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# Liquid water uptake in unconfined Callovo Oxfordian clay-rock studied with neutron and x-ray imaging

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## Abstract

The Callovo Oxfordian clay-rock (COx) is studied in France for the disposal of radioactive waste, because of its extremely low permeability. This host rock is governed by a hydromechanical coupling of high complexity. This paper presents an experimental study into the mechanisms of water uptake in small, unconfined, prismatic specimens of COx, motivated by the comprehension of cracking observed during concrete/COx interface sample preparation. Water uptake is monitored using both x-ray tomography and neutron radiography, the combination of these imaging techniques allowing material deformation and water arrival to be quantified respectively. Given the speed of water entry and crack propagation, relatively fast imaging is required: 5 minute x-ray tomographies and ten-second neutron radiographies are used. In this study, pairs of similar COx samples from the same core are tested separately with each imaging technique. Two different orientations with respect to the core are also investigated. Analysis of the resulting images yields with micro- and macro-scale insights into hydro-mechanical mechanisms to be obtained. This allows the cracking to be interpreted as a rapid breakdown in capillary suction (supposed large both to drying and rebound from *in-situ* stress state) due to water arrival, which in turn causes a loss of effective stress, allowing cracks to propagate with ease, which in turn deliver water further into the material.

## 1 Introduction

The management of radioactive waste is an important environmental issue, in particular in the countries where nuclear power is generated. In Eastern France, the French National Radioactive Waste Management Agency (Andra) is studying the behaviour of a deep geological repository in Callovo Oxfordian clay-rock (COx). The permeability of the Callovo Oxfordian stratum is extremely low, it is however governed by complex thermo-hydro-mechanical behaviour, which is continuously under investigation with both laboratory and in-field experiments ([1]-[4]).

During the operation phase, tunnels will be excavated and ventilated, until their final closure. In specific sections, swelling clay like bentonite, will be emplaced in between two concrete plugs in order to seal the tunnel and apply pressure on the rock wall, in sections where the concrete lining will be removed ([5, 6]). The study of the mechanical behaviour of the concrete/COx interface in the lab using among others the newly-developed SInC box [7], requires representative and reproducible concrete/COx samples to be created under controlled conditions.

During the preparation of these samples when fresh concrete has been poured directly onto the unconfined COx, macro-cracks have been detected before starting mechanical experiments. Many authors have studied the effect of drying and wetting cycles on different clay-rocks; on Callovo Oxfordian clay-rock samples with saline solution ([8, 9, 10]) and *in situ* on Tournemire

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45 clay-rock ([11, 12]) and on Callovo Oxfordian clay-rock ([13]). Guillon et al [10] have performed  
 46 static atmosphere drying tests which highlighted the hydro mechanical coupling and mechanical  
 47 anisotropy of the samples. Both wetting and drying paths were explored, allowing the potential  
 48 hysteretic behaviour of the samples. During imbibition, the swelling of the sample is mainly  
 49 achieved by the interaction of water with swelling clay sheets (illite/smectite interstratified, [14]).  
 50 The opening of some cracks can also be responsible for the dilation of the sample. At a smaller  
 51 scale, Wang et al [15] also studied the evolution of the COx clay-rock under hydric cycles using  
 52 techniques based on the combination of environmental scanning electron microscope (ESEM) and  
 53 digital image correlation (DIC) [9], in order to quantify local strain field. The observation is carried  
 54 out on zones of several hundred micrometers, so that the evolution of such material under hydric  
 55 loadings can be studied at the scale of inclusion-matrix composite. A heterogeneous strain field  
 56 is evidenced at the microscale during wetting. Nonlinear deformation is observed at high relative  
 57 humidity (RH) which is related not only to damage, but also to the nonlinear swelling of the  
 58 clay mineral itself, controlled by different local mechanisms depending on relative humidity. This  
 59 sensitivity to variations in water content leads to enhanced variations of mechanical parameters in  
 60 response to water saturation.

61 In the case of poured concrete/clay-rock interface, the cause of these macro-cracks must be  
 62 further investigated in order to understand whether they reflect site conditions, and furthermore  
 63 since their presence may affect the representativeness of the measured mechanical response in the  
 64 lab. In the lab (and presumably on site) the exposed, drying face of the clay-rock is rapidly wetted  
 65 by the free water of the fresh concrete. To gain some insight into the causes of this phenomenon, a  
 66 number of *in-situ* wetting experiments have been carried out with high-speed lab x-ray tomography  
 67 (in Laboratoire 3SR) and neutron radiography (at the ILL instrument D50-tomo or “NeXT”, an  
 68 instrument that has recently come online in Grenoble [16]).

## 69 2 *In-situ* imaging techniques

70 Full-field imaging is a very precious tool in experimental mechanics especially for the observation  
 71 and study of inhomogeneous phenomena. The use of penetrating radiation allows the inside of  
 72 studied specimens to be revealed, which is a great improvement over photography for the repre-  
 73 sentativeness of the measured field. The attenuation of penetrating radiation beam (of a given  
 74 energy) as it travels through matter can often be modelled using the Beer-Lambert law, here given  
 75 for a specimen made of one material:  
 76

$$I = I_0 e^{-\mu_m \rho x} \quad (1)$$

77 where  $I$  is the resulting beam intensity after interaction with the material,  $I_0$  is the reference  
 78 beam intensity without interaction (no sample),  $\mu_m$  is the mass attenuation coefficient ( $\mu_m = \mu/\rho$ ,  
 79 where  $\mu$  is the linear attenuation coefficient which is a material property),  $\rho$  is the density of the  
 80 material and  $x$  is the linear distance that the beam travels inside the specimen.

81 Furthermore, the use of penetrating radiation enables the use of tomographic techniques, which  
 82 give access to the attenuation field – a 3D “image” of  $\mu$ .

83 In this work we present full-field measurements of water imbibition into a clay-rock observed  
 84 with two highly complementary techniques using different types of penetrating radiation – x-ray  
 85 tomography and neutron radiography – both of which will be briefly described below.

### 86 2.1 X-ray tomography

87 X-rays are massless photons with high-energy that interact with the electron cloud surrounding  
 88 each atom’s nucleus. Therefore the probability of an x-ray photon being attenuated is proportional  
 89 to the atomic number  $Z$ , which itself is roughly proportional to the density of the material.

90 Tomography consists in sampling the unknown attenuation field within a specimen from a large  
 91 number of different orientations, which allows an accurate reconstruction of the desired attenuation  
 92 field using back-projection.

93 Tomography was developed with x-ray radiation, and x-ray tomography remains the most de-  
 94 veloped form of tomography with high-performance laboratory scanners and dedicated synchrotron  
 95 installations.

106 Within mechanics in general, and geomechanics in particular, it has become a *de-facto* standard  
107 for time-resolved 3D analysis. For example, [17] presents the *in-situ* deformation mechanisms in  
108 Callovo-Oxfordian clay-rock in triaxial compression using x-ray tomography with important results  
109 regarding strain localisation. The measurement of strain in this material is possible due to the  
110 natural inhomogeneities within the rock, that present a convenient pattern tracking with image  
111 correlation. The observed patterns reveal that the different components that make up the rock  
112 must have significantly different densities.

113 X-ray tomography (using a lab-based micro-focus scanner) will be used in this work to acquire  
114 a number of 3D x-ray attenuation fields during the imbibition process, allowing the deformation  
115 of the rock to be measured.

116 However, due to the relatively low density of water, this technique will have difficulty measuring  
117 the invasion of water into the sample.

## 108 2.2 Neutron radiography

109 Neutrons are neutral massive particles that interact directly with the nucleus of atoms. Neutrons  
110 generally have a much higher probability of interaction with light atoms like hydrogen, even though  
111 attenuation can be radically different between isotopes of the same atom (for example deuterium  
112 has a much lower attenuation than hydrogen).

113 Neutrons principally interact with nuclei by being scattered or absorbed; when a neutron is  
114 scattered its speed and direction change, whereas when it is absorbed a wide range of radiations  
115 can be emitted or fission can be induced in some specific elements. The nucleus may rearrange its  
116 internal structure and release one or more gamma rays, charged particles may also be emitted. The  
117 sensitivity of neutrons to hydrogen make it an ideal complement to x-rays, significantly facilitating  
118 the detection of water throughout the imbibition process. It is important to note that in the  
119 interaction of neutrons with water, the principal mechanism is elastic scattering.

120 To obtain low-noise radiographies at the pertinent resolution for this phenomenon, neutron  
121 imaging is slow (even compared to a lab-based x-ray scanner), and so it has been elected to  
122 perform neutron radiography for this study, since the time required for neutron tomography was  
123 estimated to be too long.

124 Since a 3D field of neutron attenuation will not be reconstructed, great care has been taken  
125 to fully characterise the interaction of neutrons with water in the experimental conditions used  
126 for this test. Figure 1 shows a key step in this procedure where a hollow aluminium wedge has  
127 been scanned with neutrons before and after filling with H<sub>2</sub>O, in order to fit the attenuation  
128 coefficient  $\mu_m$  of neutrons to water, net of the whole imaging setup. The left of the Figure presents  
129 a schematic showing the setup. It is important to note that due to scattering, in neutron imaging  
130 the sample is kept as close to the detector as possible. The middle of the Figure shows the recorded  
131 radiographies of  $I/I_0$  before and after filling the wedge with water. The change in attenuation of  
132 the beam between these two images is then extracted along the profile shown in violet in Figure  
133 1 middle-bottom. Together with the knowledge of the geometry of the wedge, this allows the  
134 relationship between the thickness of water in the direction of the beam ( $x$  in Beer-Lambert) and  
135 its attenuation ( $I/I_0$ ), as shown in violet in Figure 1 right-bottom. This experimental data is fitted  
136 with the Beer-Lambert law (equation 1) with  $\mu$  being the only free parameter. The fit is made in  
137 the range  $[0, 2.3]$ mm, that is to say until half of the “mean-free-path length” ( $\lambda$ ) of neutrons with  
138 wavelength of 1.8 Å in H<sub>2</sub>O (close to the peak of 3Å in the beam used), as given in [18]; the fit  
139 gives  $\mu = 0.310$ .

140 It is immediately apparent that for more than 3mm of H<sub>2</sub>O there is a significant deviation  
141 between the fitted Beer-Lambert absorption and the measured attenuation data, this is likely due  
142 to scattering of the neutron beam. For short path lengths in the water (well below the mean-path-  
143 length of 4.6mm), the attenuation of the beam is low, therefore there is a significant amount of  
144 transmission ( $I/I_0$ ). The non-transmitted beam is scattered and detected elsewhere, but since this  
145 is a small amount, the relationship between distance in water and attenuation is modelled well by  
146 Beer-Lambert. For longer path lengths, for example 10mm, a 5% of the beam is expected to be  
147 transmitted. This implies that almost 95% of the beam will be scattered, and the scattered beam  
148 itself will be scattered again in a phenomenon known as multiple scattering. Some of this diffused  
149 beam will be detected at the position of the transmitted beam, thus increasing the measured flux to  
150 19%. The effect of scattering is clearly visible in Figure 1 right-top, where the measured intensity  
151 in red green and blue zones *increases* when water is added.

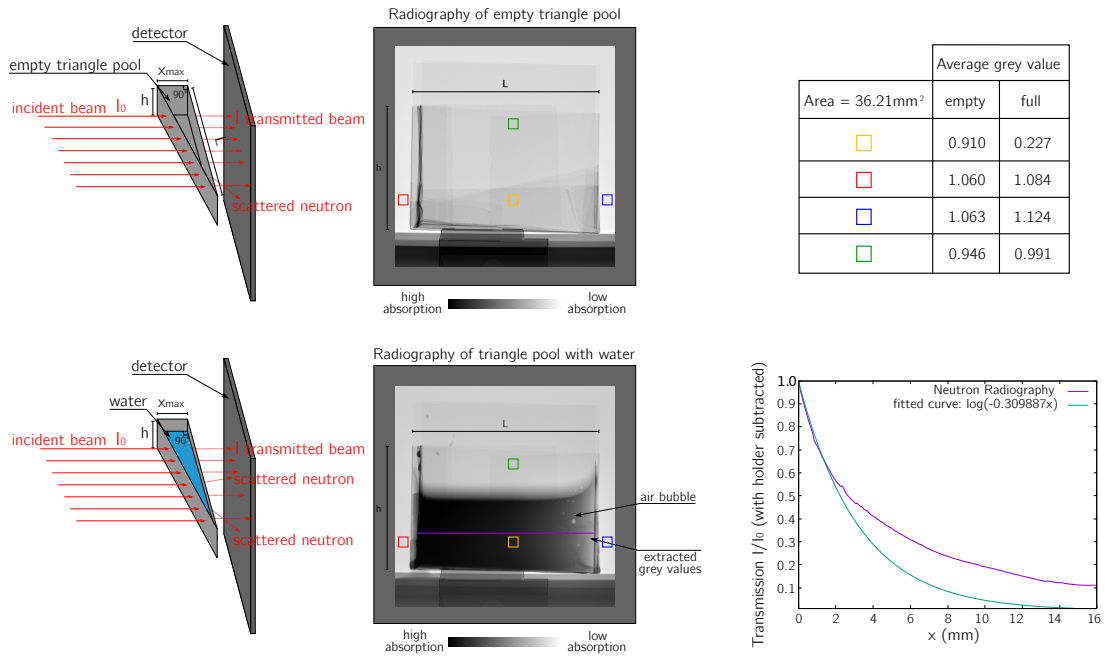


Figure 1: Calibration of the beam absorption in  $H_2O$  with neutron path length, on the left, experimental schematic, in the middle experimental data, top-right grey value in some zones of interest, bottom-right the fit of the Beer-Lambert law to the acquired data

152 We therefore have a good degree of confidence in the Beer-Lambert law for up to 3mm of water  
 153 in the direction of the beam.

### 154 3 Experimental Campaign

155 During the preparation of concrete/ $CO_x$  interface specimens for mechanical testing, fresh concrete  
 156 is poured directly onto the free surface of an unconfined cylinder of  $CO_x$  (78mm in diameter, 50mm  
 157 height), causing cracking within one day.

158 This phenomenon was studied in a preliminary study using time-resolved x-ray tomography as  
 159 shown in Figure 2 a); cracks visible after 1h are highlighted in red in the vertical slices through the  
 160 x-ray tomography volumes middle and bottom. The free-water in the concrete was suspected as  
 161 the root cause of the cracking in the  $CO_x$ . In order to confirm the availability of free water as the  
 162 main mechanism for cracking a second experiment (Figure 2 b) was carried out, where the concrete  
 163 was replaced by tap water provided to the top part of a  $CO_x$  specimen of the same dimensions  
 164 as a). Again, cracks after one hour are highlighted in red, visually confirming the repeatability of  
 165 the phenomenon. In both experiments cracking is clearly visible, and continues to an impressive  
 166 degree after an hour, however in both cases the entry of water into the specimen is not visible,  
 167 weakening the conclusions and hampering further interpretation.

168 Neutron imaging was selected as an ideal complement to x-ray tomography for the detection  
 169 of water entry. Given the limitations of neutron radiography discussed above, prismatic specimens  
 170 with one small dimension are clearly convenient.

171 Given the change of geometry of the specimen, experiment b) was repeated on prismatic speci-  
 172 men as shown in Figure 2 c). Finally, in order to avoid possible lateral leakage, water was supplied  
 173 from below from a saturated sponge Figure 2 d). Again in these prismatic specimens, after an  
 174 hour of water supply, cracking is clearly visible in correspondence with previous experiments. Due  
 175 to one smaller dimension, the rate of cracking also appears higher. Experiment d) was therefore  
 176 selected to be performed under x-ray tomography and neutron radiography.

177 For the selected imbibition experiment, a fresh and presumably saturated cylindrical core –

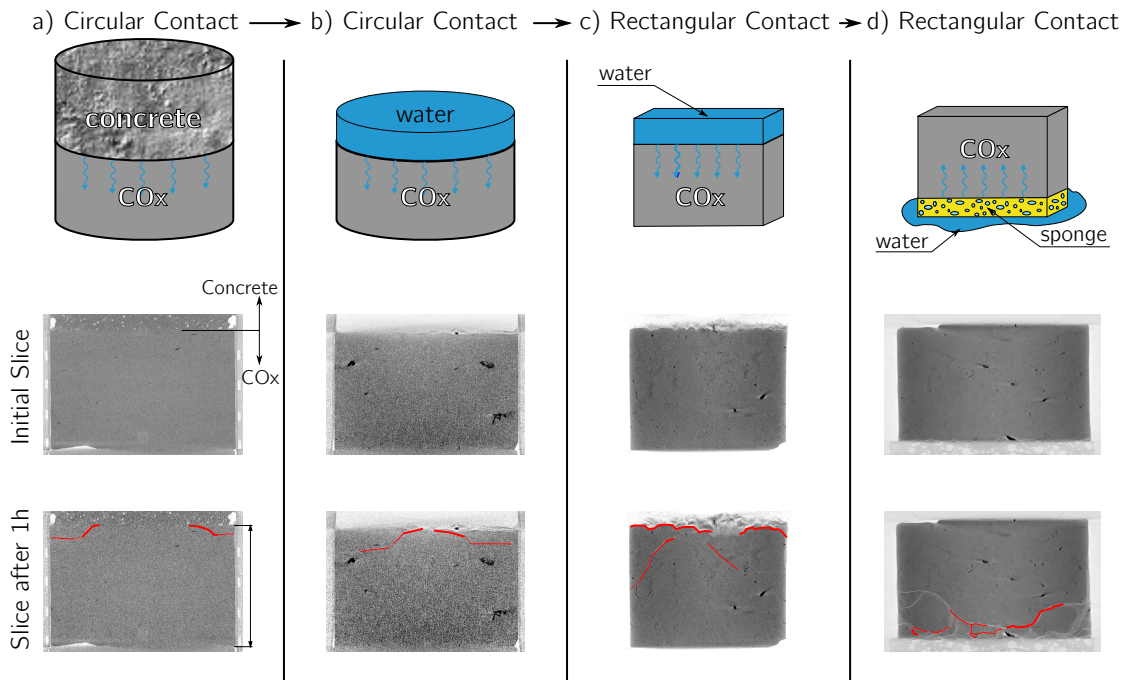


Figure 2: Illustration of the different steps of simplification which led to neutron scanning, with top: a sketch, middle: a vertical slice through an x-ray scan of an undeformed specimen, bottom: same specimen after an hour

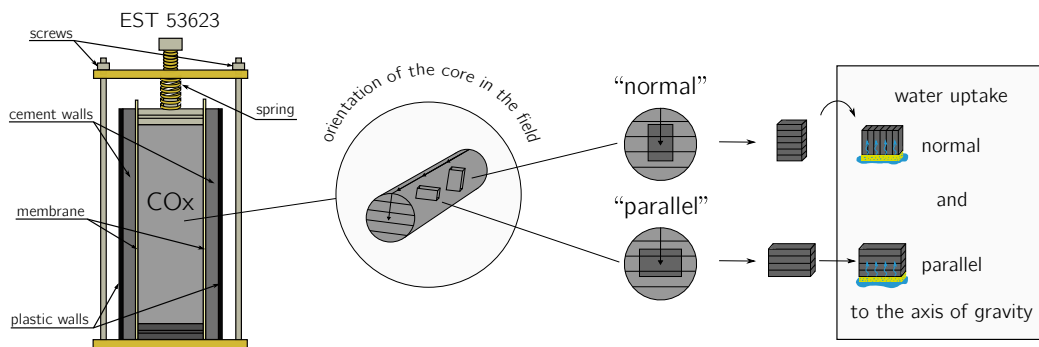


Figure 3: Description of the preparation of specimens from the COx core EST 53623 from Andra. From left to right: sealed COx core, laterally with a cement and a plastic wall and vertically pressurised with a compressed spring; extracted COx core; orientation of extraction of the two types of specimens, normal and parallel

178 EST 53623 – was opened. The long axis of the extracted core is normal to the axis of gravity.  
 179 Four prismatic specimens, two normal and two parallel to the reported *in-situ* horizontal plane,  
 180 were immediately cut with a diamond wire saw and rectified on all six faces with a belt sander  
 181 (see Figure 3). Massive offcuts were used to measure the initial water content, giving an initial  
 182 water content of 6.8% which is within the statistical range of water contents for saturation for this  
 183 material, as reported in [19].

184 Four practically identical imbibition tests were performed independently under either neutron  
 185 radiography or x-ray tomography – although in the future both imaging modalities will be available  
 186 on the NeXT instrument. Samples destined for neutron radiography were scanned with x-ray  
 187 tomography before imbibition.

188 For x-ray tomography, the scanner in Laboratoire 3SR is used. The scanner has a micro-focus  
 189 source with Tungsten target, the acceleration of electrons into the target is set at 100 kV, and  
 190 the resulting beam is not filtered. An amorphous silicon detector with CsI scintillator is used

Specimen Name	Dimensions (mm)	Mass (g)	Time to experiment after cutting (h)
Parallel-X	40.98 × 32.60 × 14.50	45.29	3
Parallel-N	44.35 × 32.02 × 14.51	48.24	8½
Normal-X	34.85 × 31.92 × 15.04	38.88	5½
Normal-N	36.20 × 32.03 × 14.13	38.65	6½

Table 1: Table describing the specimens tested

191 in 2x2 binning and given the non-cylindrical shape of the specimen 1200 projections (more than  
192 strictly necessary) are taken. The geometrical pixel size is 70  $\mu\text{m}/\text{px}$  and the total scan time for  
193 one tomography is 5 min. Reconstructions are performed with the XAct software provided by  
194 RX-Solutions (Annecy, France), with appropriate beam-hardening corrections applied.

195 For neutron radiography the NeXT instrument on the D50 beamline at the Institut Laue-  
196 Langevin in Grenoble is used. The neutron beam has wavelengths [1,10]  $\text{\AA}$  with a peak at 3  $\text{\AA}$ ,  
197 making this a cold-neutron beam. For geometrical sharpness a 15 mm pinhole is used at around  
198 10m distance to the detector (giving a practically parallel beam) and a 5 mm thick graphite diffuser  
199 is used to homogenise the beam. A 50  $\mu\text{m}$  thick LiF scintillator is used and a 2048x2048 S-CMoS  
200 camera is used in 2x2 binning giving a pixel size of around 55  $\mu\text{m}/\text{px}$ . Ten second exposure is  
201 chosen used for each radiography as a compromise between image contrast and process speed.  
202 In all that follows radiographies are corrected by  $I_0$ , by the darkfield of the camera, gamma-ray  
203 streaks (which leave high-valued pixels in some radiographs) are removed by searching for high  
204 outliers, and radiographs are averaged with a median filter in groups of six, giving a total exposure  
205 time of 1min per corrected radiography.

206 Regardless of the modality, each COx sample was held from the top with double-sided sticky  
207 tape. The bottom of the sample rests on a commercial kitchen sponge, cut to shape for the  
208 experiment, which was saturated rapidly with room-temperature tap water.

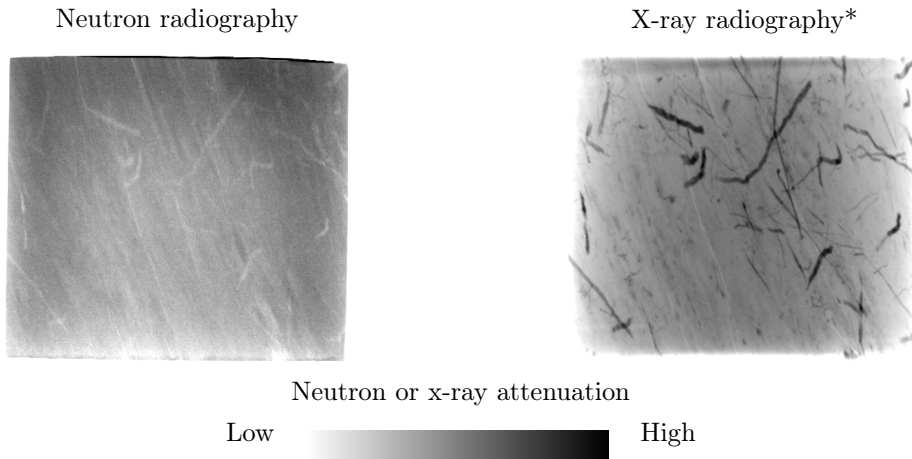


Figure 4: Neutron and x-ray radiography of sample Normal-N through the thinnest direction with holders masked from the image before water imbibition.

\*Since the x-ray data is collected with a divergent beam which gives different magnification through the depth of the specimen, the reconstructed 3D volume is virtually projected in parallel geometry in the same direction as the neutron radiography to obtain easier-to-compare attenuation images

209 Previous use of both modalities on geomaterials, for example by [20] show the utter com-  
210 plementarity of these two imaging techniques for the investigation of sand is a state of partial  
211 water-saturation. Figure 4 shows a comparison of the attenuation of the neutron and x-ray beams  
212 used in this work of sample “Normal-N” before imbibition. The inclusions in the clay matrix clearly  
213 attenuate more x-rays than the matrix, whereas they attenuate fewer neutrons than the matrix.

## 214 4 Acquired data and processing

215 Figure 5 presents a number of neutron radiographies for the “-N” samples and central vertical slices  
216 through the reconstructed x-ray tomography volumes for the “-X” samples. It is important to note  
217 that averaging together of the neutron radiographies over 5 min offers significant denoising, but in  
218 intervals where there is significant water movement can blur the radiography.

### 219 4.1 X-ray tomographies

220 In the vertical slices presented in Figure 5, x-ray tomography is clearly able to follow the deforma-  
221 tion of the solid COx, but is not sensitive to water. Furthermore, in both “parallel” and “normal”  
222 samples, some pre-existing cracks are visible.

223 In the 3D+t x-ray tomography timeseries acquired for samples Normal-X and Parallel-X during  
224 imbibition, a mask for the bounding surface of the sample is created in each time step. The objective  
225 of this mask is to define the “external” volume of the sample by filling cracks and removing the  
226 sample holder. An initial mask is obtained by applying a greyscale threshold on the reconstructed  
227 16bit images of [15400,65535] – see Figure 6 for the meaning of the greyvalues with respect to the  
228 histogram of reconstructed greyvalues. Cracks are filled by four cycles of morphological dilation of  
229 the selected solid phase with a radius of 3 pixels and four cycles of erosion; the number of cycles  
230 is selected as half of the maximum crack opening in pixels plus 1 pixel margin. The measure of  
231 volume of the mask in every timestep gives the outside volume of the deforming sample.

232 The detection of large cracks *i.e.*, larger than 2 pixels can simply be done with a threshold.  
233 However, subpixel crack detection is more sensitive to noise and the beam hardening effect which are  
234 present in the images. A simple program was developed in Python using Numpy and Scipy[21, 22]  
235 for the detection of subpixel cracks: for any pixel within the mask described above, if its value is  
236 less than 12000 is it considered a void. For values higher than this threshold, they are considered  
237 cracks if their value is 2200 units lower than the local mean, which is itself calculated as a moving  
238 linear window  $\pm 9$  pixels (*i.e.*, 1.33 mm) centred on the pixel of interest, avoiding values lower than  
239 12000 (cracks) and higher than 26000 (dense inclusions). Since the boundary of the specimen is  
240 detected as a crack, the first two pixels on the boundary of the identified crack map are finally  
241 cropped (by masking with a twice-eroded overall sample mask).

### 242 4.2 Neutron radiographies

243 Neutron radiographies clearly reveal the entrance of water into the COx matrix. The principle  
244 for the analysis of the water entering the COx is based on the reasoning that the attenuation  
245 coefficient  $\mu$  of the COx in its initial state of saturation will not change if the sample swells at  
246 constant mass (for example if it expands without losing or gaining water) – the mass attenuation  
247 coefficient  $\mu_m = \mu/\rho$  will however change to reflect a decrease in density. Thus in a radiography  
248 of a sample swelling in all directions at constant mass, the swelling in the direction of the beam  
249 does not change the overall attenuation of the beam (changes in size in the direction of the beam  
250 are undetectable since increases in volume – and thus the length for the beam to cross – are can-  
251 celled out by the reduction in density), whereas swelling in the imaging plane will reduce the mass  
252 attenuation coefficient allowing more of the beam to cross the sample. If the increase in projected  
253 area in the detected radiography of the sample is taken into account, it is easy to show that:

$$254 \quad Area_{initial} \times \mu_{initial} = Area_{swelled} \times \mu_{swelled} \quad (2)$$

255 Thus, at constant mass, for any swelling or contracting:

$$256 \quad Area \times \mu = const. \quad (3)$$

257 It is important to note that since the reference  $I_0$  images are acquired in air, when air invades  
258 the specimen, the projected area to be used is everything within the boundary of the specimen.

259 We are obviously not in a case of swelling at constant mass, since water invades the specimen,  
260 thus any departure from the above reasoning is assumed to be due to the entry of water. To this  
261 end, throughout the neutron radiographies acquired during water imbibition, the projected area of  
262 the specimen is carefully selected in the images using a mask for each radiography. Starting from



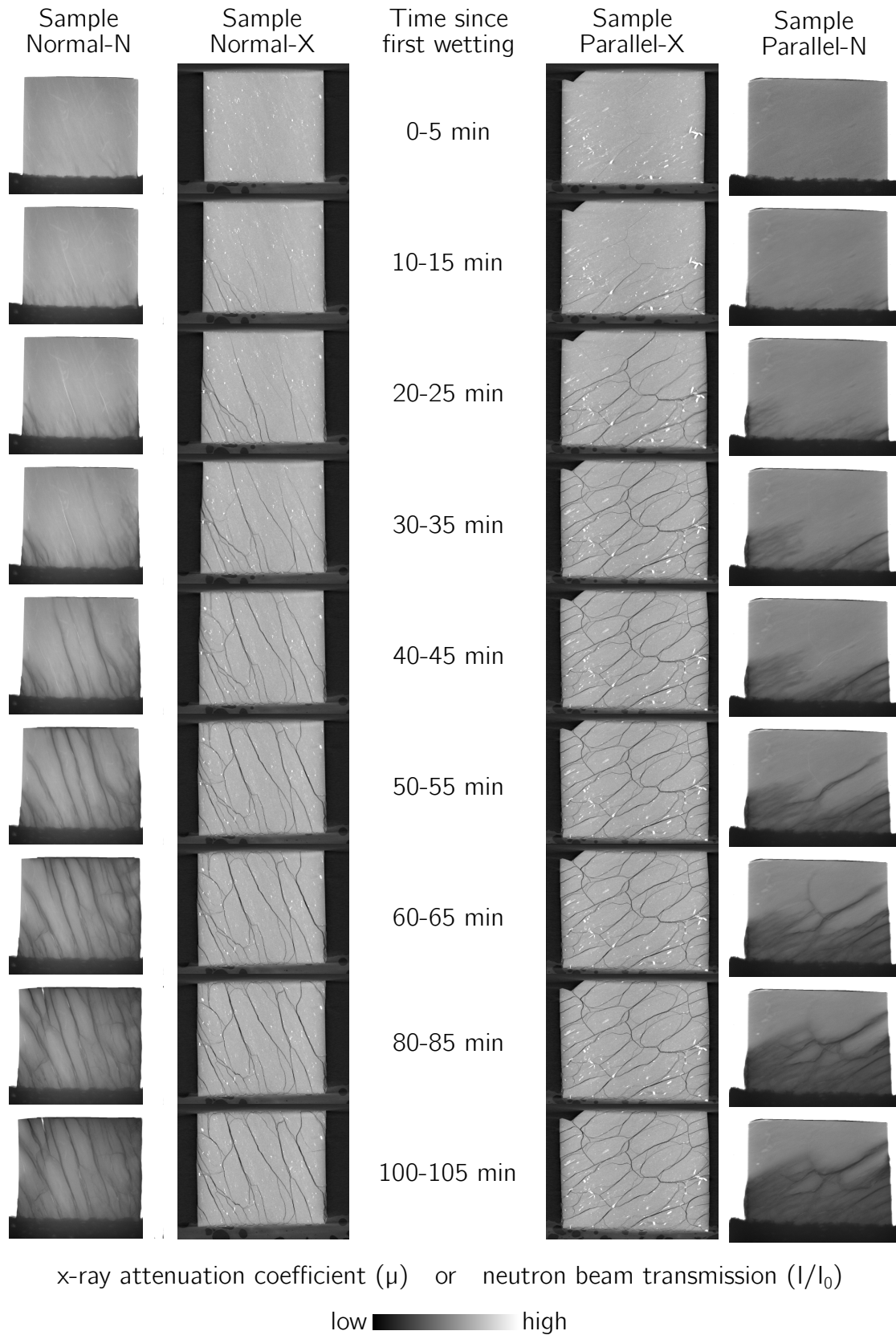


Figure 5: Selected *in-situ* images acquired during imbibition for the four samples in Table 1. X-ray data is presented as a vertical slice in the middle of the specimen, whereas neutron radiographies are averaged together in 5-minute intervals in order to match the x-ray tomography times. The structure holding the sample from the top has been manually erased from neutron radiographies. Accuracy on times is  $\pm 90$ s

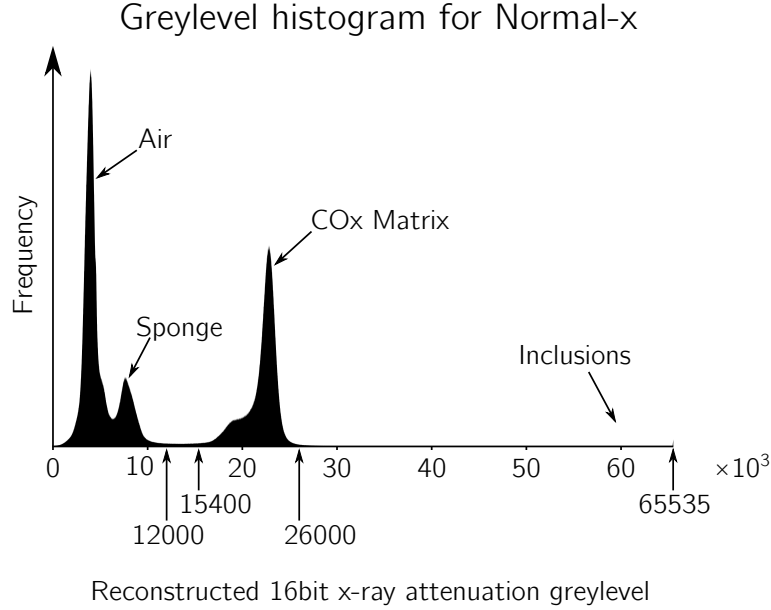


Figure 6: Greylevel histogram of the reconstructed x-ray tomographies

263 the initial dry image, the expected average attenuation in each subsequent radiograph is calculated:  
 264

$$\mu_{current} = \frac{\mu_{initial}}{Area_{initial}} \times Area_{final} \quad (4)$$

265 This is compared to the measured mean attenuation value, and any difference is ascribed to the  
 266 entrance of water into the specimen. The change in attenuation can thus be directly linked to a  
 267 mean “thickness” of water entering the masked area using the fit in Figure 1. The mean thickness  
 268 of water over the projected area of the specimen thus gives a relatively direct measurement of  
 269 water volume entering the specimen with one minute resolution.

## 270 5 Sample-scale measurements

271 In this section, measurements at the scale of the sample are presented. These measurements use the  
 272 analysis of the acquired x-ray tomography and neutron radiography data detailed in the previous  
 273 section in an average sense.

274 Figure 7 shows the evolution of sample-wide quantities that can be obtained: x-ray tomography  
 275 data is used to measure the change in volume of the external boundary of the specimen (using  
 276 the mask) and the volume of visible cracks using the method described above, and neutron radiog-  
 277 raphies are used to measure water entering the specimen. The evolution of these three volumes  
 278 is shown from the beginning of imbibition process in Figure 7 for both types of sample: Parallel  
 279 (squares) and Normal (circles), normalising each by the initial volume of the sample as obtained  
 280 from x-ray tomography. It is important to note that the sub-pixel sensitivity of the crack detection  
 281 is expected to significantly overestimate crack volume for smaller-than-pixel crack openings. A  
 282 small volume of cracks is detected even at the beginning of the test, which on close inspection of  
 283 the x-ray tomography images makes sense, since some hairline cracks are present in the samples  
 284 before imbibition, presumably due to the stress release with respect to site conditions.

285 The measurement of the change in water volume inside the boundary of the specimen is obtained  
 286 from the analysis of the neutron radiographies. The change in attenuation of the specimen (taking  
 287 into account its change of area/volume) is assumed to be entirely ascribable to water entering the  
 288 specimen, and is converted to a volume using the fit to Beer-Lambert in Figure 1. The maximum  
 289 increase of attenuation measured (396 min after imbibition for Parallel-N) corresponds to a path  
 290 length of neutrons in water of 2.13 mm which is just within the area of confidence of the fit. This  
 291 measurement cannot be verified directly due to experimental difficulties (the sample falls apart  
 292 after 2h imbibition and in neutron radiography the sample becomes radioactive and cannot be

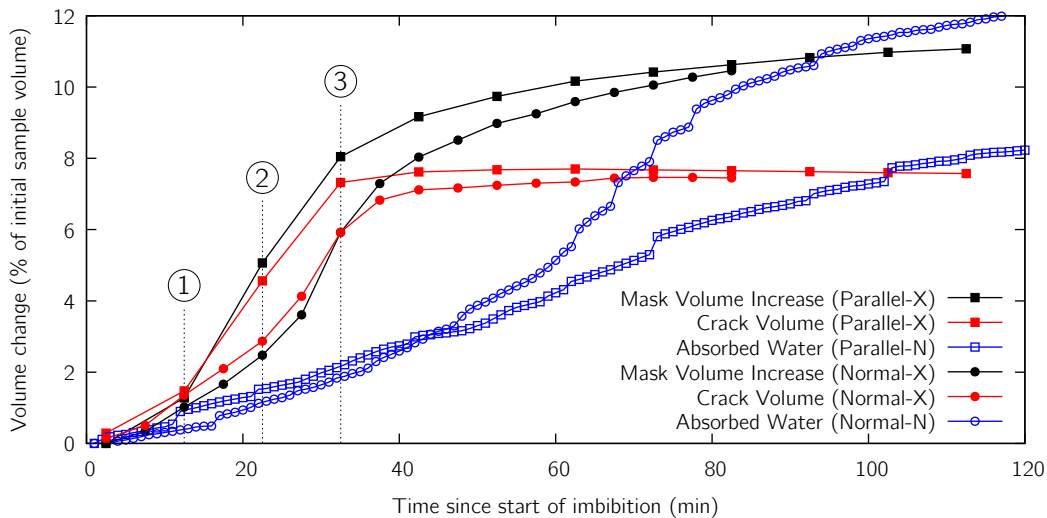


Figure 7: Macroscopic measurements of sample evolution from x-ray tomography (mask and crack volumes) and Neutron radiography (water volume). It is important to note that for the x-ray scan the time is taken as the middle of the 5-minute scan, and that Normal and Parallel samples are not identical. Accuracy on times is  $\pm 90$ s

293 handled immediately) but the measured volumes correspond satisfactorily to the low-quality mass  
 294 measurements after x-ray tomography tests.

295 The upshot of Figure 7 is that the imbibition process causes a rapid change of volume (for  
 296 both directions of exposed surfaces) in the COx samples tested, with the peak rate between 10 and  
 297 40 minutes. In this early stage, it appears that (again for both tested orientations) most of the  
 298 sample's increase in volume is due to large cracks that appear and develop. In both samples tested,  
 299 after about 50 minutes crack volumes then become relatively constant in the observed time. The  
 300 water, which causes the cracks, steadily enters the sample at a significantly lower rate than the  
 301 crack propagation, however continues to do so after the cracks have stopped increasing in volume,  
 302 thus the phase starting from 50 min onwards appears to be one of water uptake into the matrix  
 303 without cracking *at the scale observed*.

## 304 6 Local measurements

### 305 6.1 Local observations

306 Full-field imaging also allows local processes within the sample to be followed. Figure 8, for example  
 307 shows the relative success of the crack identification algorithm presented above in detecting small  
 308 cracks in three selected x-ray tomography images, representing points of significant cracking activity  
 309 (marked as 1,2,3 in Figure 7). For both types of samples, a central, vertical slice through the original  
 310 greyscale volumes is presented, alongside the same slice through the detected crack field, as well  
 311 as a 3D rendering of the entire detected crack field.

312 The success of the crack detection algorithm means that, for example, the structure of the  
 313 cracks (size and number of connections) could be analysed with ease.

314 Another local measurement of key importance is the interplay between cracking and water  
 315 distribution in the sample. The sample-scale comparisons presented in Figure 7 seem to indicate a  
 316 good correspondence between x-ray and neutron measurements, however since (slightly) different  
 317 samples are followed with x-rays and neutrons, a direct *local* comparison cannot be made. The  
 318 cracking that can clearly be seen in x-rays can also be seen with neutron radiography (although in  
 319 this case its volume cannot be measured directly).

320 Figure 9 presents neutron radiographies for sample Normal-N with a higher time resolution  
 321 (and therefore more noise) than Figure 5 with two zooms into areas of the sample: the larger  
 322 area (top line) presents minute-averaged radiographies showing the development of three cracks  
 323 (A, B and C) in the middle of the sample, whereas the smaller area (bottom line) shows ten-

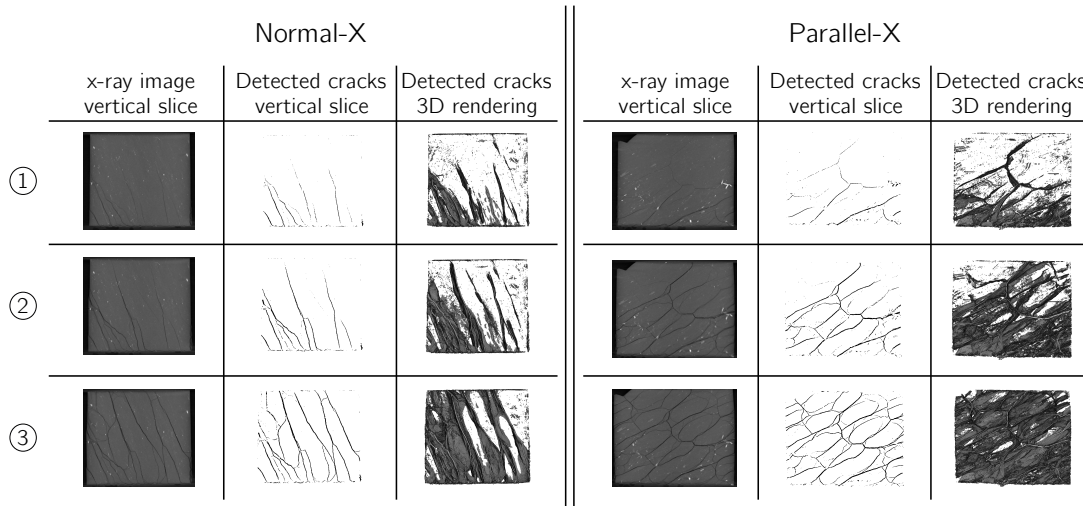


Figure 8: Crack state for the two types of samples, during the three time-steps indicated in Figure 7, when the the crack propagation is rapid

324 second radiographies of crack C. The top line of Figure 9 shows some interesting features of crack  
 325 development with the opening of a dry crack, which is then filled with water from both sides  
 326 (clearly indicating a structural effect of connected water in the specimen) for crack A, followed by  
 327 B and C. The bottom line of Figure 9 aims to present the kinematics of crack opening, followed  
 328 by what must be capillary rise. This relatively simple conclusion would have been very difficult to  
 329 be certain about with x-ray tomography due to the low contrast that water presents with respect  
 330 to air.

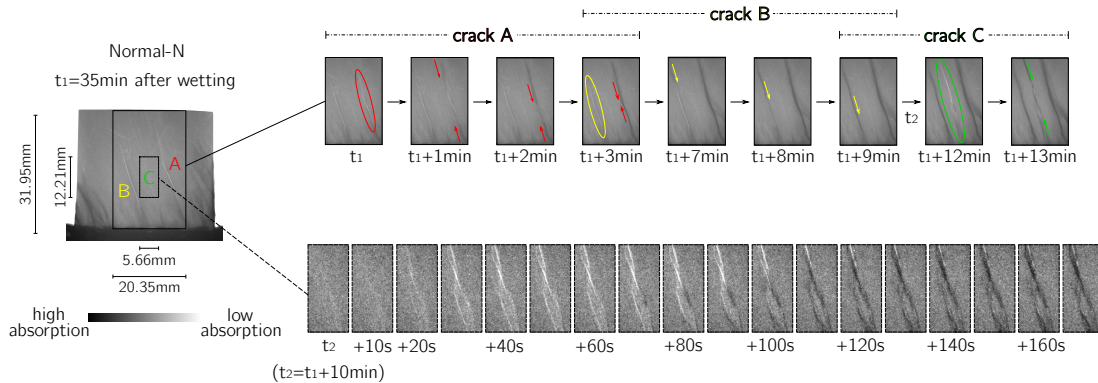


Figure 9: Neutron imaging observation of the opening of three different cracks (A, B, C) in time and demonstration of the sequence of the phenomena: mechanical cracking followed by water penetration for the Normal-N sample. Upper row: Sequence of the three different cracks and water penetration in time. Lower row: Sequence of crack C opening and water penetration in time

331 Figure 10 also presents neutron radiographies of cracks opening followed by water invasion,  
 332 however for the sake of an illustrative example, an inset central vertical slice from x-ray tomography  
 333 is presented where a clear air-water front can be seen within an open crack. Given the size and the  
 334 contrast of the x-ray inset, it is once more obvious that neutrons allow a much clearer observation  
 335 of the water front.

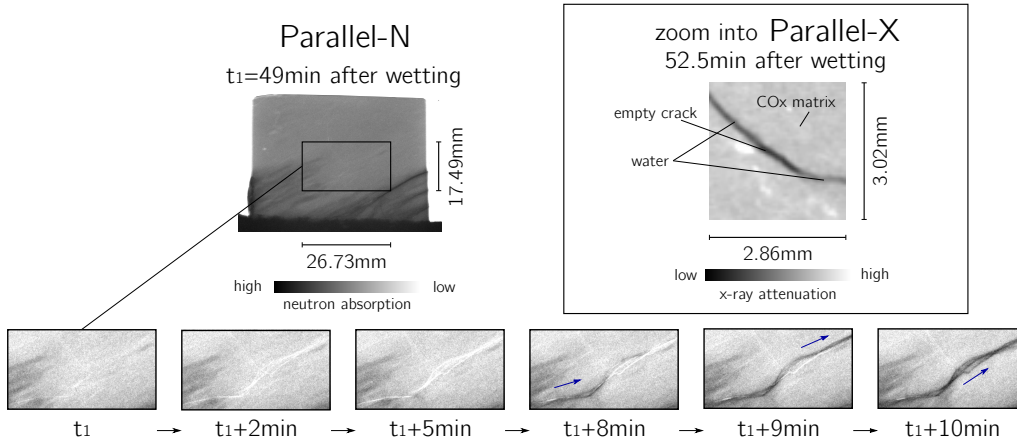


Figure 10: Neutron imaging observation of crack opening and water penetration for the Parallel-N sample. Inset vertical slice from Parallel-X sample

## 336 6.2 Digital Volume Correlation

337 The time-resolved x-ray tomographies, of which vertical slices are shown in Figure 5, are ideally  
 338 suited to following the solid material's evolution as the specimen deforms. The identification of  
 339 cracks and the specimen's external volume have been presented above, however a finer measurement  
 340 of material evolution using Digital Volume Correlation (DVC) is also possible to the fine texture  
 341 present in the images. Here we use a new development library called RANDOMWAFFLE. In this case,  
 342 a regular mesh of nodes spaced 18 pixels (*i.e.*,  $70 \mu\text{m} \times 18 = 1.26 \text{ mm}$ ) apart defines measurement  
 343 nodes. For each node, a representative sub-volume centred on its 3D position is extracted from the  
 344 3D volume which represents the reference configuration. The DVC code then attempts to match  
 345 this reference sub-volume in the next 3D volume. Subpixel accuracy is obtained with 3rd order  
 346 image interpolation, and a transformation operator is obtained, mapping each 3D point into the  
 347 next image.

348 Given the significant disturbance created by the cracks, DVC is performed *incrementally* be-  
 349 tween pairs of images. Strains are calculated using TOMOWARP2[23] using a large strain hy-  
 350 pothesis, and using only the displacements from DVC. The increments of the trace of the locally-  
 351 calculated strain tensor are presented in Figures 11 and 12, and referred to as volumetric strain.  
 352 It is important to note that during cracking, the transformation operator is obviously unable to  
 353 capture this behaviour, causing large apparent strains in the cracks.

354 Figure 11 illustrates vertical slices through the field of incremental volumetric strain for the  
 355 "parallel" sample. The disturbance due to cracking is evident, presenting saturated values of strain,  
 356 which in this experiment is most striking between the second and third increments. However, before  
 357 and after cracking, swelling is clearly measurable within the sample.

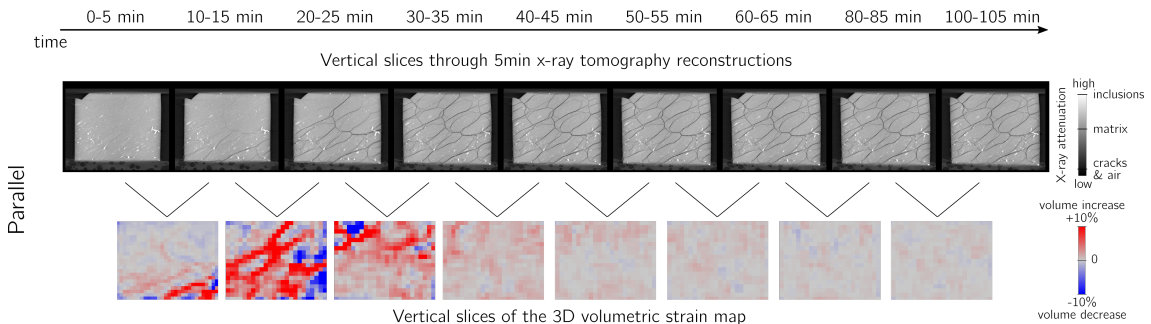


Figure 11: Series of vertical slices with time (parallel orientation). Top: x-ray tomography reconstructed images, Bottom: incremental volumetric strain maps (from incremental DVC)

358 Figure 12 also shows x-ray images and incremental volumetric strain maps, and adds the corre-  
 359 sponding neutron radiography images. It is important to note that the samples are similar but not  
 360 identical. This being said there is a good correspondence between neutron and x-ray datasets for  
 361 this test. Figure 12 presents an important perspective for further exploration of the direct mea-  
 362 surement of hydro-mechanical coupling in this material: the combination of techniques presented  
 363 allows a change of solid volume to be related to water entry. When this “multimodal” scanning  
 364 is available simultaneously for the same specimen, this will allow the important measurement of  
 365 the local change in water content, by updating the local value of solid density from the volumetric  
 366 strain.

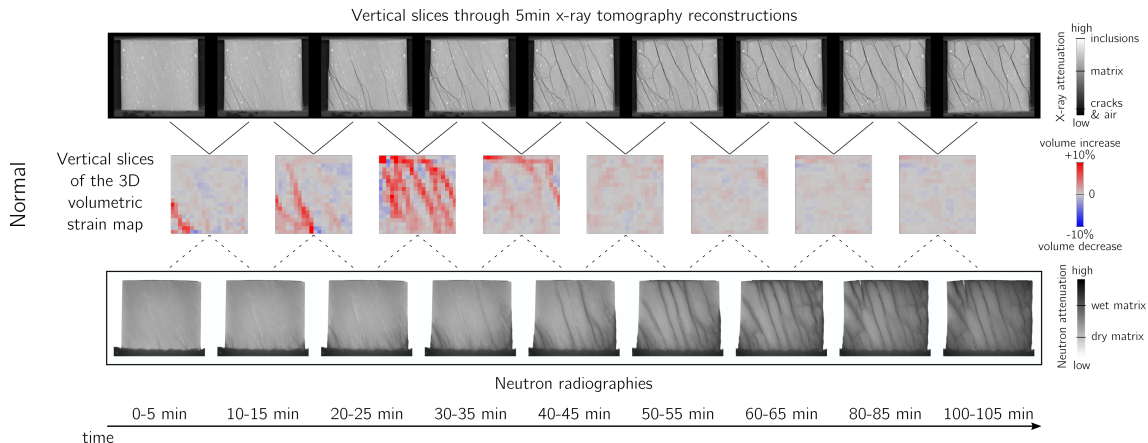


Figure 12: Series of vertical slices with time (parallel orientation). Top: x-ray tomography reconstructed images, Middle: incremental volumetric strain maps (from incremental DVC), Bottom: Neutron radiographies of similar sample

## 367 7 Discussion

368 The observed processes can be explained purely as a hydro-mechanical interaction. The samples  
 369 studied – as well as those prepared for testing in the SInC box – are almost certainly in a state  
 370 of partial saturation due to both water losses (evaporation during sample preparation in the lab,  
 371 and possibly during transport) as well as an increase of volume of the soil skeleton due to elastic  
 372 rebound due to unloading from the site stress condition. As reported in [24], the suction stresses  
 373 generated in COx even for small changes of water content are significant, for example 150 MPa of  
 374 suction is required to dry their samples from 6% water content to 2.5%. The suction necessarily  
 375 present in the specimens studied here applies an internal effective stress, which holds the sample  
 376 together despite the presence of cracks (from the tunnel’s EDZ and/or induced by coring and sample  
 377 preparation). Thus, when an unconfined specimen is placed in rapid contact with an abundance of  
 378 free water, natural capillary rise leads to a local breakdown of suction. The breakdown of suction  
 379 locally decreases the effective stress and leads to swelling. This differential swelling allows existing  
 380 cracks to open, and potentially new ones to form, with an preferred orientation depending on the  
 381 clay-rock’s orientation in the bedding plane.

382 As is clearly shown throughout the measurements presented in this work, a large degree of  
 383 cracking occurs when the COx samples are put into contact with water. Figures 9 and 10 are  
 384 strong evidence for the propagation of cracks in the samples before the arrival of water around  
 385 the crack. This is consistent with the hypothesis that suction is keeping the sample together, and  
 386 as soon as it is removed by the presence of free water, effective stress falls dramatically and the  
 387 sample disintegrates.

388 The macroscopic volume evolutions presented in Figure 7 show, for both sets of samples studied  
 389 with *in-situ* x-ray tomography, a rapid increase of the external volume initially due to cracks that  
 390 are detectable at the resolution used. The crack volume detectable at this scale stabilises between  
 391 7 and 8% for both sample orientations, whereas the external volume continues to increase due to  
 392 swelling or cracks below the resolution. The water volume arriving into similar samples studied

393 with neutron radiography appears to exceed the total volume increase of the sample, which is  
394 perfectly consistent with a state of partial saturation in the initial sample. This final conclusion  
395 however is weak, due to different samples being studied.

## 396 8 Conclusions and perspectives

397 This paper presents some of the sample-scale and local-scale measurements that can be obtained on  
398 the Callovo Oxfordian clay-rock with the highly complementary contrast mechanisms of x-ray and  
399 neutron beams. The uptake of liquid water in unconfined COx specimens has been studied, allowing  
400 a direct measurement of the rate of sample volume increase (both globally and locally), crack  
401 growth and water uptake. Even though the unconfined testing conditions are not representative  
402 of the *in-situ* under stress state, the cracking response of the clay-rock appears to be governed by  
403 hydro-mechanical interactions. The cracking observed in the preparation of samples to simulate  
404 the clay-rock/concrete interface, is therefore interpreted as a breakdown of suction (and therefore  
405 effective stress) due to water penetration.

406 The principal crack orientation of the “normal” samples appears to be perpendicular to the  
407 cracks of the “parallel” samples, revealing the anisotropy of the Callovo Oxfordian clay-rock. A  
408 favourable cracking orientation could be suspected for the “parallel” samples, more results are  
409 however necessary for a clearer understanding.

410 The measurements resulting from the combination of the two imaging techniques reveal a rapid  
411 crack opening and propagation in the COx samples within 30-40 minutes of contact with free water,  
412 which then appears to stabilise. The water penetrates the samples significantly more slowly, but  
413 steadily, reaching the samples’ increased volume (filling open cracks) after around 120 min.

414 With the truly combined simultaneous neutron and x-ray tomography setup expected to come  
415 online on the NeXT instrument on the D50 beamline at the ILL in early 2018, a full coupling  
416 between measurements *of the same sample* can be done, allowing for example, the comparison of  
417 water uptake (neutrons) to volumetric strain obtained with DVC on the x-ray tomography volumes.

418 A clear path for this combination of tools is to look at this material (ideal for neutrons since  
419 its hydraulic behaviour is key, all while having low water content *in-situ*) under stresses of interest  
420 for the site.

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