Proceedings of the 7th GACM Colloquium on Computational Mechanics for Young Scientists from Academia and Industry October 11-13, 2017 in Stuttgart, Germany

X-ray imaging of water transport in porous materials: New possibilities by phase and dark-field contrast

Fei Yang[1,2,3], Michele Griffa[1]*, F. Prade[4], R. Kaufmann[2], A. Bonnin[5], A. Hipp[6], H. Derluyn[7†], P. Moonen[8], M. Boone[9], J. Herzen[4,10], R. Mokso[5‡], F. Pfeiffer[4,10,11], F. Beckman[6], P. Lura[1,3]

Micro Abstract
In this contribution, we show examples of X-ray phase and dark-field contrast X-ray imaging, based on refraction and ultra-small angle scattering of the transmitted X-ray photons, respectively, as applied to cement-based materials during changes in their water content due to distinct processes. We overview what can be gained by using such imaging methods compared with what achievable with standard, attenuation contrast ones.

[1]Concrete/Construction Chemistry Laboratory, Swiss Federal Laboratories for Materials Science and Technology, Empa, ETH Domain, Dübendorf, Switzerland

[2]Center for X-Ray Analytics, Swiss Federal Laboratories for Materials Science and Technology (Empa), ETH Domain, Dübendorf, Switzerland

[3]Institute for Building Materials, Swiss Federal Institute of Technology Zurich (ETHZ), Zürich, Switzerland

[4]Chair of Biomedical Physics, Department of Physics and School of BioEngineering, Technical University Munich, 85748 Garching, Germany

[5]Swiss Light Source, Paul Scherrer Institute, Villigen, Switzerland

[6]Helmholtz-Zentrum Geesthacht, Geesthacht, Germany

[7]UGCT/PProGRess, Dept. of Geology, Ghent University, Ghent, Belgium

[8]Univ. Pau et Pays Adour, LFCR-IPRA and DMEX-IPRA, 64000 Pau, France

[9]UGCT/Dept. of Physics and Astronomy, Ghent University, Ghent, Belgium

[10]Department of Diagnostics and Interventional Radiology, Klinikum rechts der Isar, Technical University Munich, 81675 München, Germany

[11]Institute for Advanced Study, Technical University Munich, 85748 Garching, Germany

***Corresponding author**: michele.griffa@empa.ch
† Current affiliation: Univ. Pau et Pays Adour, CNRS, TOTAL, LFRC-IPRA, UMR 5150, 64000 Pau, France
‡ Current affiliation: MAX IV Laboratory, Lund University, Lund, Sweden

Introduction

Photoelectric absorption and Compton scattering are the two physical processes assumed as the main contributors to the macroscopic attenuation of a X-ray beam transmitted through a specimen. Such attenuation is what creates contrast between distinct material phases (or between regions of distinct density of the same phase) in standard X-ray images (both radiographs and tomograms). Standard, i.e., attenuation-based, X-ray imaging is one of the most used 3D imaging methods for investigating mechanical and fluid (especially water) transport properties of highly heterogeneous porous materials, both natural and man-made ones [1–4]. Related with the investigation of water transport properties, standard X-ray imaging typically requires substituting the liquid water with a water-based salt solution where the salt is made of high atomic number elements and acts as a contrast agent. Compared with normal water such a solution bears higher X-ray attenuation, thus higher contrast is achieved between regions differing in its content. The need to use such a contrast agent strongly limits what can be actually investigated: in the case of a chemically reactive water transport process, the ions in the solution may strongly alter the physics and chemistry of the reactions of the liquid with the solid substrate, thus perturbing the system which should be, supposedly, imaged without too much perturbation. We present in this contribution the implementation, optimization and application to the investigation of water transport in porous materials of other X-ray imaging approaches which do not require the use of contrast agents. Our original driving goal was studying water transport processes in hydrating cement-based materials and respective shrinkage/cracking mechanisms without perturbing the cement hydration reactions. The approaches we present in this contribution rely on two other types of X-ray - matter interaction processes which create different types of contrasts in the final, respective radiographs/tomograms: X-ray refraction (phase contrast) and X-ray small angle scattering (dark-field contrast). Both approaches bear an advantage compared with standard, attenuation contrast X-ray imaging, in addition to not requiring contrast agents: they reach higher sensitivity in detecting strong heterogeneities, e.g., cracks. As examples we show here results from X-ray phase contrast tomographic microscopy of pore-scale 3D visualization of pure water evaporative drying in stones and mortars and from X-ray dark-field contrast imaging of water capillary imbibition in cracked mortar specimens and water release from internal curing particles embedded in high performance cement matrices.

1 Experimental methods and measurements

The first type of measurements we performed were X-ray phase contrast tomographic microscopy ones obtained both with the free-space propagation approach, at the TOMCAT beamline of the Swiss Light Source (Paul Scherrer Institute), achieving an effective spatial resolution of about 10 μm on cylindric specimens of about 5 mm in diameter and 10 minutes of tomographic temporal resolution [5], and with the Talbot interferometry approach at the P07 beamline of DESY/Helmholtz-Zentrum Geesthacht, getting effective spatial resolution of about 5 μm on 5 mm-diameter mortar specimens and 1.5 hours of tomographic temporal resolution [6]. The second type of measurements were X-ray dark-field contrast imaging by Talbot-Lau (TL) interferometry with laboratory-scale interferometers custom developed and implemented at the Chair for Biomedical Physics, Technical University Munich, and at Empa's Center for X-ray Analytics.

2 Examples of results

By theoretical calculations, we initially hypothesized that phase contrast images, based upon information retrieved from the decrement (in respect to unity) δ of the real part of the X-ray complex index of refraction, $n = 1 - \delta + i\beta$, should bear, at the same radiation dose level, up to a ten-fold increase in contrast, compared to standard attenuation-contrast images, based upon β. We confirmed such hypothesis in proof-of-concept studies [5,6] (see Figure 1), where we experimentally showed that phase contrast imaging could resolve water content changes in the absence of any contrast agents and could help with mapping their spatial distribution within the fraction of the pore space with size above the spatial resolution of the imaging system used. When we targeted the visualization of water content changes in the fraction of the pore space with size smaller than the spatial resolution, we showed that that dark-field contrast imaging, based, e.g., upon Talbot(-Lau) interferometry, allows qualitatively visualizing and discerning between regions with different water contents by producing different degrees of X-ray small-angle scattering, thus different dark-field contrast levels [7].

Conclusions

X-ray phase and dark-field contrast imaging provide new opportunities for the spatial-temporal mapping of water content changes in porous materials in the absence of any contrast agent added to the water itself, which is a severe limitation when the water transport process is chemically

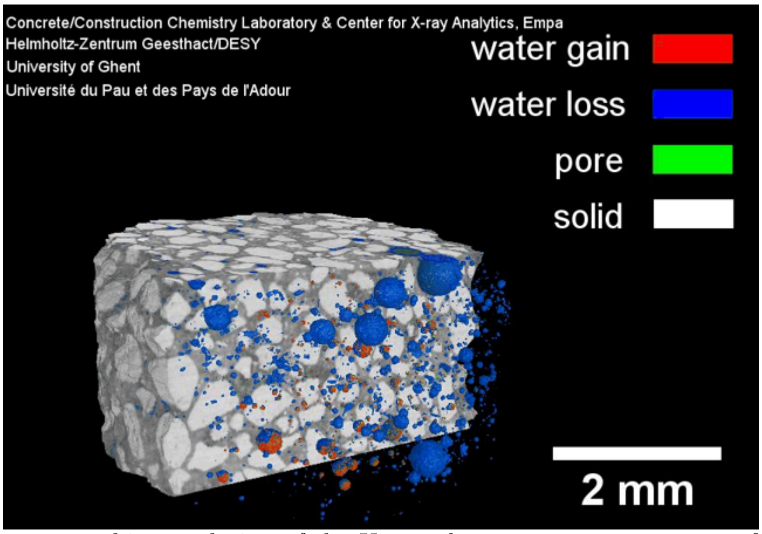

Figure 1. 3D computer graphics rendering of the X-ray phase-contrast tomogram of a mortar specimen at the end of an evaporative drying process. The 3D rendering of the binary images (masks) of pores classified as having undergone either water loss or water gain (blue or red, respectively) during the drying, based upon image analysis of the time-differential phase contrast tomograms, is superimposed on top of the phase contrast tomogram. Adaptation from [6].

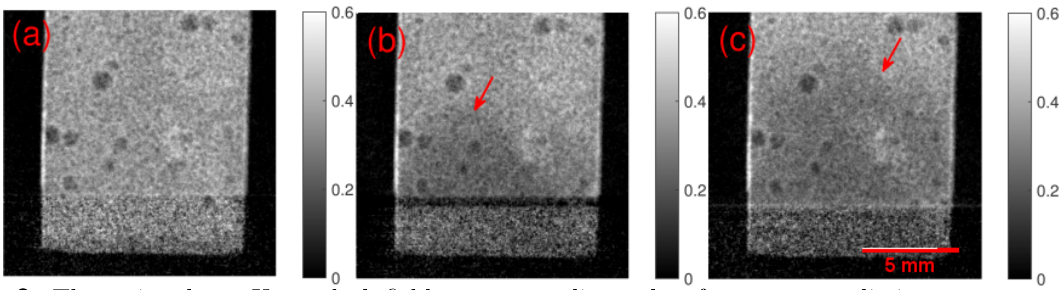

Figure 2. Three time-lapse X-ray dark-field contrast radiographs of a mortar at distinct moments during water capillary imbibition from the bottom. These radiographs were obtained by performing time-lapse Talbot-Lau interferometry measurements at Empa's Center for X-ray Analytics. The pixel value scale is in arbitrary units of $9 \cdot 10^{-12)}$. Larger values indicate stronger cumulative small-angle X-ray scattering, while lower values a weaker one. The red arrows point to the wetting front, visible by the naked-eye, which leads to a decrease in small-angle scattering, i.e., smaller dark-field signal.

reactive, i.e., the solid substrate of the porous material chemically interacts with water and get changed by it. The latter is the case in early-age (i.e., up to several days from casting) cement-based materials, which were the main subjects of interest in our study. We report in this contribution some examples of our work which has shown the higher sensitivity of phase contrast imaging to water content changes in pores with size above the spatial resolution of the images as well as examples of how to locate regions inside the pore space where water content changes occur even when the pore size is below the spatial resolution, in the latter case exploiting X-ray dark-field contrast imaging. Both contrast modalities, complementary to the attenuation contrast of standard X-ray imaging, also allows for better resolving and locating, respectively, microstructural features, e.g., cracks, which are relevant for the investigation of the water transport process themselves, thus empowering the exploitation of X-ray images for computational modeling development and validation.

Acknowledgements

We acknowledge the financial support by the Swiss National Science Foundation via projects Nr. 143782 and Nr. 162572, by the Helmholtz Virtual Institute for New X-ray analytical Methods in Materials science (VI-NXMM, http://roentgenbildgebung.de), and by the DFG Cluster of

Excellence Munich-Centre for Advanced Photonics (MAP) and the DFG Gottfried Wil-helm Leibniz program. This work was carried out with the support of the Karlsruhe Nano Micro Facility (www.kit.edu/knmf), a Helmholtz Research Infrastructure at Karlsruhe Institute of Technology. We thank Pascal Meyer and Jürgen Mohr, Institute of Microstructure Technology at the Karlsruhe Institute of Technology, for the supply of the gratings of the Talbot-Lau interferometers at the Technical University of Munich and at Empa's Center for X-ray Analytics within the framework of the VI-NXMM.

References

[1] V. Cnudde and M. Boone. High-resolution x-ray computed tomography in geosciences: A review of the current technology and applications. *Earth-Science Reviews*, 123:p. 1–17, 2013.

[2] E. Maire. X-ray tomography applied to the characterization of highly porous materials. *Annual Review of Materials Research*, Vol. 42(1):p. 163–178, 2012.

[3] E. Maire and P. Withers. Quantitative x-ray tomography. *International Materials Reviews*, Vol. 59(1):p. 1–43, 2014.

[4] D. Wildenschild and A. Sheppard. X-ray imaging and analysis techniques for quantifying pore-scale structure and processes in subsurface porous medium systems. *Advances in Water Resources*, Vol. 51:p. 217–246, 2013.

[5] F. Yang, M. Griffa, A. Bonnin, R. Mokso, C. Di Bella, B. Münch, R. Kaufmann, and P. Lura. Visualization of water drying in porous materials by x-ray phase contrast imaging. *Journal of Microscopy*, Vol. 261(1):p. 88–104, 2016.

[6] F. Yang, M. Griffa, A. Hipp, A. Derluyn, P. Moonen, R. Kaufmann, M. Boone, F. Beckmann, and P. Lura. Advancing the visualization of pure water transport in porous materials by fast, talbot interferometry-based multi-contrast x-ray micro-tomography. In *Proceedings of Developments in X-Ray Tomography X, Conference dates, San Diego (CA), USA.*, pages 99670L/1–18, San Diego, 2016. SPIE.

[7] F. Yang, F. Prade, M. Griffa, I. Jerjen, C. Di Bella, J. Herzen, A. Sarapata, F. Pfeiffer, and P. Lura. Dark-field x-ray imaging of unsaturated water transport in porous materials. *Applied Physics Letters*, Vol. 105(15):p. 154105/1–5, 2014.