Enhancing X-ray imaging of liquids in porous materials

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Abstract

Standard, attenuation-based X-ray imaging of liquids in porous materials requires adding to the liquid a “contrast agent”, i.e., a substance containing e.g. Cs, Ba or I, to increase its linear attenuation coefficient. This requirement is a strong limitation for imaging chemically reactive liquid transport processes, e.g., water transport in concrete and in polymer electrolyte membrane fuel cells (PEMFC) during operation. We present one new approach to X-ray imaging of weakly attenuating liquids, e.g. pure water, in materials with pore space dominated by small pores (below hundreds/tens of microns). This approach does not require any contrast agent but exploits the intrinsic, sub-pixel scale X-ray multi-refraction due to the microstructural heterogeneity of the materials themselves. We show one implementation based upon laboratory-scale Talbot-Lau interferometry with a polychromatic source, applied to radiography of pure water imbibition in concrete samples. This approach has the potential to become complementary to neutron and magnetic resonance imaging of water in porous materials.
1 Introduction

In porous materials, e.g., several building materials, water ingress can cause unwanted mechanical deformation/cracking or high degrees of pore saturation which might result in damage upon freezing [1]. The total water content can be measured gravimetrically, but the distribution of the wetting front inside the materials needed so far methods like MRI [2] or neutron imaging [3], which show only a moderate spatial resolution. High spatial resolution X-ray imaging was only possible with contrast agents, which can change the chemistry of the material-fluid interactions and which are therefore only used if absolutely necessary [4, 5]. In this work an imaging method using ultrasmall angle scattering at the material's microstructure instead of absorption contrast is introduced to visualise the water spatio-temporal distribution.

2 Method

Imaging based on ultra-small-angle scattering is called X-ray dark-field imaging, similar to optical dark field imaging. It visualises the strength of the multiple-scattering at the material’s microstructure, well below the spatial imaging-resolution of the X-ray system [6, 7, 8]. Water inside the pores of the material reduces the multiple-scattering since the difference of the refractive index between material and pores is reduced [9]. This method is therefore well suited to image water- ingress into porous materials. The dark-field images are acquired by a X-ray Talbot-Laue grating interferometer (see figure 1) [10], which simultaneously gives the absorption-, phase-contrast- and dark-field-image of an object. The simultaneous acquisition of three radiographs with different contrasts represents a key advantage of this technique, together with the possibility of implementing it with standard industrial X-ray sources.

In order to prove the feasibility and the potential of X-ray dark-field imaging of water in a porous materials a water capillary uptake experiment with mortar samples and a Talbot-interferometer was designed. Liquid capillary uptake consists in the unsaturated flow driven by capillary forces at boundaries between liquid and bulk. Mortar was chosen as test material because of the relevance of water transport in several processes related with its microstructural development during cement hydration and with its durability.

Three identical samples of $10 \times 20 \times 2 \text{ mm}^3$ were fabricated and dried at 50 °C for 48 hours (preconditioning). One sample was then heated up to 200 °C for one hour in order to induce thermal micro-cracking. Another sample was conditioned at 120 °C for three hours. The third sample went untreated. After the conditioning a 0.07 mm thick capton film tape was applied to each sample’s lateral surface, except for the bottom, which was put in contact with demineralised water. This tape provides high transparency to X-rays...
Figure 1: Principle setup of a Talbot-interferometer. The first grating acts as a phase-shifting grating generating an interference pattern. The second grating in front of the detector is an analyser grating to measure the interference pattern with the help of a phase-stepping procedure.

and reduces the evaporation from the lateral surfaces. With such a setup the water transport mostly happens along the samples vertical direction. The overall experiment lasted about 13 h. During the first hour images were taken every 11 s, while in the following twelve hours every 131 s. A higher sampling frequency was required during the first hour in order to improve the capture of the initial stages of the uptake.

3 Results

The three samples were expected to differ in terms of their liquid sorptivity, as it relates to the amount of dehydration and thermal micro-cracking. Figure 2 gives an impression of the obtained images for the three samples. The wetted part of each sample is clearly visible in the dark-field images ((a) and (b) in figure 2). In those images the wetted regions appear darker than the non-wetted ones, confirming the initial hypothesis about the scattering reduction by water permeating the material porosity.

The capillary uptake is characterised by a wetting front proceeding from bottom to top, which is not completely uniform along the sample width (see figure 3). This is mainly due to the microstructural inhomogeneity of the porous material, specifically the presence of large spherical pores created by air entrapped during the mixing of the mortar, but boundary effects play also a role in determining the level of inhomogeneity for the wetting front. The red lines in figure 3 correspond to different time instants, starting at about 2.9 min after immersion into water, and the time gap between two successive lines corresponds to 25 radiographic sampling steps (i.e., 11 s at first and 131 s after one hour). These wetting fronts confirm the increase of sorptivity with increasing thermal preconditioning of the samples, as expected. The full
Figure 2: X-ray radiographs at start and after one hour of the capillary uptake. (a) and (b) dark-field radiographs. Brighter pixel values correspond to larger scattering strength. (c) and (d) attenuation radiographs. Brighter pixel values correspond to larger X-ray attenuation. The bright horizontal lines visible towards the bottom in (c) and (d) are artifacts created by evaporation from the water surface during the experiment.

time-series of the wetting front was compiled into a video sequence, which can be downloaded from

www.calcolodistr.altervista.org/en/work/...sunto/Suppl_Mats_Movie_CapillaryUptake.gif

Figure 3: Wetting front positions (red lines) overlaid on top of a central region of interest of the last absorption radiograph, for the three different samples. For each sample, the first wetting front is plotted at approximately 2.9 min after water immersion. The time gap between two successive fronts is about 4.6 min.

In order to quantify this result the contrast-to-noise ratio $CNR$ between a region at the bottom (mostly wet) and a region at the top (mostly dry) was calculated for each sample and for both types of images:

$$CNR = \frac{|I_t - I_b|}{\sqrt{\sigma_t^2 + \sigma_b^2}}, \quad (1)$$

where $I_t, b$ is the average intensity in the top and bottom region, respectively, and $\sigma$ denotes the standard deviation in these regions. A large $CNR$ value
indicates a good separation between wet and dry regions. Figure 4 clearly indicates that the CNR values increase with time for dark-field contrast images, whereas they stay roughly the same for absorption based images.

![Figure 4: Contrast to noise ratio CNR (see equation 1) as a function of water uptake time for three samples and absorption (abs, in blue) and dark field imaging (dfi, in red) contrast.](image)

### 4 Conclusion

It could be shown that X-ray dark field imaging is suitable to image water fronts during capillary uptake processes in porous objects like building materials. This was proven with a water uptake experiments with mortar samples and a Talbot-Lau X-ray interferometer using a conventional industrial X-ray tube. The experiment lasted for 13 hours and used a time series of 2D radiographies of the samples. Further details of this experiment can be found in [9].

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### References


