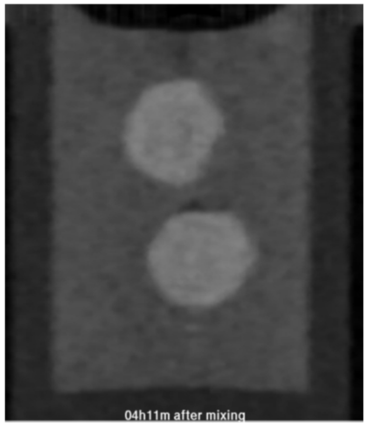
Source: NIST 2003

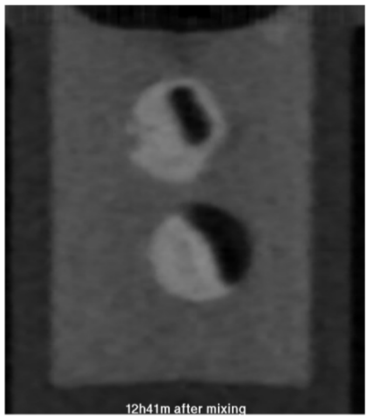


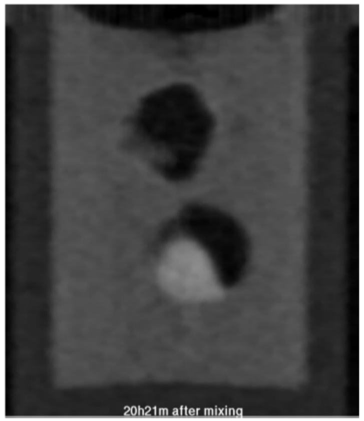
Water release from SAP and transport to paste






4 hours

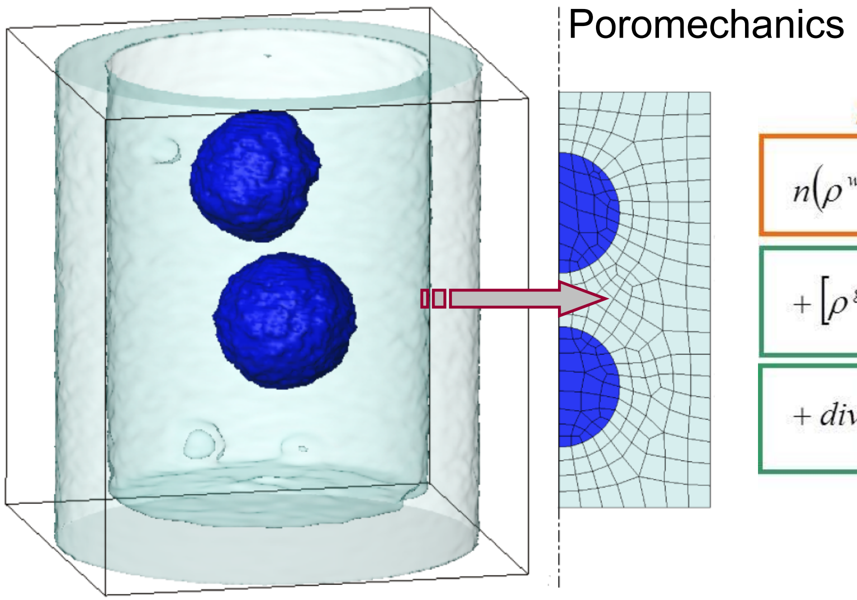

12 hours


20 hours

Trtik, Lura et al. RILEM 2010

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

Water transport during IC: meso-level



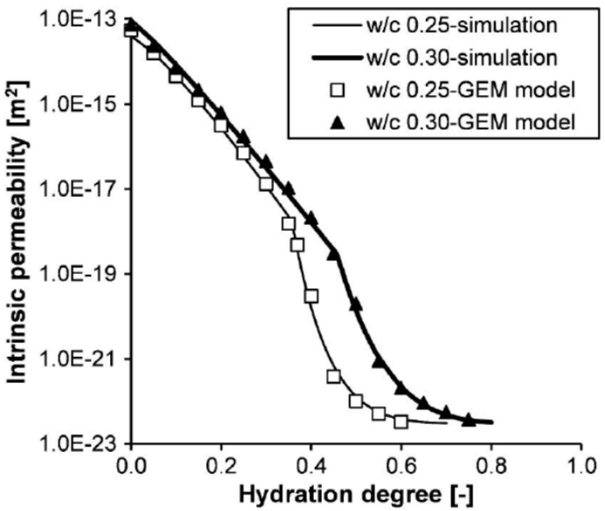
Accumulation terms

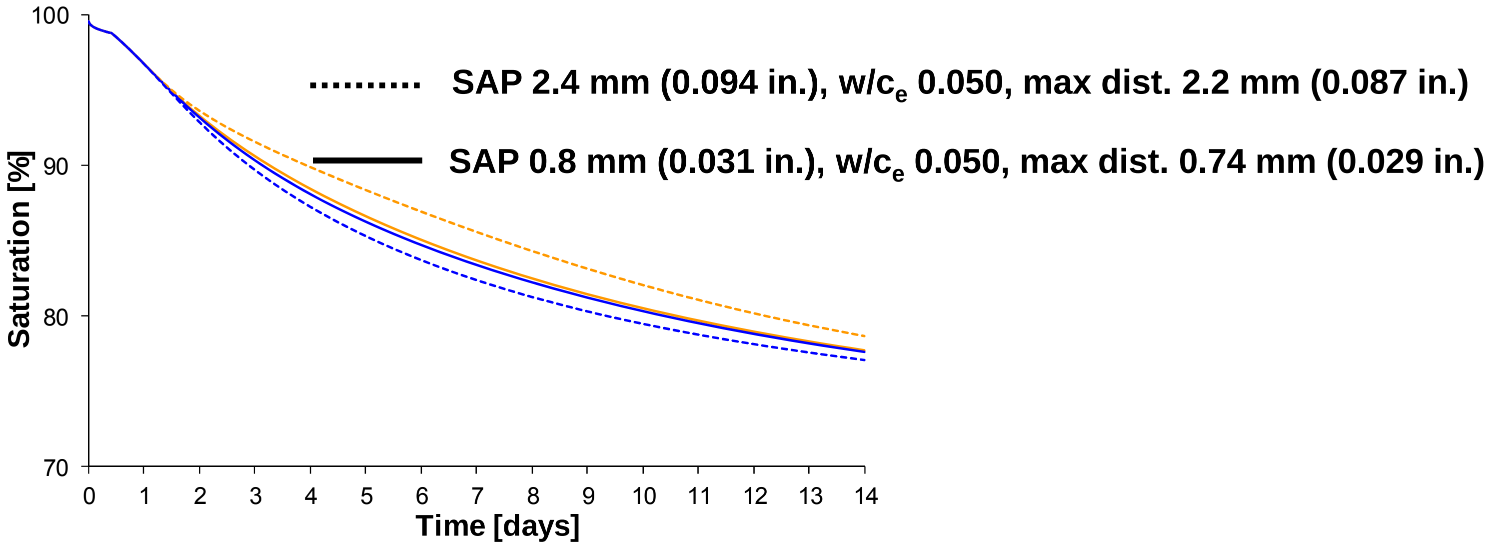
$$n\left(\rho^{w}-\rho^{gw}\right)\frac{\partial S_w}{\partial p^c}\frac{\partial p^c}{\partial t}-\left\{\beta_s\rho^{gw}(1-n)(1-S_w)+\left[(1-n)\beta_s+n\beta_w\right]\rho^{w}S_w\right\}\frac{\partial T}{\partial t}+(1-S_w)n\left(\frac{\partial\rho^{gw}}{\partial T}\frac{\partial T}{\partial t}\right)+$$

$$+\left[\rho^{gw}(1-S_w)+\rho^{w}S_w\right]div\frac{\partial \mathbf{u}}{\partial t}-div\left[\rho^{g}\frac{M_aM_w}{M_g^2}\mathbf{D}_d^{gw}\,grad\left(\frac{p^{gw}}{p^g}\right)\right]+div\left[\rho^{gw}\frac{k\mathbf{I}k^{rg}}{\mu^g}\left(-grad\ p^g\right)\right]+$$

$$+div\left[\rho^{w}\frac{k\mathbf{I}k^{rw}}{\mu^w}\left(-grad\ p^g+grad\ p^c\right)\right]=\frac{\rho^{gw}}{\rho^s}(1-S_w)\dot m_{hydr}+\frac{\rho^{w}}{\rho^s}S_w\dot m_{hydr}-\dot m_{hydr}$$

Flux terms **Source terms**





Wyrzykowski, Lura, Pesavento, Gavin, JMCE 2012

Modelling IC: macro-level

Water conservation equation with additional mass source term

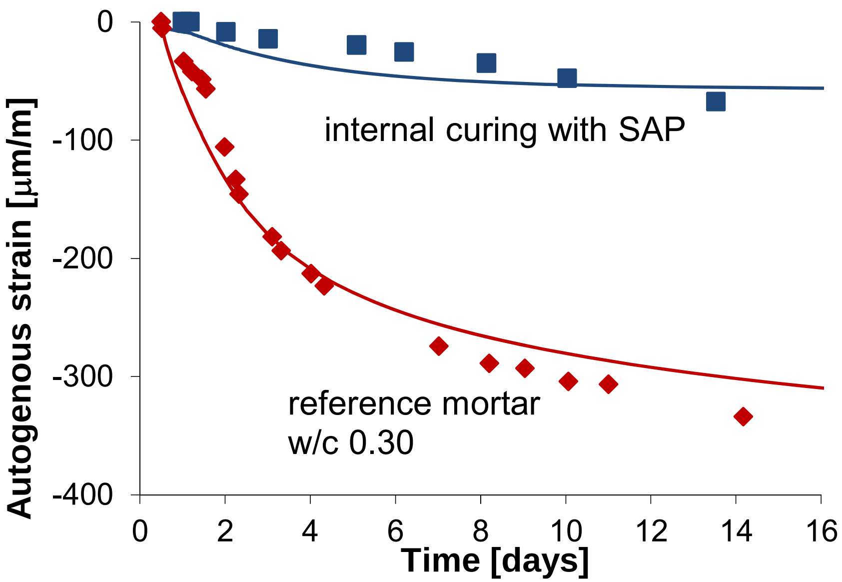
$$n\left(\rho^{w}-\rho^{gw}\right)\frac{\partial S_{w}}{\partial p^{c}}\frac{\partial p^{c}}{\partial t}=\frac{\rho^{gw}}{\rho^{s}}\left(1-S_{w}\right)\dot{m}_{hydr}+\frac{\rho^{w}}{\rho^{s}}S_{w}\dot{m}_{hydr}-\dot{m}_{hydr}+\dot{m}_{IC}$$

Sorption isotherm for SAP (LWA)

Capillary suction

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

Mortars with SAP w/c 0.3 + 0.04 → **Possible application to LWA**



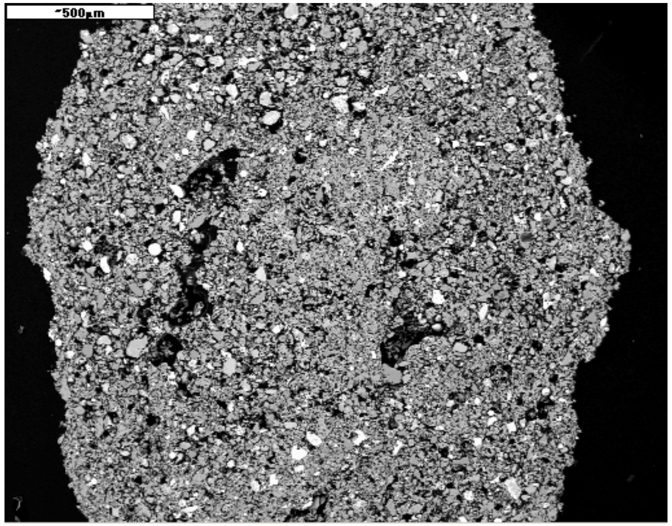
Wyrzykowski, Lura, Pesavento, Gawin, CCR 2011

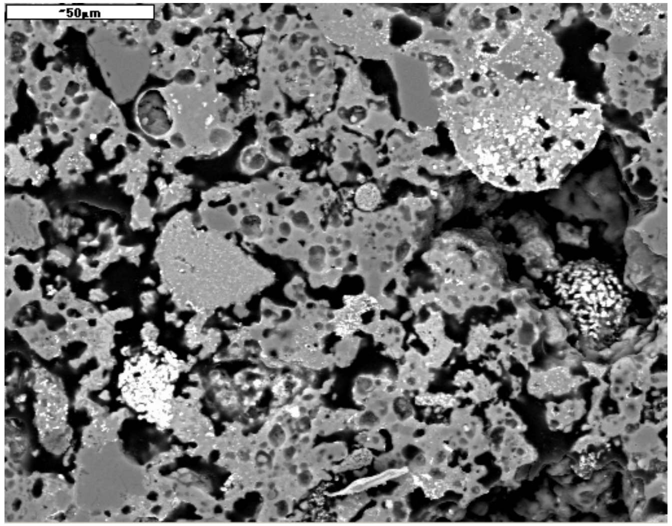
Bio-LWA for internal curing

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%

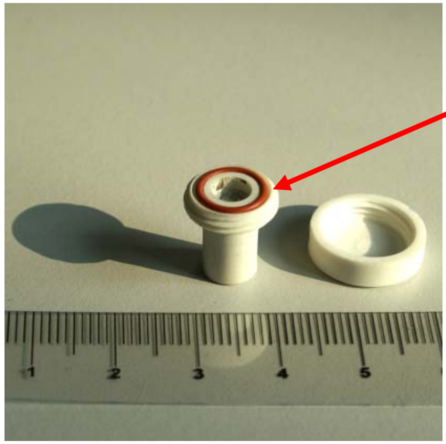
Lura, Wyrzykowski, Tang, Lehmann CCR 59, 2014

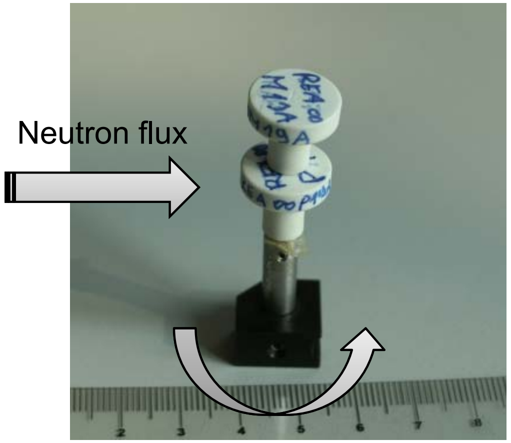


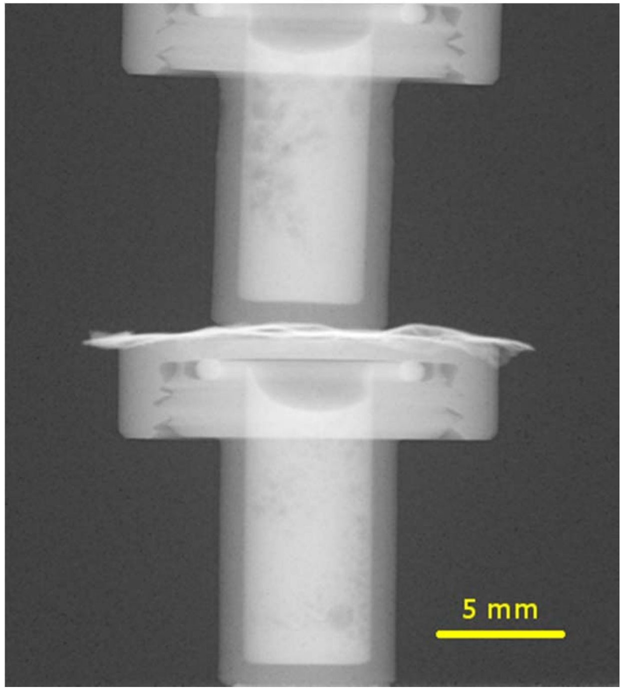


Neutron tomography of mortars with LWA

- 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







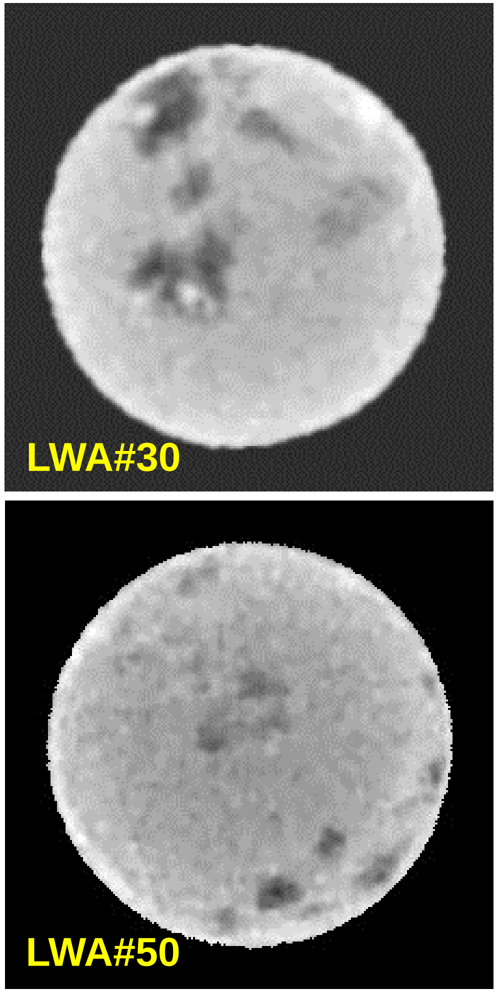
Neutron tomography @ SINQ (PSI), cold neutrons ICON beamline

Lura, Wyrzykowski, Tang, Lehmann CCR 59, 2014

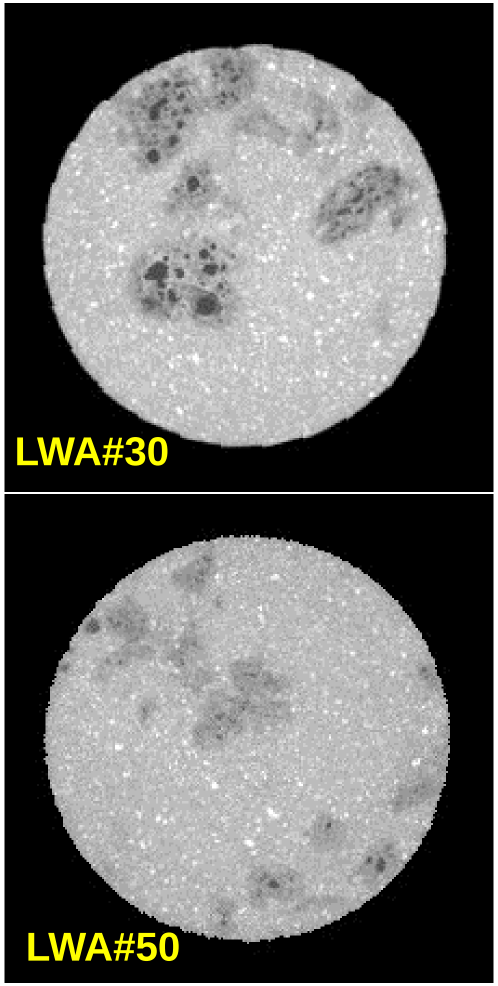
Neutron and X-ray microtomography (segmenting of LWA)

- **Slices from the reconstructed 3-D data**

Neutron tomography



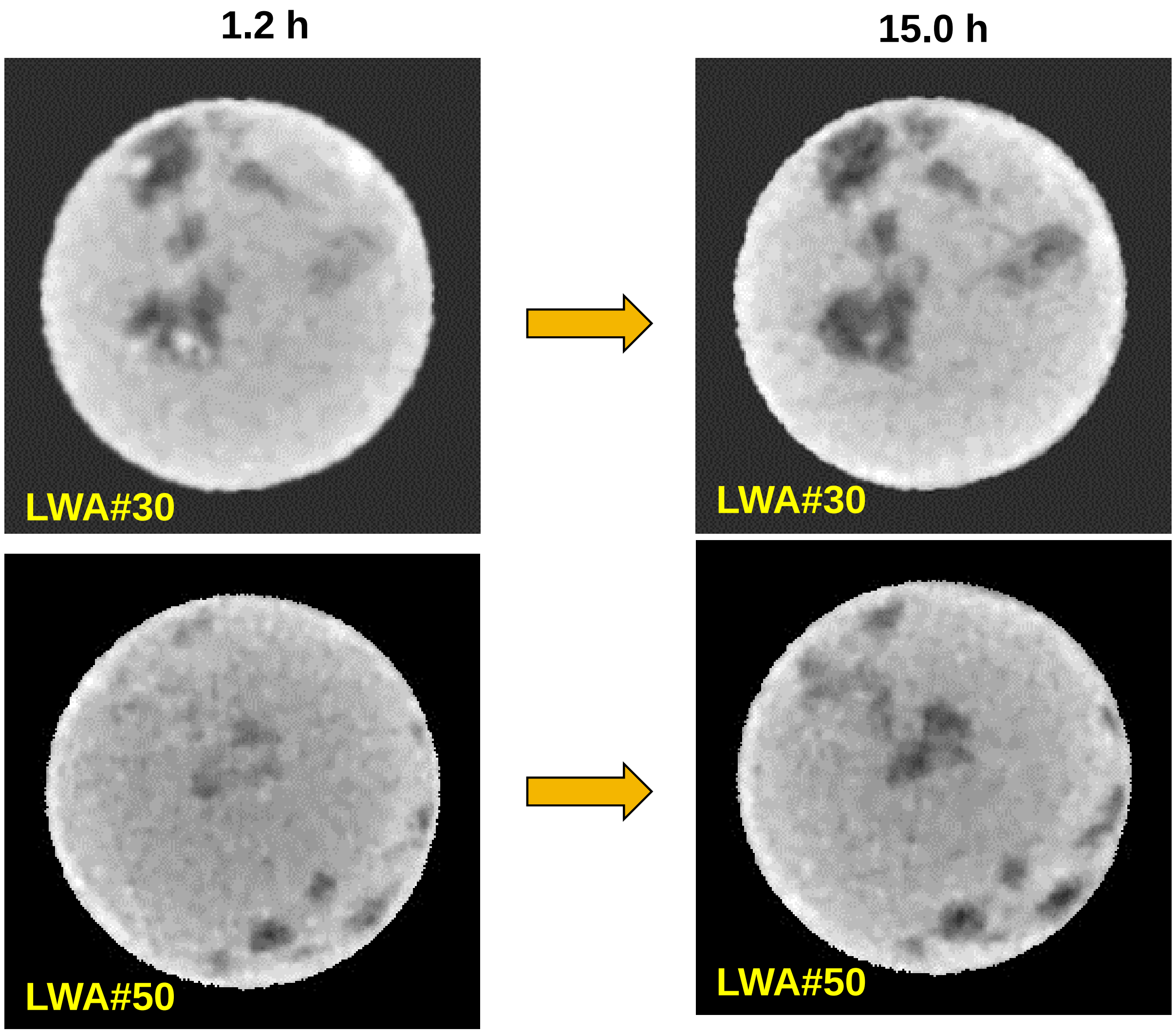
X-ray tomography

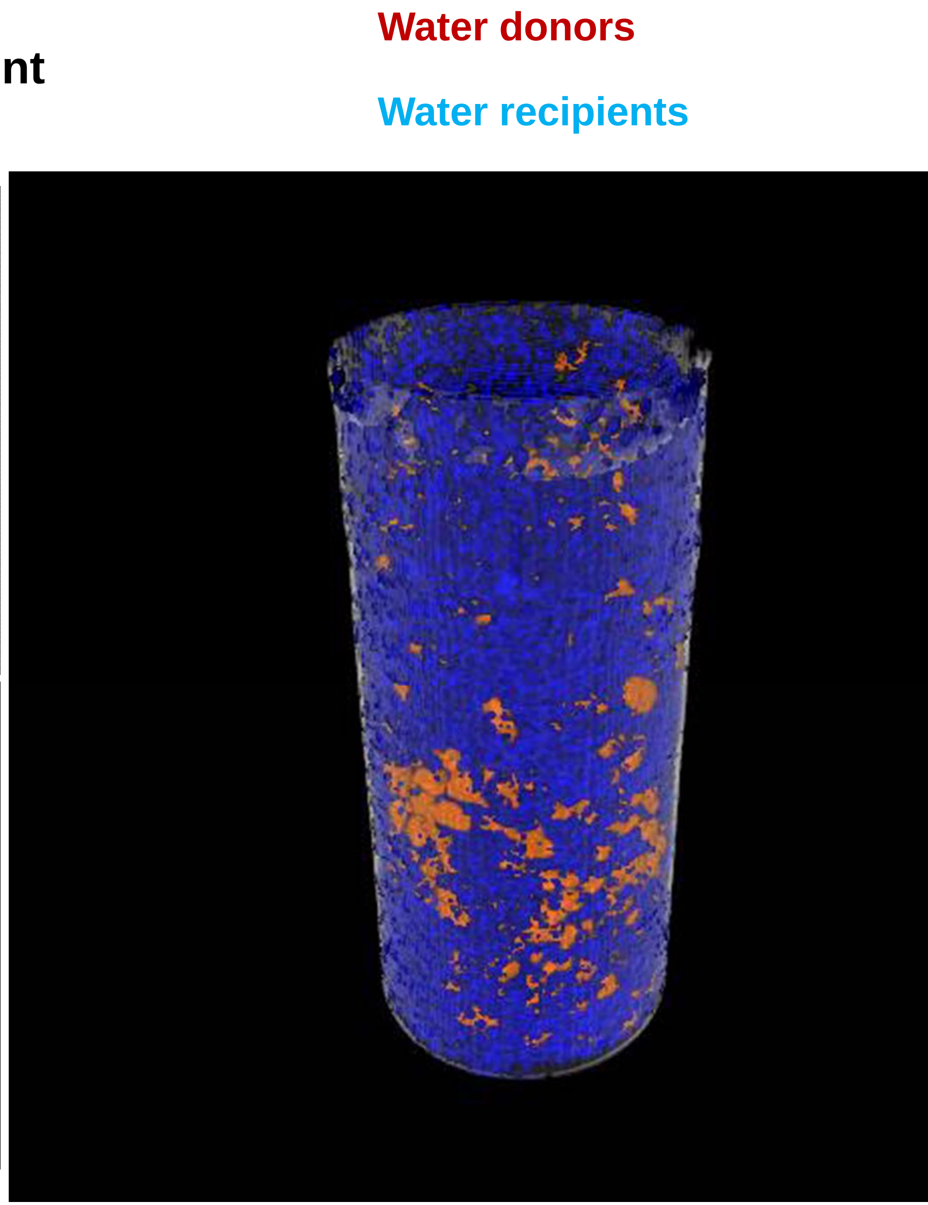


Lura, Wyrzykowski, Tang, Lehmann CCR 59, 2014

Neutron tomography of mortars with LWA (4)

- **Subtraction images to find the changes in water content**

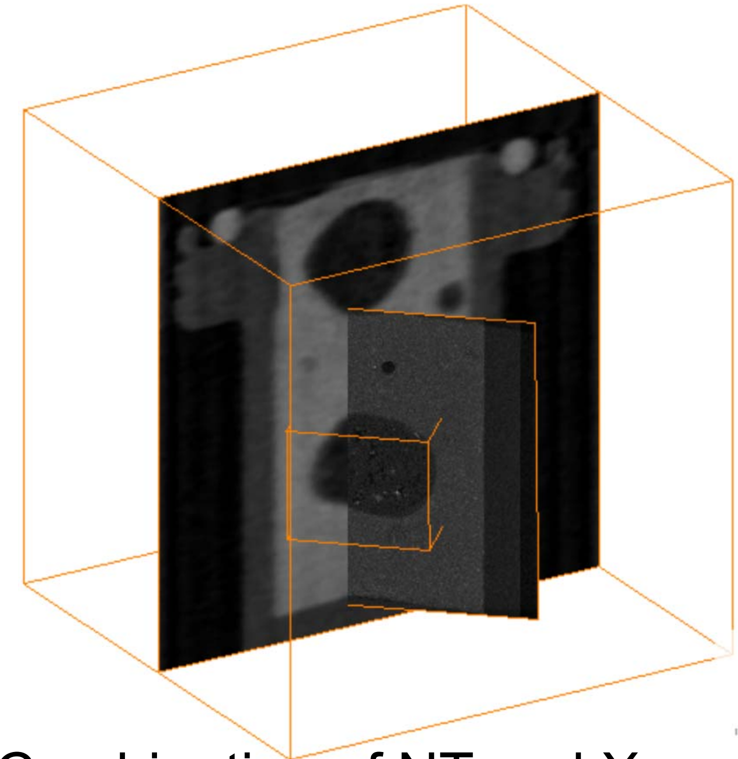


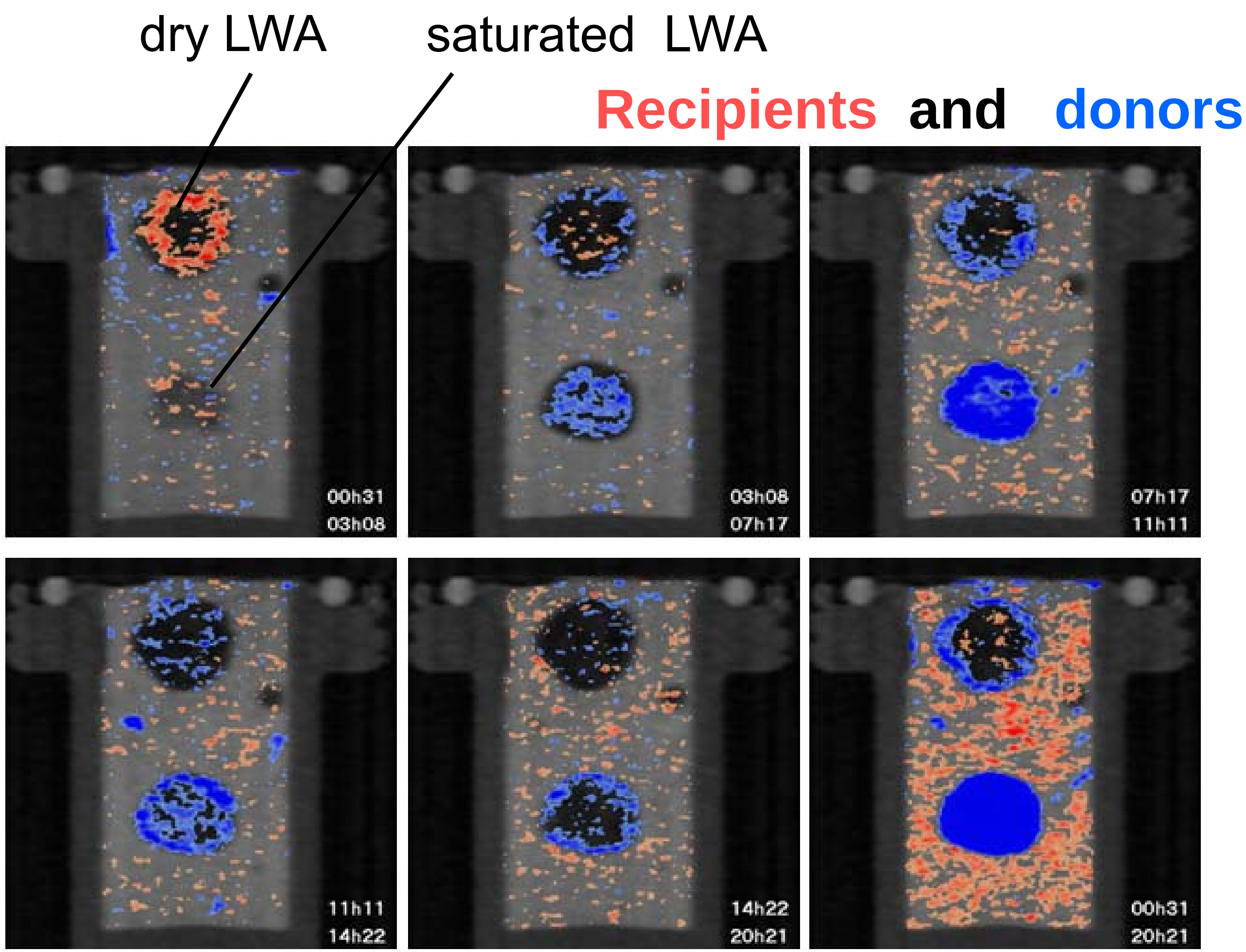


Lura, Wyrzykowski, Tang, Lehmann CCR 59, 2014

Internal curing with expanded shale LWA (Liapor)

Trtik, Münch, Weiss, Kaestner, Jerjen, Josic, Lehmann, Lura, Nucl Instrum Methods Phys Res A 2011

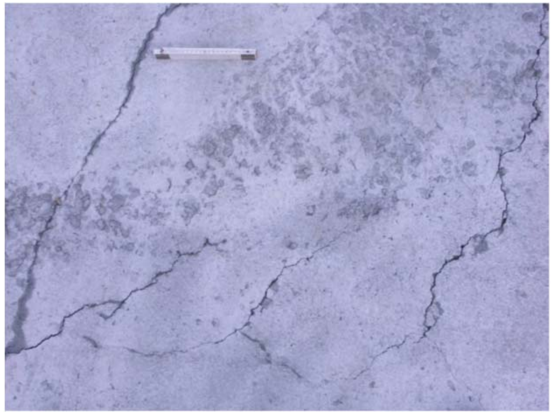




- Combination of NT and X-ray μCT to detect LWA boundaries
- Upper LWA dry, absorbs water from paste, then releases
- Lower LWA saturated, only release

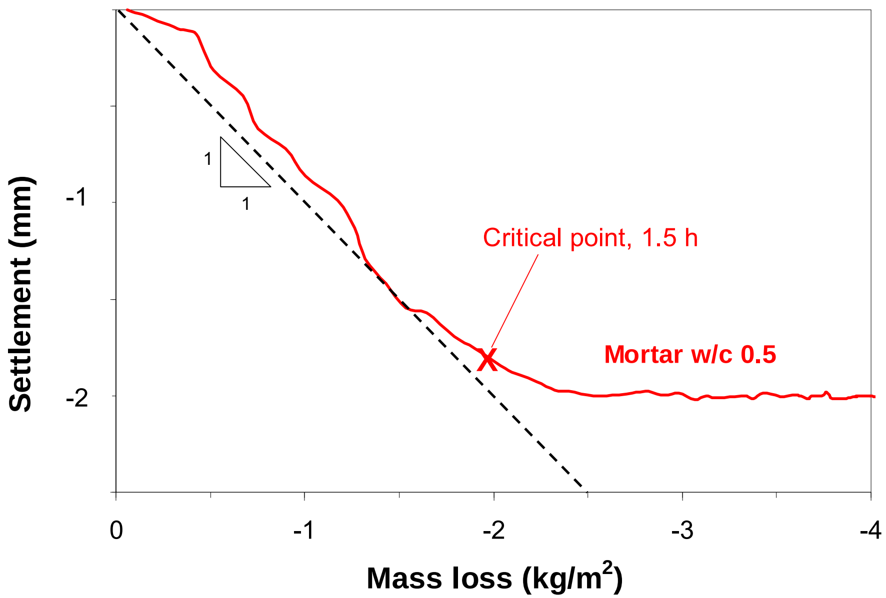
Measurements at ICON

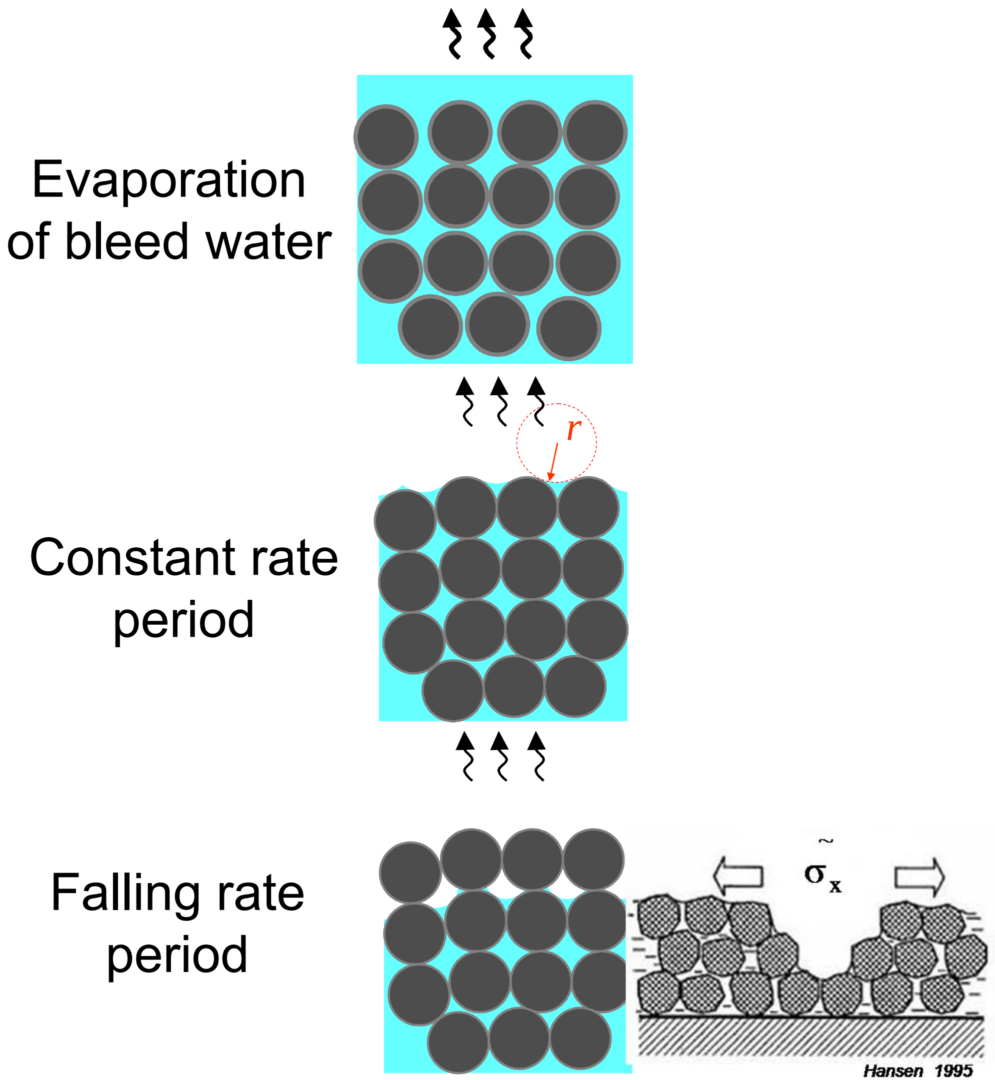
Plastic shrinkage settlement and cracking



Cracks go through concrete slab, water seeps

Photos by A. Leemann





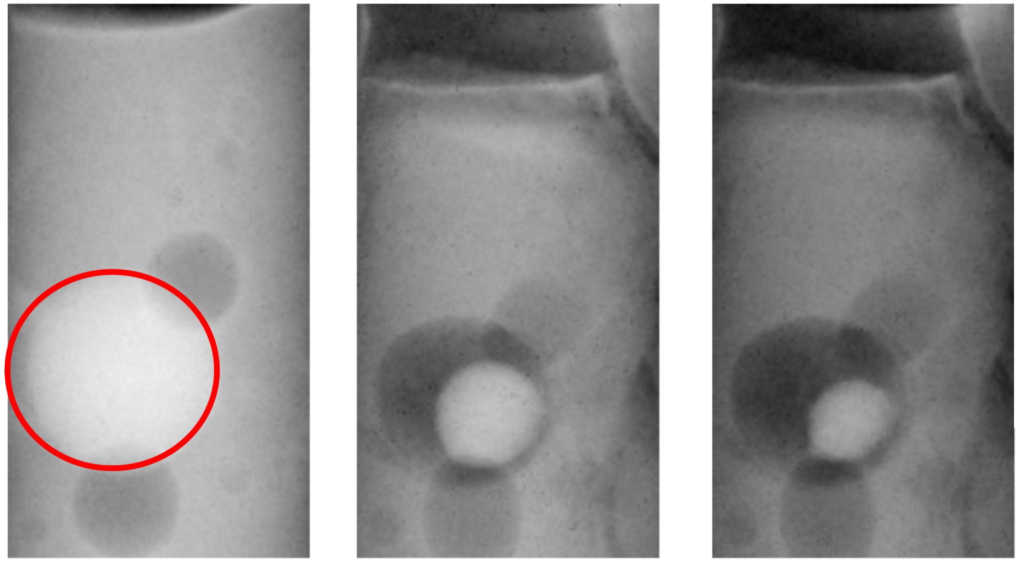
Lura, Weiss et al. ACIMJ 2007

Water release from SAP in fresh pastes

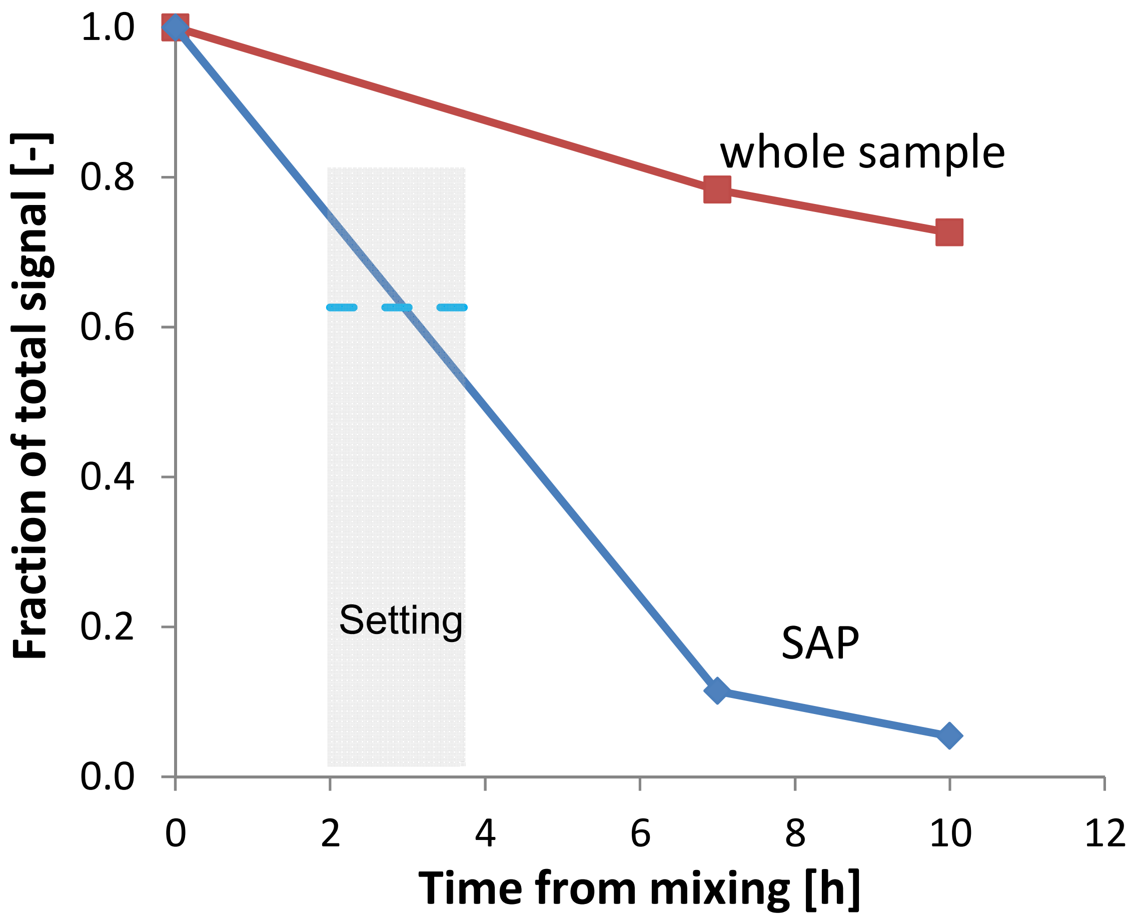
Cement paste w/c 0.3

Temperature: 24 ± 0.5°C

Relative humidity: 25 ± 3 % RH

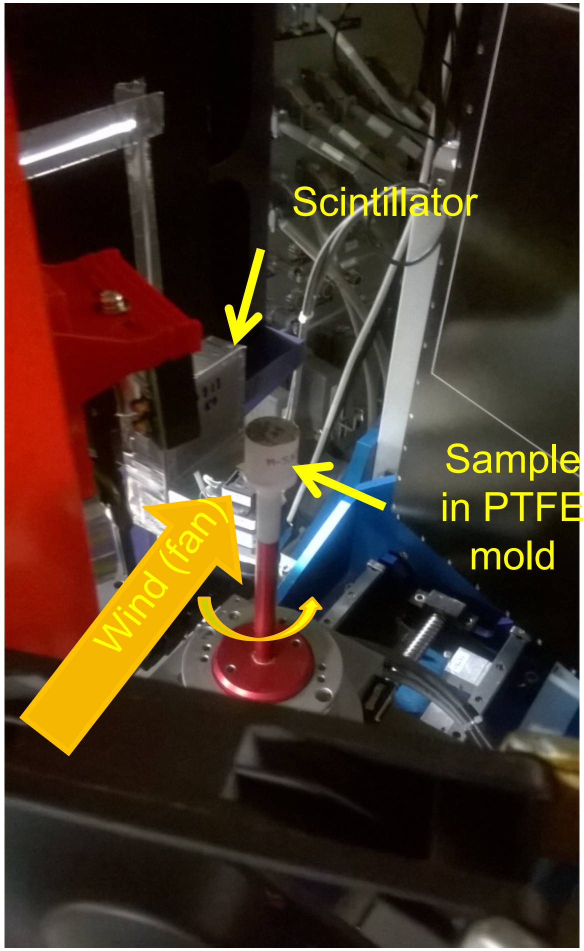


0 hours   7 hours   10 hours



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

Ghourchian, Griffa, Wyrzykowski, Münch, Lura et al., unpublished 2016

Reconstruction from golden ratio sequence

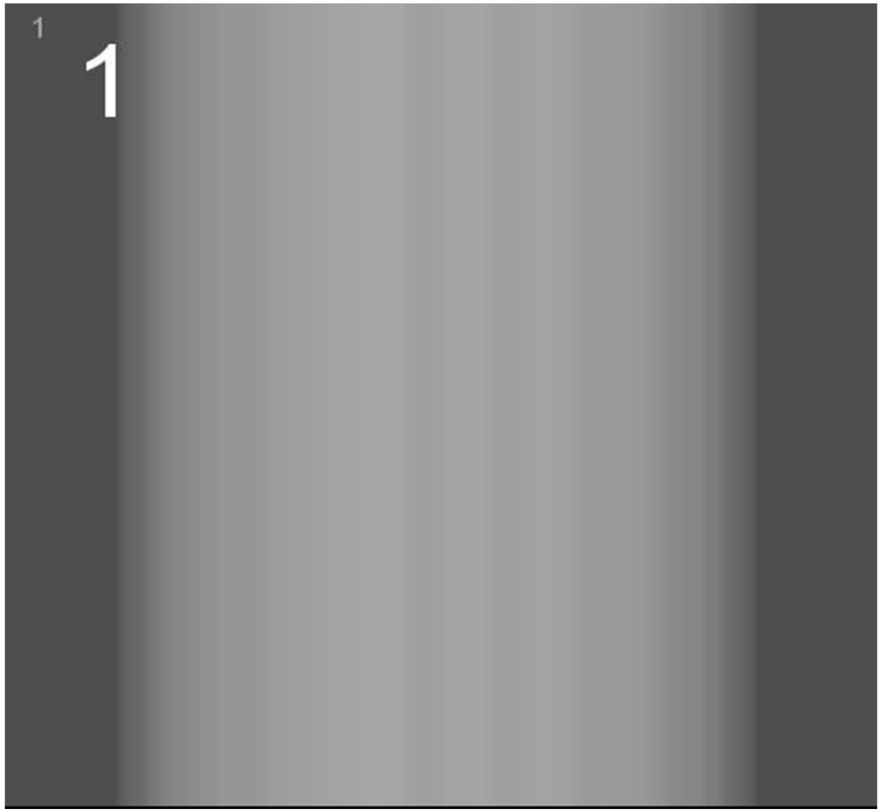
- Observe the sample at oblique angles
- The golden ratio ϕ gives the next angle θ

$$\phi = \frac{1+\sqrt{5}}{2}$$
$$\theta_i = mod(i \cdot \phi \cdot \pi, \pi)$$

- 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

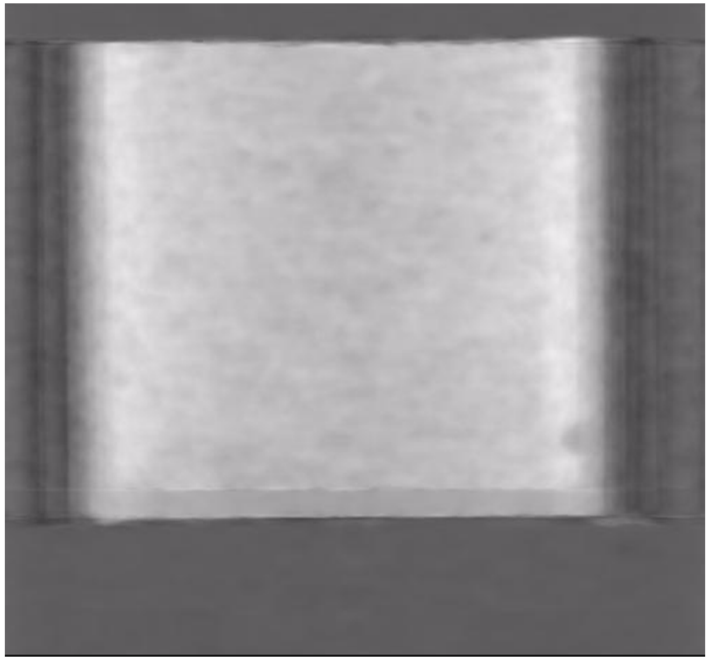


Different reconstruction methods:

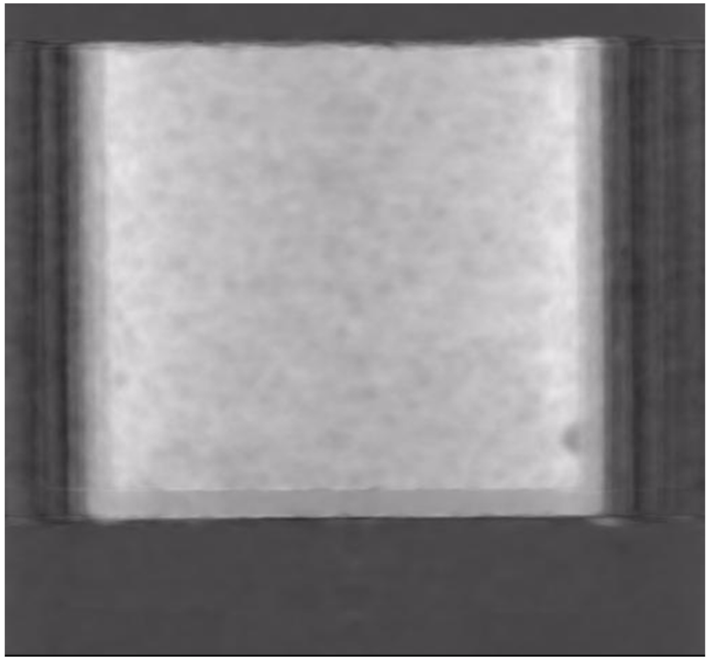
- Filtered back-projection *Kaestner 2011*
- Penalized likelihood *Ahn et al. 2006*
- Spatio-temporal regularization *Kazantsev et al. 2013*
- **SIRT** (Simultaneous Iterative Reconstruction Technique)

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

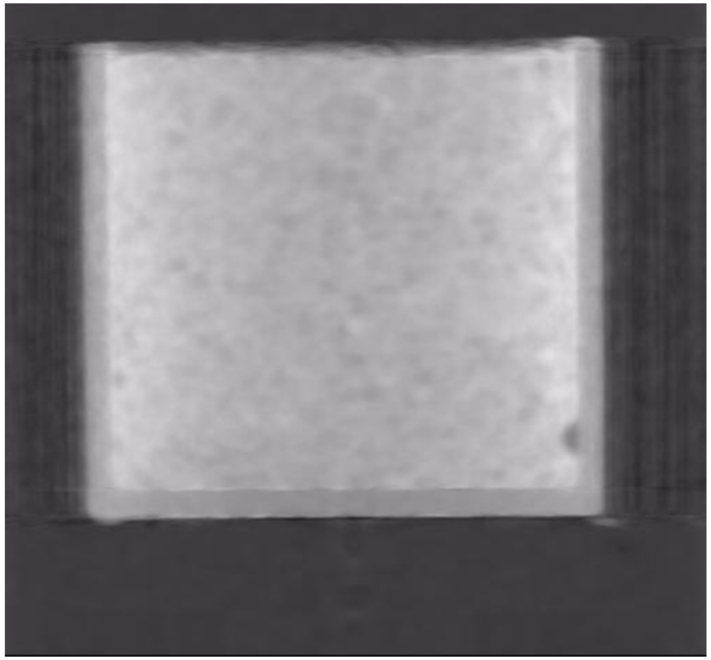
16 projections / 3-D dataset
time span – 9 min / 3-D dataset



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



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

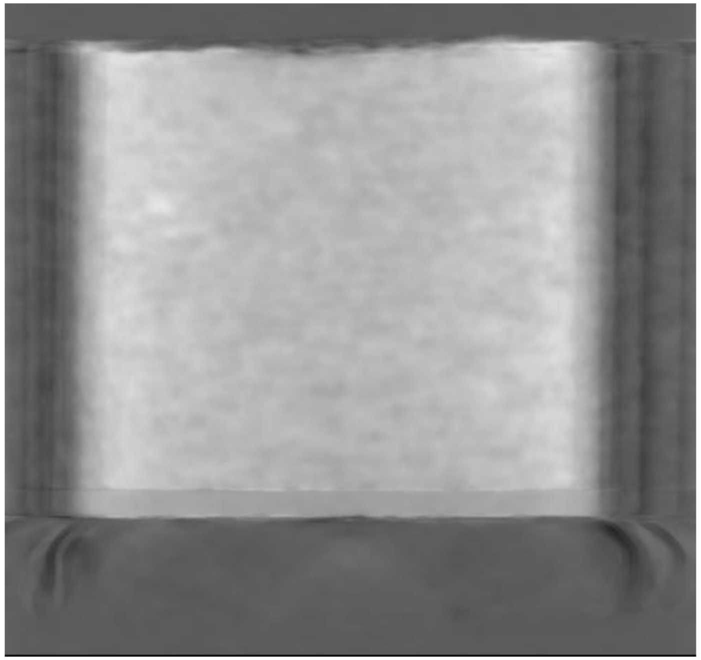


Overall time span – 7 h

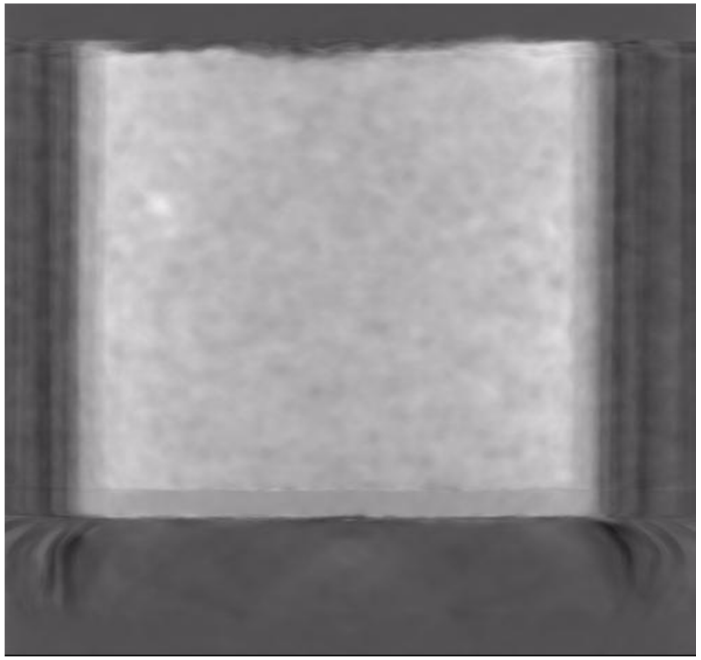
Reference mortar

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

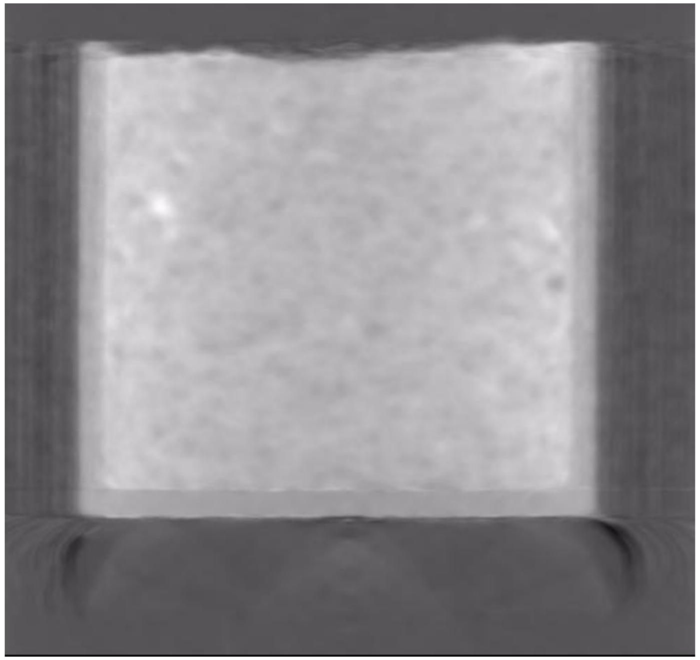
16 projections / 3-D dataset
time span – 9 min / 3-D dataset



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



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



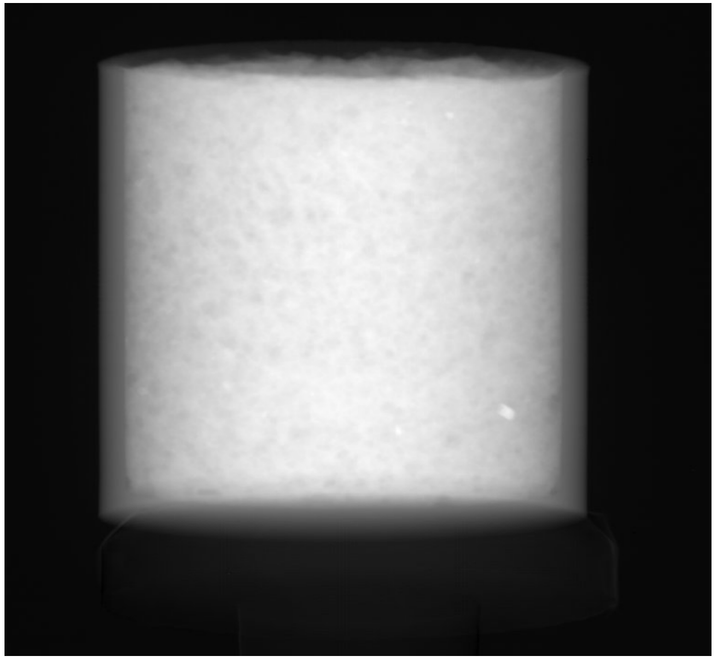
SAP mortar

Overall time span – 6 h

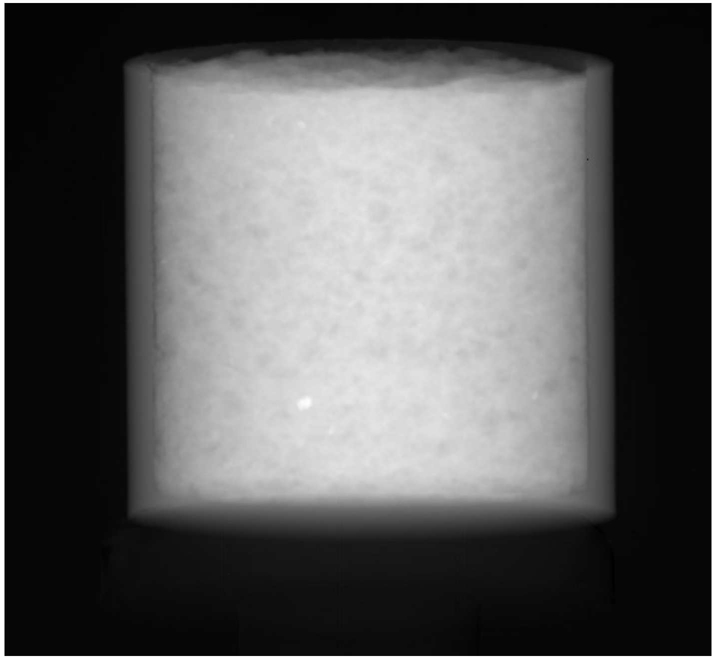
X-ray – in processing

Projections

Time 0 h

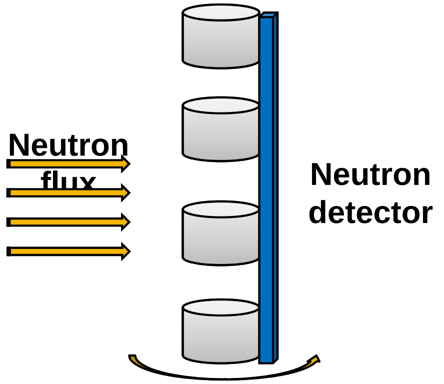


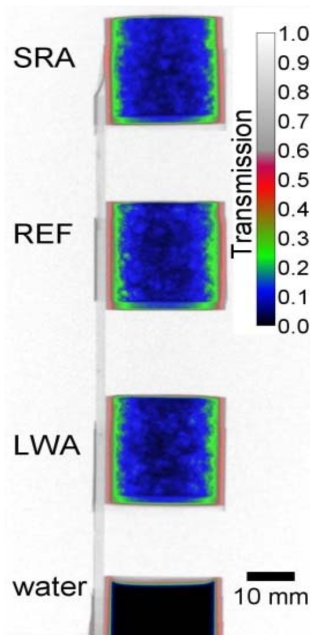
Time 6 h

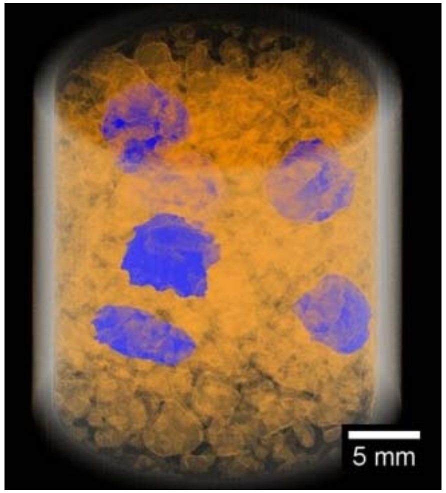


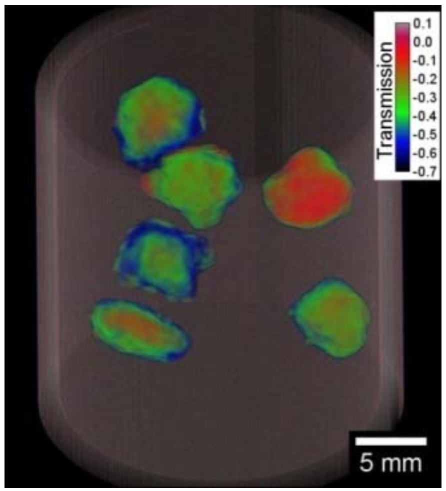
Drying of fresh concrete (plastic shrinkage)

Neutron tomography study of water transport – SRA, LWA

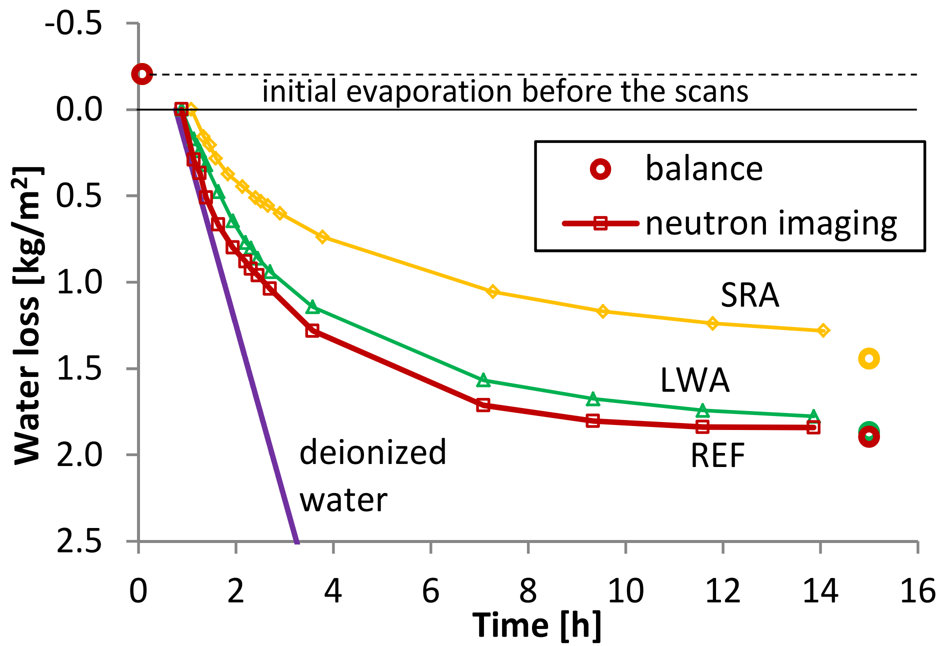


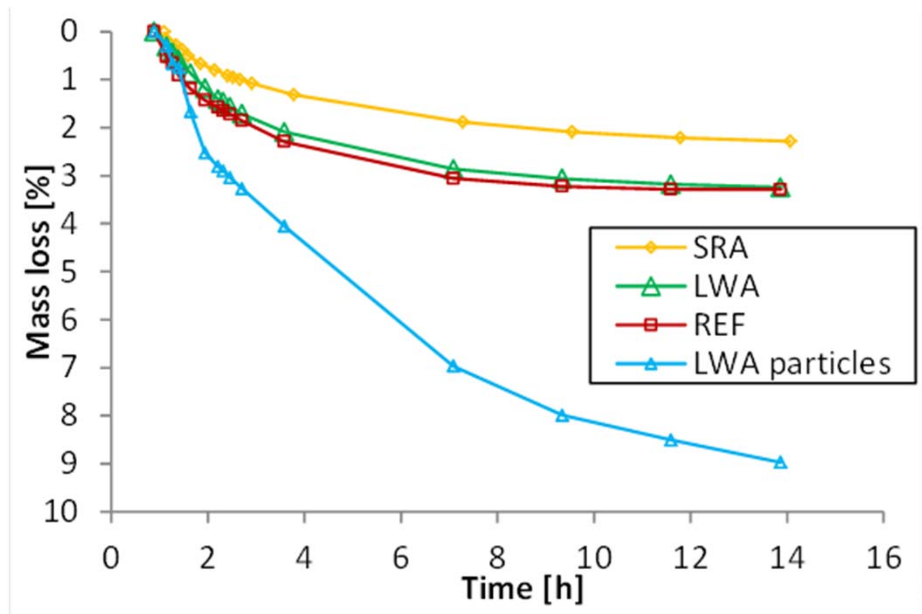






Measurements at NEUTRA

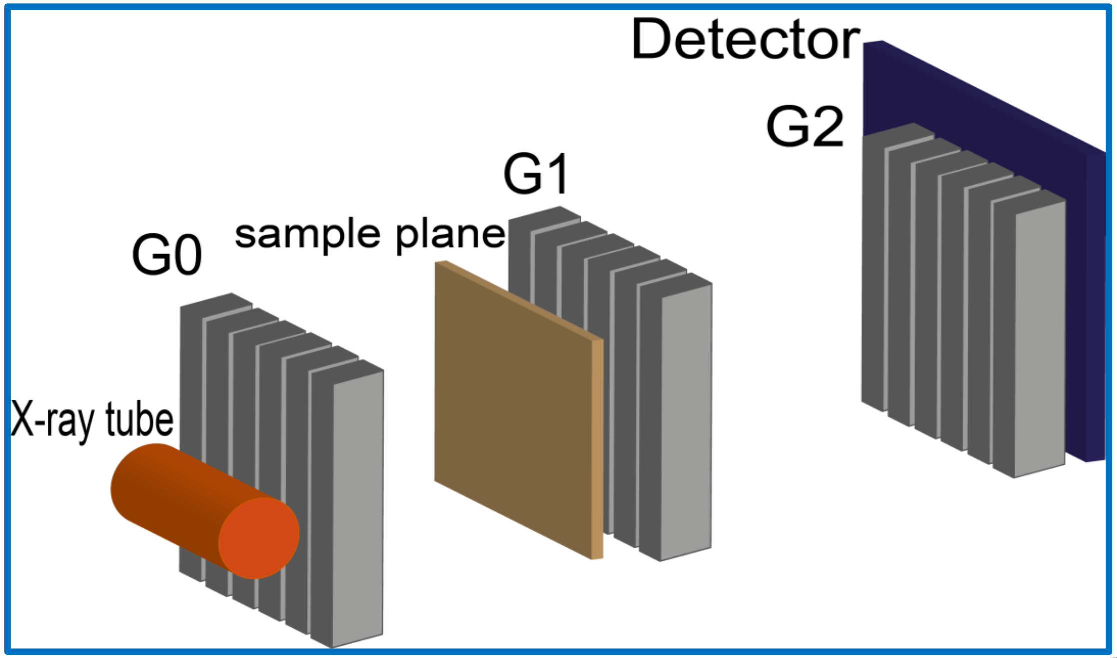




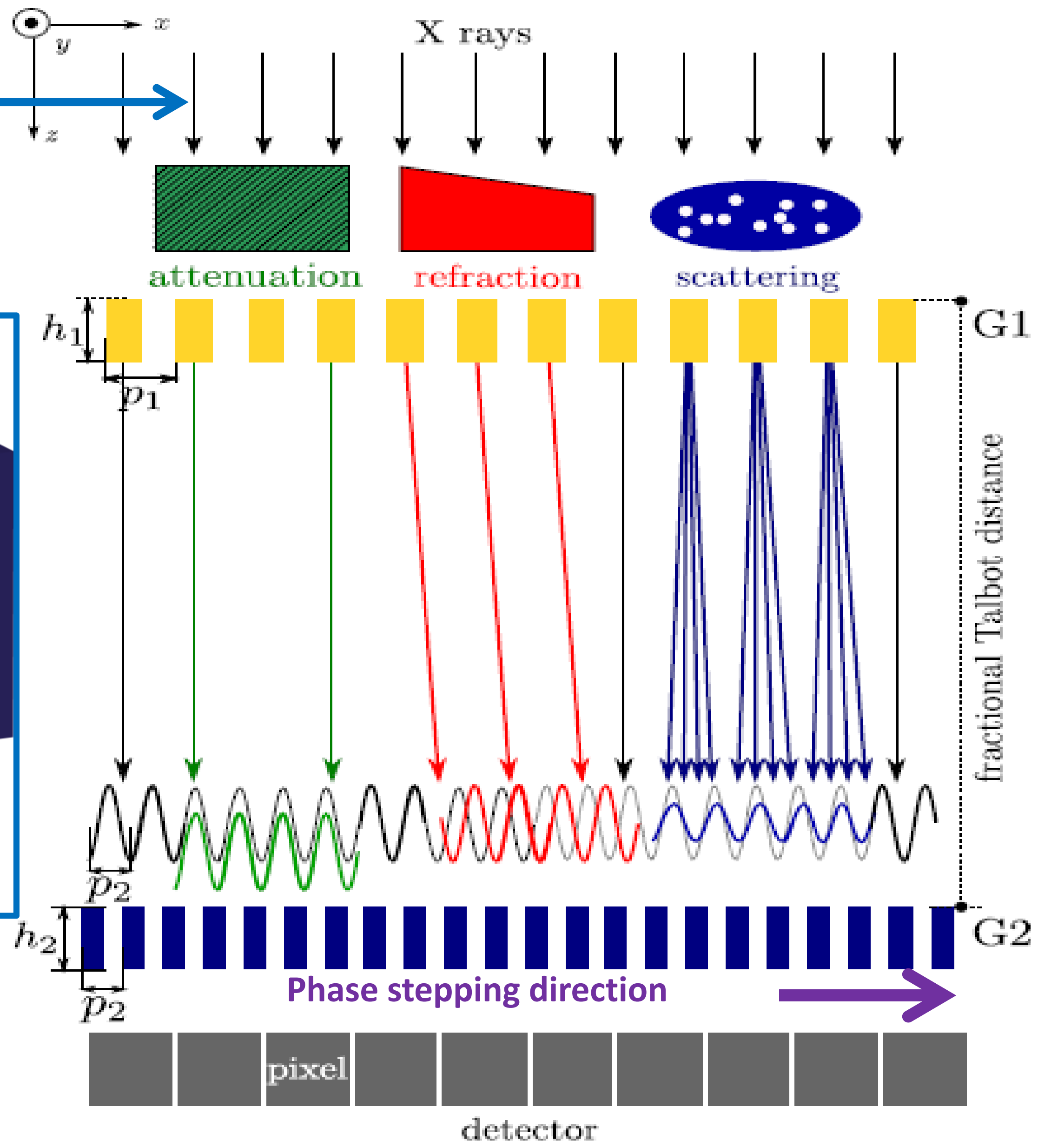
Wyrzykowski, Trtik, Münch, Weiss, Vontobel, Lura CCR 2015

X-ray phase-contrast imaging

Talbot-(Lau) interferometer



ideal for laboratory-scale setup

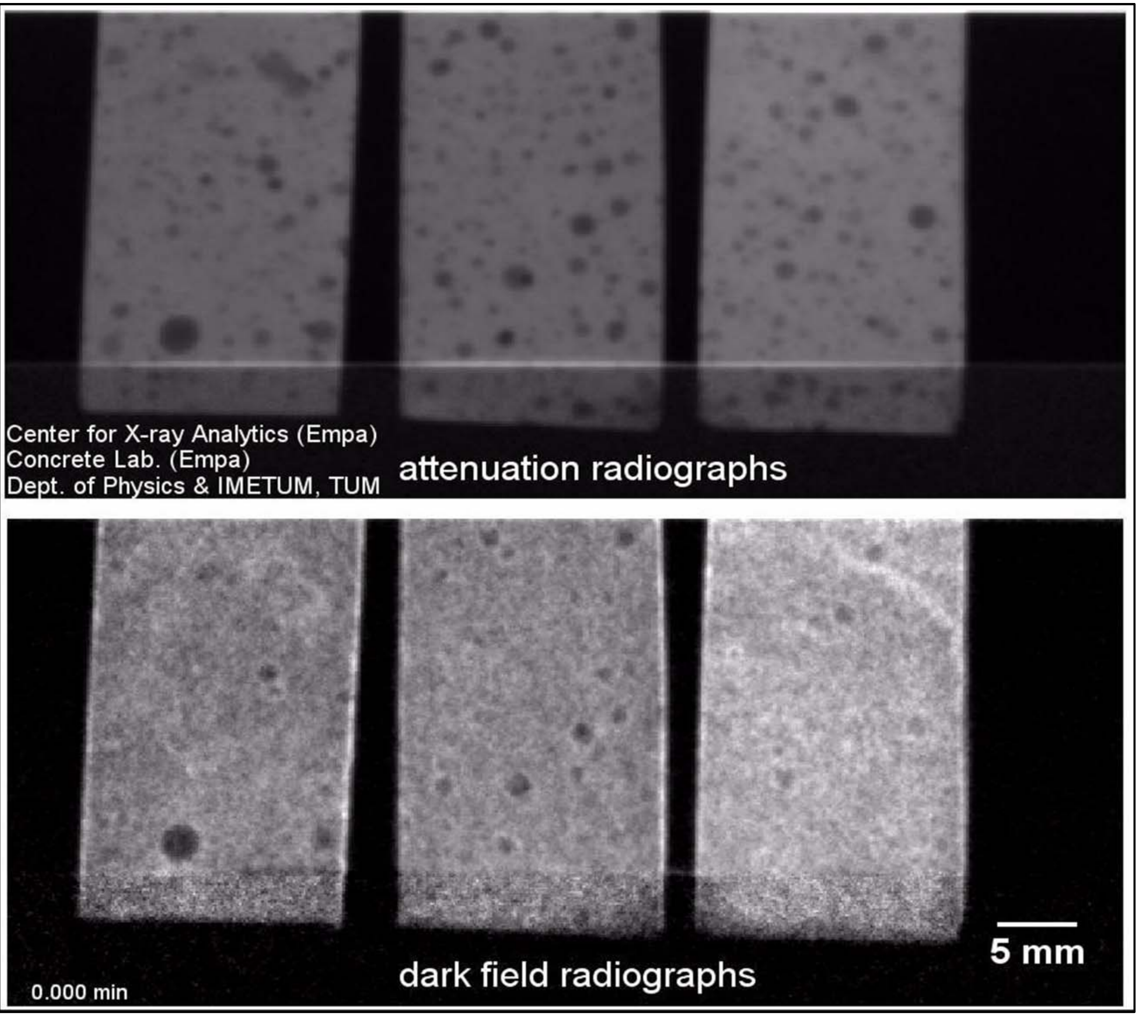


sketch of X-ray grating interferometer
adapted from *Zanette 2011*

Dark-field contrast X-ray imaging of water capillary uptake in mortars

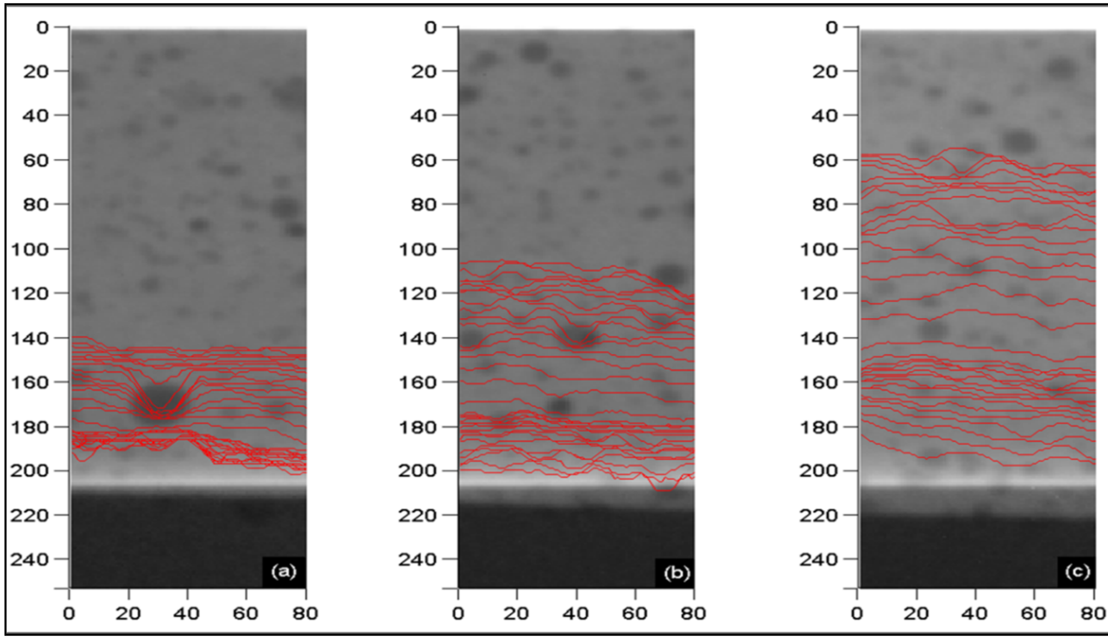
Talbot-Lau interferometer with conventional macro-focused tube

Setup at TU Munich, Germany



Samples of w/c 0.5 mortar 10x20x2mm^3
All dried in an oven at 50^{o}C for 48 h
Left sample pre-conditioned at 200^{o}C for 1 h
Middle sample pre-conditioned at 120^{o}C for 3 h

wetting front profiles extracted from dark-field radiographs

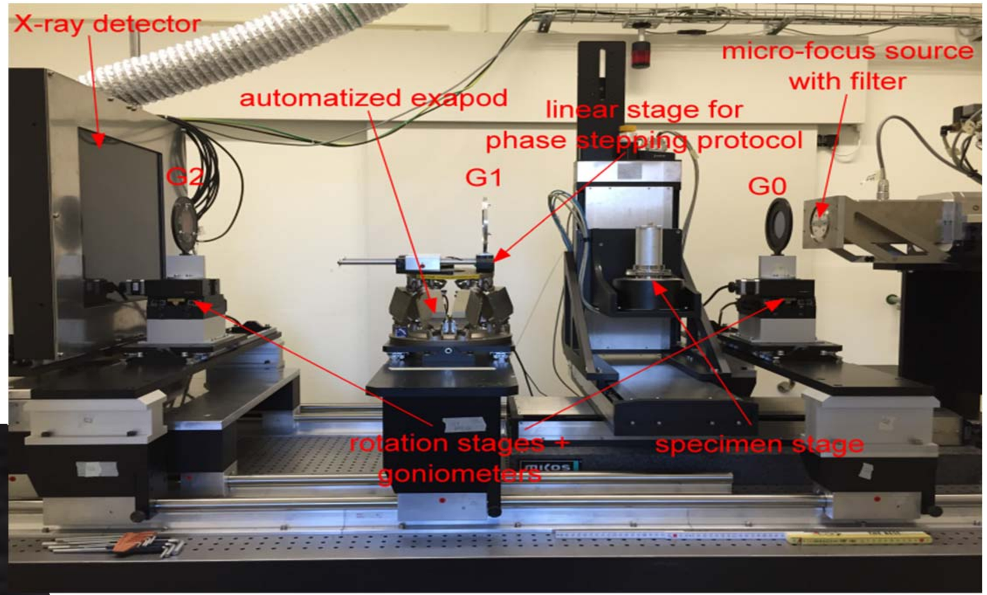


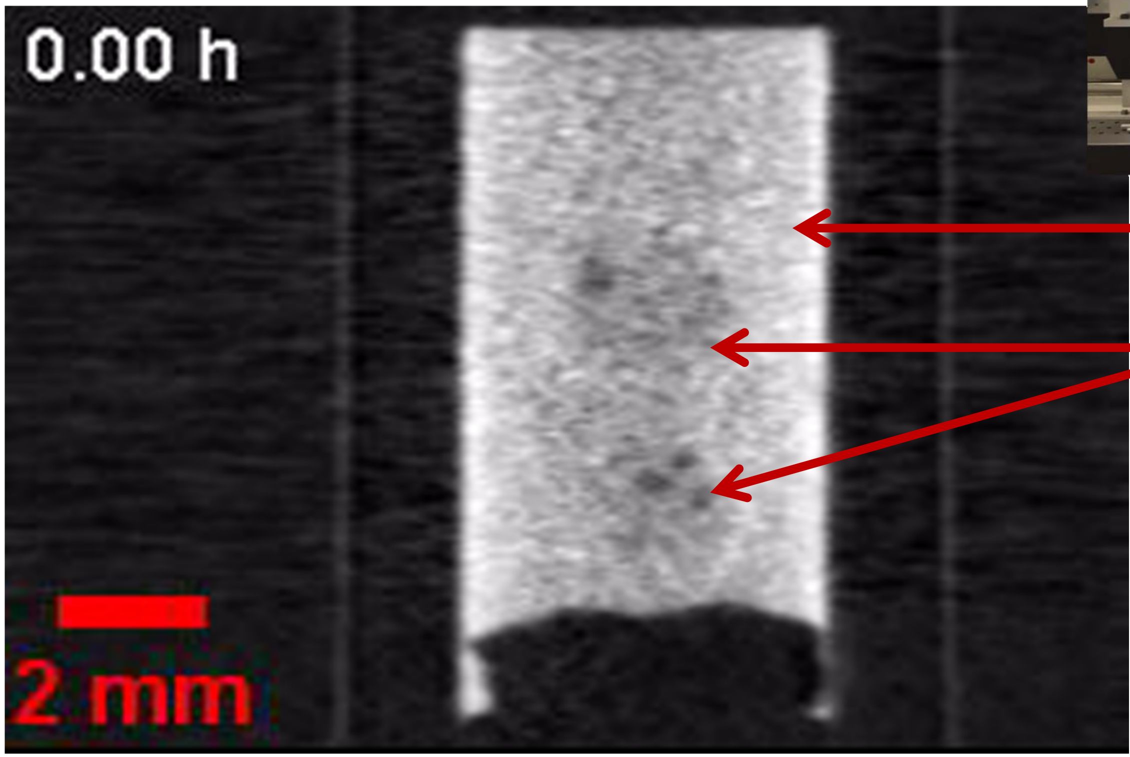
Yang, Prade, Griffa, Jerjen, Di Bella, Herzen, Sarapata, Pfeiffer, Lura Appl. Phys. Lett. 2014

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

  $\Rightarrow$ 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:

- Clarence Tang, Kritsada Sisomphon, Sakprayut Sinthupinyo – *Siam Research and Innovation,* Thailand
- Gregor Herth, Stefan Friedrich, Alexander Assmann – *BASF,* Germany
- Guillaume Jeanson – *SNF Floerger,* France

Main published results

[1] P. Trtik, B. Muench, W.J. Weiss, G. Herth, A. Kaestner, E. Lehmann, P. Lura. Neutron tomography investigation of water release from superabsorbent polymers in cement paste. Proc. Int. RILEM Conf. on Material Science (MatSci), Sept. 6-10, 2010, Aachen, Vol. III, 175-185.

[2] M. Wyrzykowski, P. Lura, F. Pesavento, D. Gawin, Modeling of water migration during internal curing with superabsorbent polymers, Journal of Materials in Civil Engineering 24 (2012) 1006-1016.

[3] P. Lura, M. Wyrzykowski, C. Tang, E. Lehmann, Internal curing with lightweight aggregate produced from biomass-derived waste, Cement and Concrete Research 59 (2014) 24-33.

[4] F. Yang, F. Prade, M. Griffa, I. Jerjen, C. Di Bella, J. Herzen, A. Sarapata, F. Pfeiffer, P. Lura. Dark-field X-ray imaging of water in porous materials. Applied Physics Letters 105 (2014) 154105.

[5] M. Wyrzykowski, P. Trtik, B. Münch, J. Weiss, P. Vontobel, P. Lura, Plastic shrinkage of mortars with shrinkage reducing admixture and lightweight aggregates studied by neutron tomography, Cement and Concrete Research 73 (2015) 238-245.