X-RAY IMAGING OF WATER TRANSPORT IN POROUS MATERIALS: NEW POSSIBILITIES BY PHASE AND DARK-FIELD CONTRAST

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Keywords: phase and dark-field contrast, Talbot-Lau interferometry, X-ray imaging, water transport, porous media

Summary: We present a systematic assessment of the new possibilities offered by X-ray phase and dark-field contrast imaging, implemented with different methods and at different experimental facilities, for visualizing and quantitatively investigating water transport processes in porous materials where contrast agents cannot be used in conjunction with standard attenuation-contrast imaging because of chemical reactivity of the transport process itself.

1. INTRODUCTION

Standard X-ray imaging, based upon the assumption of X-ray photoelectric absorption and attenuation due to Compton scattering as the main physical processes bearing contrast, has been extensively used for investigating water transport in natural and man-made porous materials. One of its limitations, in several applications, has been related with the need of increasing the water X-ray attenuation by adding “contrast agents”, i.e., by substituting it with a water-based salt solution (a “brine”) where the salt is made of high atomic number elements. This necessity has strongly limited the range of possible studies, especially in the cases of chemically reactive transport processes.

Motivated by the need of investigating water transport processes in hydrating cement-based materials and respective shrinkage/cracking mechanisms without perturbing the cement hydration reactions, we have implemented and applied X-ray imaging approaches which do not require any contrast agent.

Such approaches are based upon two other types of X-ray - matter interaction processes leading to different types of contrasts in the images: refraction (phase contrast) and small angle scattering (dark-field contrast). In addition to not requiring contrast agents, X-ray phase and dark-field contrast imaging allow achieving higher sensitivity in detecting strong heterogeneities as cracks.

In this contribution, we show examples from X-ray phase contrast tomographic microscopy of pore-scale 3D visualization of pure water evaporative drying in stones 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

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high performance cement matrices.

2. EXPERIMENTAL METHOD

We performed X-ray phase contrast tomographic microscopy both in free-space propagation mode at the TOMCAT beamline of the Swiss Light Source (Paul Scherrer Institute), achieving about 10 μm of effective spatial resolution and about 10 minutes of tomographic temporal resolution [1], and based upon Talbot interferometry at the P07 beamline of DESY/Helmholtz-Zentrum Geesthacht, achieving effective spatial resolution of 5 μm and 1.5 hours of tomographic temporal resolution [2].
We performed 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.

3. RESULTS

Theoretical calculations suggest that the 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 lead, at the same radiation dose level, up to a ten-fold increase in contrast, compared to standard attenuation-contrast images, based upon β. In proof-of-concept studies [1,2] we experimentally showed that X-ray phase contrast imaging allows indeed resolving water content changes and mapping their spatial distribution within the part of the pore space directly resolvable by the imaging system used.
For pores with size below the spatial resolution, we unexpectedly found and showed for the first time [3] that dark-field contrast imaging based, e.g., upon Talbot(-Lau) interferometry, allows (so far only) qualitatively visualizing and distinguishing between regions with different saturation degrees producing different degrees of X-ray small-angle scattering, thus different dark-field contrast levels [3].

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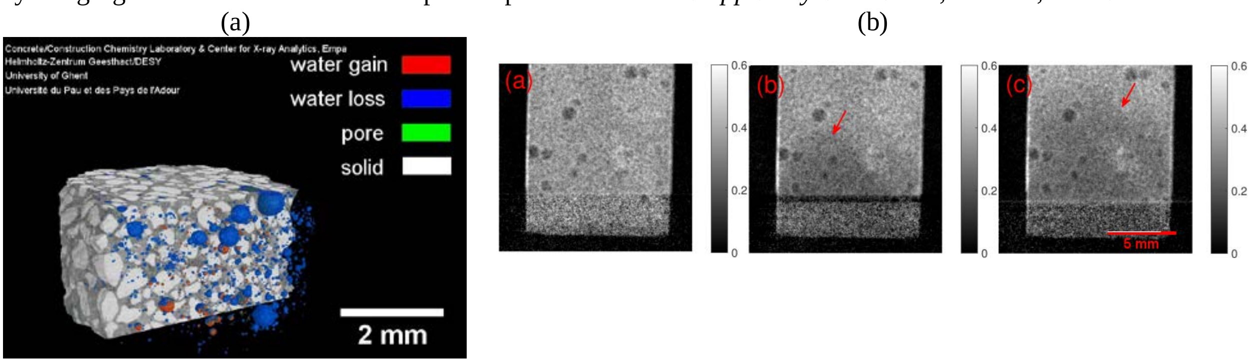

Figure 1: (a) 3D rendering of the X-ray phase-contrast tomogram of a mortar specimen at the end of an evaporative drying process, superimposed to the 3D rendering of the 3D binary images (masks) of pores classified as having undergone either water loss or water gain (blue or red, respectively) during the drying, based upon image analysis of the time-differential phase contrast tomograms. Adaptation from [2]. (b) X-ray dark-field contrast radiographs of a mortar while undergoing water capillary imbibition from the bottom, obtained by performing time-lapse Talbot-Lau interferometry measurements at Empa's Center for X-ray Analytics. The distinct insets refer to different time instants during the process. The pixel value scale is in (arbitrary) units of 9×10^{-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.