



1 **X-ray Computed Tomography Investigation of Structures in Opalinus Clay from**  
2 **Large Scale to Small Scale after Mechanical Testing**

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10

11 **ABSTRACT**

12 In the past years X-ray Computed Tomography (CT) has become more and more common  
13 for geoscientific applications and is used from the  $\mu\text{m}$ -scale (e.g. for investigations of micro-  
14 fossils or pore scale structures) up to the dm-scale (full drill cores or soil columns). In this  
15 paper we present results from CT imaging and mineralogical investigations of an Opalinus  
16 Clay core on different scales and different regions of interest, emphasizing especially upon  
17 the 3D evaluation and distribution of cracks and their impact upon mechanical testing of such  
18 material. Enhanced knowledge of the testing behavior of the Opalinus Clay is of great  
19 interest, especially since this material is considered for a long term radioactive waste  
20 disposal and storage facility in Switzerland. Hence, results are compared regarding the  
21 mineral (i.e. phase) contrast resolution, the spatial resolution, and the overall scanning  
22 speed.

23 With this extensive interdisciplinary top-down approach it has been possible to characterize  
24 the general fracture propagation in comparison to mineralogical and textural features of the  
25 Opalinus Clay. Additionally, and as far as we know, a so called mylonitic zone, located at the  
26 intersect of two main fractures, has been observed for the first time for an experimentally  
27 deformed Opalinus sample. The multi-scale results are in good accordance to data from  
28 naturally deformed Opalinus Clay samples, which enables to perform systematical research  
29 under controlled laboratory conditions. Accompanying 3D imaging greatly enhances the  
30 capability of data interpretation and assessment of such a material.

31

32 **Key words:** Claystone,  $\mu$ -CT, Opalinus Clay, Mechanical Testing, HLRW Research

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## 34 INTRODUCTION

35 In the past years X-ray Computed Tomography (CT) has become more and more common  
36 for geoscientific applications and is used from the  $\mu\text{m}$ -scale (e.g. for investigations of micro-  
37 fossils or pore scale structures; e.g. Schmitt et al, 2016, this issue) up to the dm-scale (full  
38 drill cores or soil columns; e.g. Schlüter et al, 2015, this issue). Consequently, benchtop CT  
39 equipment for material and geoscience were developed and are now frequently used  
40 because almost all geoscientific samples show 3D features which would be missed when  
41 analyzing 2D sections only (e.g. by classical microscopy). These features are for example  
42 the abundance of minerals, location of particular particles towards bedding (or texture in  
43 general), pore system, cracks, and veins. The 3D distribution of all these features can be  
44 extracted and used for a variety of numerical modeling purposes (Andrä et al., 2013).  
45 However, due to the resolution of  $\mu\text{-CT}$  devices in range of a few  $\mu\text{m}$ , it is particularly suitable  
46 to study sandstones or other rocks with large particles and less suitable for the  
47 characterization of clays. Claystones, per definition, feature grain sizes below the common  
48 CT resolution (in range of 1 – 2  $\mu\text{m}$ ) and also grain densities, i.e. absorption characteristics,  
49 which result in very challenging segmentation procedures. Although not all features can be  
50 resolved,  $\mu\text{-CT}$  was extensively used to improve the understanding of clays in sediments (oil  
51 industry), in soil science, and as barrier functions in repository systems for high-level  
52 radioactive waste (HLRW). The oil industry is particularly interested in porosity, permeability,  
53 and fluid flow in general. An overview of CT application in the oil industry and for soils is  
54 provided by Heijs et al. (1995) and Akin & Kovscek (2003). By using a medical CT, Ashi et al.  
55 (1997) analyzed texture and density of marine clays, whereas Yang et al. (2010) used CT  
56 data to support logging operations. For soils, Naveed et al. (2012) used CT to investigate the  
57 importance of macropores for the convective fluid flow. The influence of cations on pores of  
58 soils is discussed by Marchuk et al. (2013).

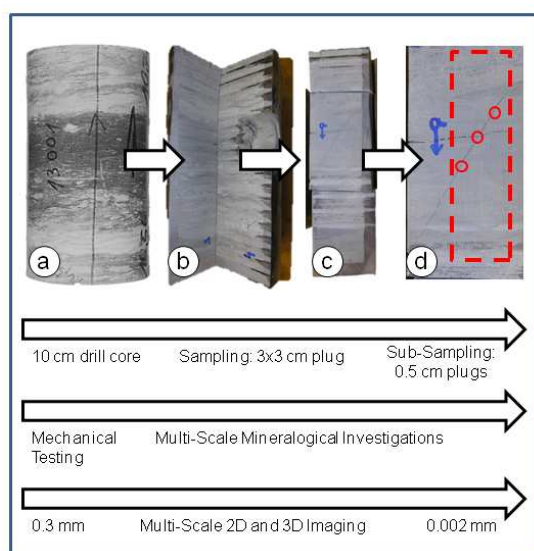
59 In HLRW research  $\mu\text{-CT}$  was used to investigate the wetting of clay pellets and for the  
60 assessment of homogeneity after wetting (van Geet et al., 2005), relations of mechanical  
61 properties and microstructure (Bésuelle et al., 2006; You et al., 2010), engineering properties  
62 such as deformation (Nakano et al., 2010), and to visualize anisotropy of deformation and  
63 the excavated damage zone (EDZ; You & Li, 2012). Keller et al. (2013) used a set of  
64 different methods (STEM, FIB, and  $\mu\text{-CT}$ ) which allowed the “characterization of the pore  
65 structure in the fine-grained clay matrix at different levels of detail” of the Opalinus Clay. The  
66 Opalinus Clay is particularly interesting because it will be the host rock and hence the main  
67 barrier for the Swiss repository for HLRW. In the Opalinus Clay, two different facies can be  
68 distinguished. The clay rich facies is referred to as “shaly facies” and hence distinguished  
69 from the “sandy facies”. To resolve differences of both facies, nanotomography was used



70 (Keller et al., 2013). Micro computed tomography is not suitable to resolve all microstructural  
 71 features of clays (micro- and mesopore-range) but rather useful to characterize the  
 72 macropore-scale which is relevant for visualizing the crack distribution, advective fluid flow,  
 73 and material heterogeneity, such as micro-bedding.

74 Especially in the field of geomechanical investigations, it is essential to get information about  
 75 the mineral composition and microstructure – before and after mechanical tests. All these  
 76 parameters have to be characterized to be able to increase the understanding of deformation  
 77 processes. While the porosity and microfabric of tectonically undeformed Opalinus Clay  
 78 (OPA) (Houben et al., 2013; Keller et al., 2011; Wenk et al., 2008) and naturally deformed  
 79 OPA (Laurich et al., 2014) have been intensively studied, little is known of the microstructure  
 80 and deformation mechanisms in experimentally deformed OPA.

81 In this study we present the investigations of an experimentally deformed OPA. The aim is  
 82 the visualization of the shear failure in various scales to get more information about the  
 83 deformation process. The deformation process is necessary for the long term safety case  
 84 analysis for HLRW repositories. Figure 1 showcases the general workflow and the main idea  
 85 for the investigation of the Opalinus Clay with a consequent multiple scale (top-down)  
 86 approach.



87

88 **Figure 1:** Generalized workflow for the multiple scale investigations of the Opalinus Clay: from  
 89 mechanical testing and imaging of the 10 cm drill core (a), to sub-sampled plugs (b & c) for  
 90 mineralogical and higher resolution imaging, to small scale samples (d) for high resolution and specific  
 91 region of interest investigations.



92 **MATERIAL & METHODS**

93 **Sample Material**

94 The investigated specimen (file 13001, drilling BLT-A6) derives from the Underground Rock  
95 Laboratory (URL) Mont Terri, St. Ursanne, Switzerland, and belongs to the sandy facies of  
96 the Opalinus Clay (Figure 2). The core sample has a diameter of 100 mm and a length of  
97 180 mm. The drilling is orientated perpendicular to the bedding.

98 The sample has been sealed by a special vacuum-bag to prevent the material from drying as  
99 best as possible, in order to obtain the original saturation condition for the mechanical  
100 testing. After the testing it was necessary to stabilize the sample with resin, since shear-  
101 failure was fully developed. The specimen was then stepwise sub-sampled for X-ray CT,  
102 mineralogical and geochemical investigations on different scales (from dm to mm of sample  
103 size).



104

105 **Figure 2:** Schematic overview of the Mont Terri Underground Rock Laboratory, showing the sampling  
106 location of the Opalinus Clay used for this study (modified after XXXX, YYYY).



## 107 **Mechanical Testing**

108 The claystone was tested by triaxial strength testing until a failure was developed. The test  
109 was executed in deformation controlled mode with a deformation rate of  $d\varepsilon/dt = 10^{-5}$  1/s and  
110 carried out under undrained condition (Gräsle & Plischke, 2010). After the mechanical testing  
111 the core was embedded in a resin to stabilize the specimen.

## 112 **Mineralogical and Geochemical Investigations**

113 XRD pattern were recorded using a PANalytical X'Pert PRO MPD  $\Theta$ - $\Theta$  diffractometer (Cu-K $\alpha$   
114 radiation generated at 40 kV and 30 mA), equipped with a variable divergence slit (20 mm  
115 irradiated length), primary and secondary soller, Scientific X'Celerator detector (active length  
116 0.59°), and a sample changer (sample diameter 28 mm). The samples were investigated  
117 from 2° to 85° 2 $\Theta$  with a step size of 0.0167° 2 $\Theta$  and a measuring time of 10 sec per step.  
118 For specimen preparation the top loading technique was used.

119 For XRF analysis of powdered samples, a PANalytical Axios spectrometer was used  
120 (ALMELO, The Netherlands). Samples were prepared by mixing with a flux material (lithium  
121 metaborate Spectroflux, Flux No. 100A, Alfa Aesar) and melting into glass beads. The beads  
122 were analyzed by wavelength-dispersive XRF. To determine loss on ignition (LOI), 1000 mg  
123 of sample material were heated to 1030 °C for 10 min.

124 The organic carbon (OC) content was measured with a LECO CS-444-Analysator after  
125 dissolution of the carbonates. Carbonates had been removed by treating the samples several  
126 times at 80 °C with HCl until no further gas evolution could be observed. Samples of 170-180  
127 mg of the dried material were used to measure the total carbon (TC) content. TIC was  
128 calculated by the difference of TC-TOC. The samples were heated in the device to 1800 -  
129 2000 °C in an oxygen atmosphere and the CO<sub>2</sub> was detected by an infrared detector. The  
130 device was built by LECO (3000 Lake Avenue, St. Joseph, Michigan 49085, U.S.A).

131 The CEC was measured using the Cu-Triethylenetetramine method (Meier & Kahr, 1999).

132 Both the smoothed drillcore section (21 mm x 18 mm) and the three polished core heads ( $\varnothing$   
133 5 mm) were analyzed for element distribution patterns by an energy-dispersive X-Ray  
134 fluorescence spectrometer, the EDXRF microscope M4-Tornado from Bruker-nano. The  
135 instrument is equipped with a Rh-tube generating a polychromatic beam, focused by a poly-  
136 capillary lense to a spot of a diameter of 17  $\mu$ m and two Xflash Silicon Drift Detectors (SDD).  
137 Take off angle for the tube in moving direction and the detectors is 51° incident and takeoff  
138 angle respectively, and the arrangement of the detectors to the tube is in 90° and 270°,  
139 respectively. Measuring time was 2 ms at 50 kV, 600  $\mu$ A and no filters were applied. The



140 stepsize for the overview was 25  $\mu\text{m}$  and for the core heads 5  $\mu\text{m}$ . False colour evaluation  
141 was performed by using the M4-tornado software esprit.

142 The polished three drillcore heads were investigated with an Environmental Scanning  
143 Electron Microscope (ESEM, type FEI Quanta 600 FEG) coupled with an energy dispersive  
144 X-ray (EDX) detector (two 30  $\text{m}^2$  Xflash Silicon Drift Detectors (SDD), Bruker-nano).  
145 Measurement conditions were 25kV, approximately 200  $\mu\text{A}$ , 4  $\mu\text{m}$  spot size, 19 times  
146 magnification at 11.4 mm working distance, and 2 minutes acquisition time.

#### 147 **X-Ray Computed Tomography**

148 The Opalinus Clay sample was first scanned with the speed|scan CT 64 located at the GE  
149 facility in Ahrensburg (Germany). Based upon a medical CT system, the CT 64 consists of a  
150 dust protected radiation protection cabinet with an integrated, rotating ring-shaped scanning  
151 device (gantry) and sample transport system for moving components through the scan ring.  
152 The system may accommodate samples of up to 900 mm in length and 500 mm in diameter.  
153 The CT datasets are automatically generated in the so-called helix scan mode with a high  
154 performance rotating anode X-ray tube and a 64 channel multi-line detector rotating around  
155 the sample. Its unique technique allows an overall cycle time of typically 1 minute per  
156 inspected sample (Ambos et al., 2014). Limitation of this type of equipment is the spatial  
157 resolution with typical 0.5 to 1 mm. Nevertheless, due to the very high power of the X-ray  
158 tube (72 kW), different mineral phases can be very distinctively observed. The scan was  
159 recorded with 140 KV and 140 mA within 13 seconds at a spatial resolution of 312  $\mu\text{m}$ .

160 Second, a CT scan of the same sample was recorded with the v|tome|x L300 system at the  
161 GE facility in Wunstorf (Germany). For technical applications one main goal is the detection  
162 of failures at smallest dimension possible. This approach led to the development of tubes  
163 with small focal spot to enable sharp images at high magnifications. One side effect of this is  
164 that the tube power is hereby limited. The only way to get enough information on the detector  
165 is to increase the scan time, typically from 30 min to 2 hours. In practice this delivers a  
166 resolution of approximately 60  $\mu\text{m}$  for a 10 cm core diameter, which is a factor of 10 better  
167 compared to “medical” CT scanners or such as the speed scan CT. Accordingly, the  
168 differentiation of mineral phases is significantly worse than for the high power system as  
169 described before. The scan parameters were 270 KV and 0.3 mA and the scan duration was  
170 145 min. With this system a spatial resolution of 57.5  $\mu\text{m}$  has been achieved.

171 For smaller cores (1 to 10 cm) this type of scanning device is still suitable, but when we get  
172 down with sample size to the mm range and thus want to achieve a resolution of a few  
173 microns there is a need to use so called nanofocus tubes with a focal spot size below 1  $\mu\text{m}$ .  
174 For the hereby described studies on 3 mm plugs a nanotom m system (GE Measurement &



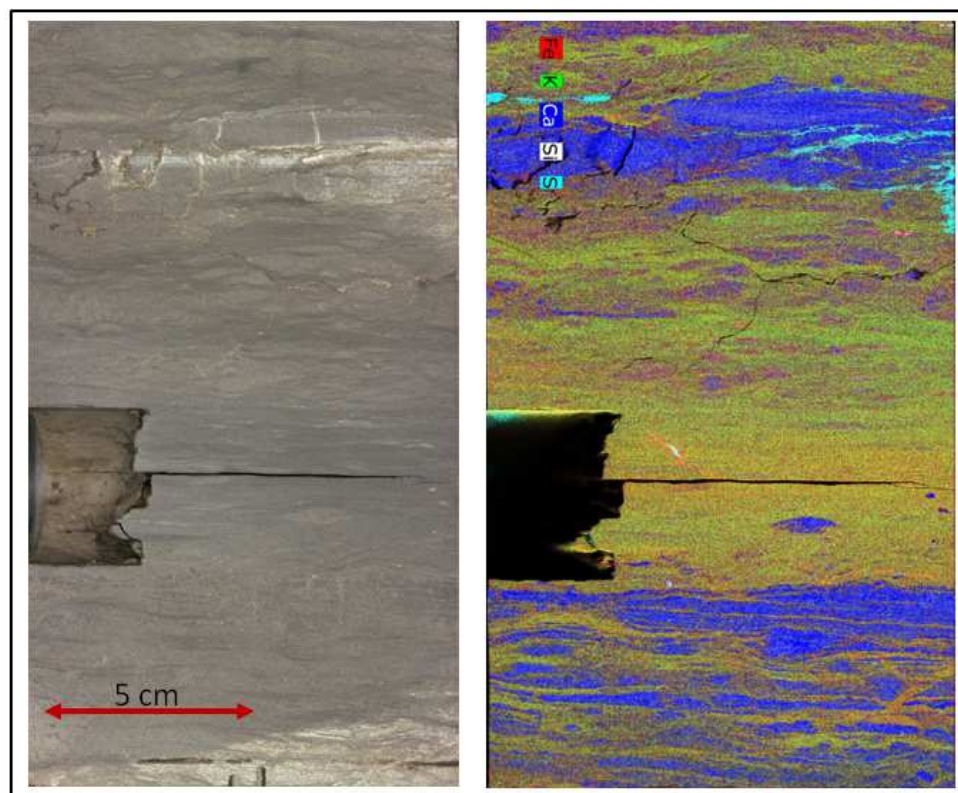
175 Control, phoenix(x-ray) was used. For the 3 cm x 3 cm plug and for the smallest samples  
176 which feature a diameter of 3 mm to 4 mm, a spatial resolution of 2.8  $\mu\text{m}$  has been achieved.

## 177 RESULTS

### 178 Mineralogical and Geochemical Composition

179 The bulk sample is dominated by quartz and carbonates which is typical for the sandy facies  
180 of the Opalinus Clay (Kaufhold et al., 2013; Siegesmund et al., 2013). Amongst the  
181 carbonates calcite was most abundant. In addition, kutnohorite was found which can be  
182 confounded with dolomite because of similar XRD reflections. The existence of traces of  
183 dolomite in addition to calcite and kutnohorite cannot be ruled out. Siderite is present as  
184 trace mineral. Muscovite and illite could not be distinguished because of similar XRD  
185 reflections. Therefore, the presence of muscovite in addition to illite/smectite is possible. The  
186 CEC accounts for 7 meq/100g pointing towards the presence of less than 10 mass-%  
187 smectitic layers which are predominately in illite/smectite mixed layer minerals. Minor  
188 amounts of kaolinite, feldspar, and pyrite were also found. Using LECO elemental analysis  
189 0.6 mass-% of organic material was found. Assuming an average C-content of carbonate  
190 minerals of about 12 mass-% results in slightly more than 40 mass-% carbonates and 0.9  
191 mass-% of sulfur corresponds to almost 2 mass-% pyrite. This composition is in accordance  
192 with Kaufhold et al. (2013) and Siegesmund et al. (2013).

193 The aim of the present study was to investigate crack formation which could be related to  
194 microstructural features or mineralogical heterogeneities (as fine bedding, fossil shells, etc.).  
195 Therefore, the heterogeneity was investigated by  $\mu$ -XRF and SEM. First the crossing of two  
196 cracks was investigated with respect to the mineral indicator elements Si, Ca, Fe, and K. Si  
197 represents quartz, Ca can be mostly found in carbonates, Fe dominates in pyrite and/or Fe-  
198 oxohydroxides, and K indicates clay rich layers because it can be mostly found in  
199 illite/smectite mixed layer minerals being the main clay mineral of the Opalinus Clay. Results  
200 are depicted in Figure 3 . The XRF scanner results reveal the heterogeneities of the sample  
201 in the relevant scale with a resolution of a few  $\mu\text{m}$ . The bedding, horizontal in the image, is  
202 reflected by a few mm thick clay layers (green) with more carbonatic layers in between. A  
203 centimeter scaled region was found at the lower left section of the image which could be a  
204 fossil, e.g. a shell fragment. However, this microstructure feature could not be related to the  
205 cracks.

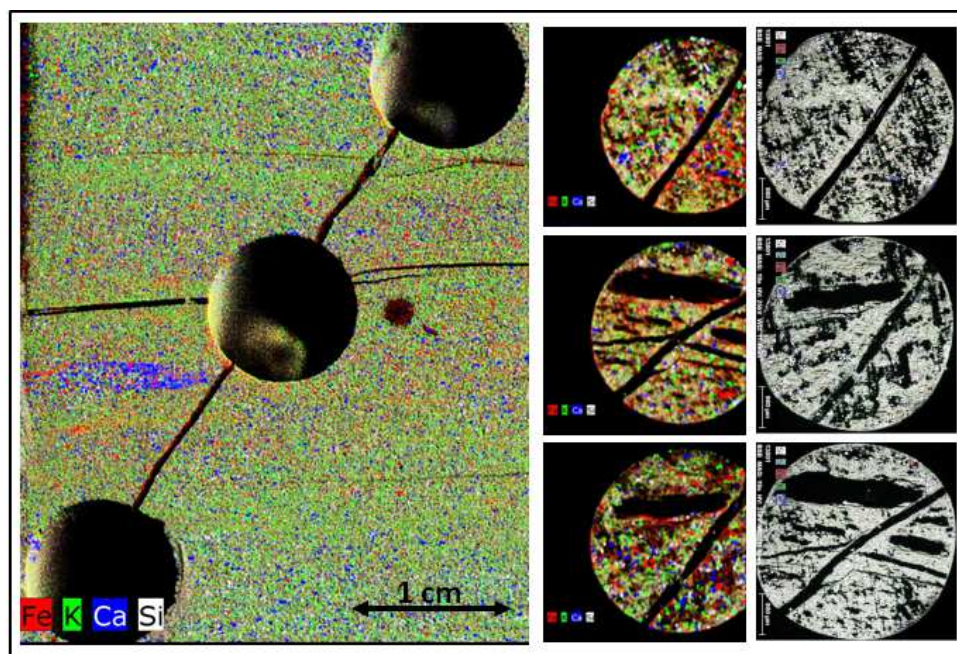


206

207 **Figure 3:** Results of the 2D mineralogical mapping on the large core sample (10 cm).

208 Therefore, magnification was increased (Figure 4). In these small sections of about 5 mm,  
209 bedding features could not be detected anymore. Instead a few 50-100  $\mu\text{m}$  thick bands of  
210 either carbonates (blue) or clays (green) could be observed with a significant angle  
211 compared to bedding. Assuming that these small lineaments were no XRF artefacts, it can  
212 be supposed that a crack started to form there. The location from where tension relief  
213 observable as crack formation started is assumed to be outside the investigated area.  
214 Therefore it can only be assumed that the small lineaments observed both with the XRF  
215 scanner as well as with the SEM could be the starting point for crack formation.



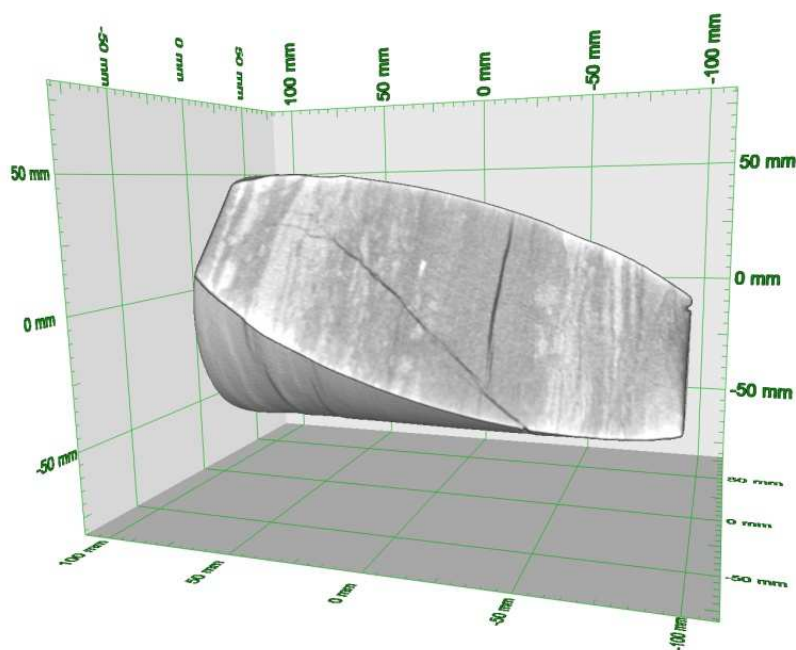


216

217 **Figure 4:** Results of the 2D mineralogical and geochemical mapping on the small samples.

#### 218 **Large Scale and High Speed X-ray CT**

219 The CT results of the speed|scan CT 64 show good contrast resolution due to its high tube  
220 power (up to 72 kW). Layering, i.e. changes in the mineralogical composition of the core, can  
221 be easily detected based on slightly changing density (see Figure 5). The clay-rich areas are  
222 characterized by darker grey values (e.g. middle section of Figure 5), carbonatic regions are  
223 indicated by higher, i.e. brighter grey values. Layering features can be qualitatively observed  
224 in about the millimeter scale. Cracks and pores can be spatially resolved down to 0.5 mm.  
225 For this core, two main fractures can be observed: a horizontal crack (fracture A), which is  
226 probably caused by de-hydration (so called diking) of the core, and a shear crack caused by  
227 the laboratory mechanical testing. Interestingly, the shear fracture is located within the clay-  
228 rich area of the OPA sample. Starting point is right at the border between clay-rich and  
229 carbonatic zone (right hand side of Figure 5). Additionally, the fracture ends in a carbonatic  
230 region (left hand side of Figure 5) and seems to fan out within that layer. The 3D data set can  
231 be virtually sliced in any direction to emphasize the specific layering or location of the crack  
232 system.

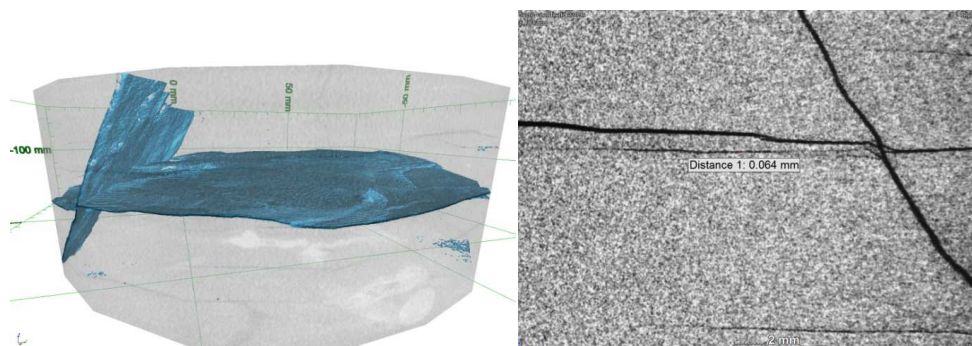


233

234 **Figure 5:** Partial 3D view of the speed|scan CT result. Within the virtual core, the layered structure  
235 (due to changes in the mineralogical composition) and two main cracks can be observed.

### 236 Large Scale and High Resolution X-ray CT

237 Compared to the faster device explained in the preceding paragraph, the CT results of the  
238 v|tome|x L300 show much better spatial resolution (down to approximately 60  $\mu\text{m}$  for 10 cm  
239 sample width). As the highest power of this system is 0.5 kW, the phase contrast is not as  
240 high but still sufficient to detect larger zones of different densities. On the other hand the  
241 fractures are much better resolved (5 times better resolution) and the delicate network can  
242 be nicely visualized (Figure 6) and studied more in detail. The effective fracture size for  
243 segmentation is in range of the achieved voxel resolution. Segmentation was performed in  
244 the central part of the sample, where the large horizontal crack is intersected by the diagonal  
245 oriented crack system. Additionally, many smaller cracks could be observed, in most cases  
246 also horizontally oriented cracks, which also might be related to diskling effects. Interestingly,  
247 a zone of higher fracture density, or at least of higher density due to the lower grey values of  
248 that region, is located near the intersection of the two main fractures (Figure 6, right hand  
249 side). Consequently, this area has been chosen for sub-sampling and 2D and 3D  
250 investigation with higher resolution.

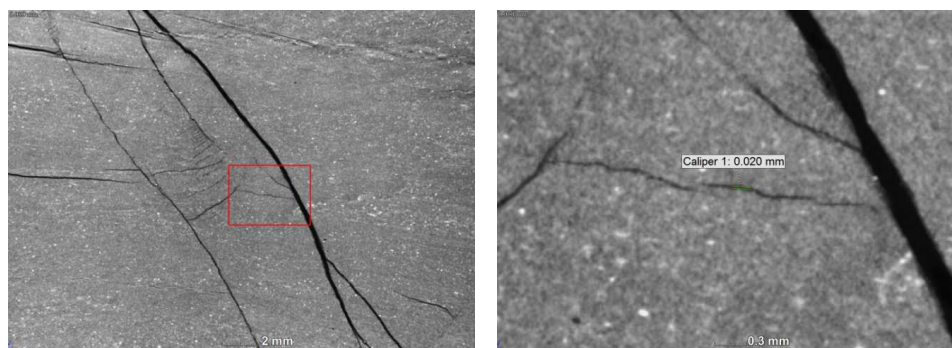


251

252 **Figure 6:** The main fractures have been segmented (left hand side) and visualized in a transparent 3D  
253 view. Despite the two major cracks, numerous tiny cracks can be detected. The fracture width can be  
254 measured down to approximately 60  $\mu\text{m}$ .

#### 255 **Small Scale and High Resolution X-ray CT**

256 In order to achieve higher image resolution and to obtain good image quality, it is mandatory  
257 to downsize the sample as a smaller voxel size can only be achieved by increasing the  
258 geometrical magnification for the hereby described CT systems. In a first step a 3 cm x 3 cm  
259 sample has been cut out and a CT scan was performed on a nanotom m system with a voxel  
260 resolution of about 18  $\mu\text{m}$ . The results (Figure 7) show significant improvement in diversity of  
261 small details. Individual carbon shells can easily be distinguished and the overall fracture  
262 pattern becomes more and more resolved. Accordingly, smaller shear fractures can be  
263 detected, which are more or less parallel oriented to the main shear crack (Figure 7, left hand  
264 side). The zoomed in view (Figure 7, right hand side) reveals the local presence of small  
265 fractures, which connect the shear cracks with each other.

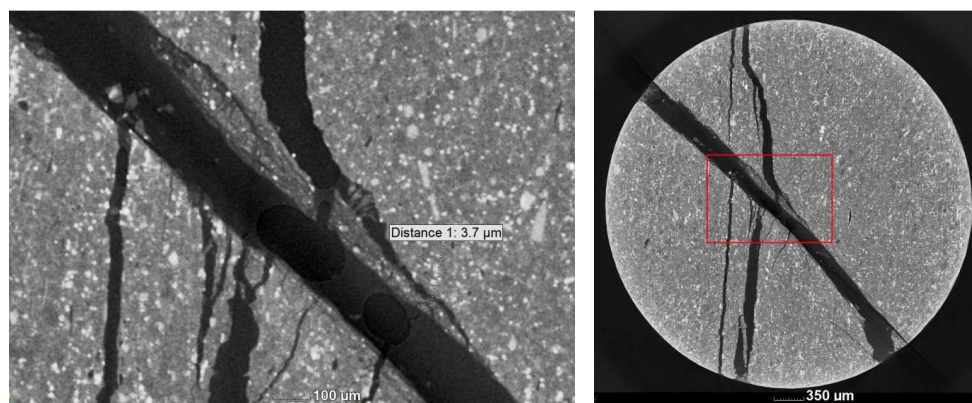


266

267 **Figure 7:** Many small scale details of the mineralogical composition are now visible, as well as  
268 numerous small fractures. The red area is zoomed in on the right hand side. Here, a small fracture is  
269 shown, which has an approximate aperture of about 20  $\mu\text{m}$ .



270 For even smaller scale, micro plugs were drilled with diameter of about 3mm and scanned on  
271 the same system with a voxel resolution of approximately 2 - 3  $\mu\text{m}$ . For this high resolution  
272 single grains can be observed as well as micro cracks and small meso-pores (Figure 8).  
273 Though no specific correlation between fracture occurrence and mineralogy can be  
274 observed, a small zone around that point, where the shear and disk-like fracture intersect  
275 each other is of very special interest. This area can be characterized as a so called mylonitic  
276 zone, i.e. an area with many small fractures and cracks where particles have been re-  
277 arranged on the fracture surface. As far as literature research reveals, this seems to be the  
278 first reported CT data set of such a zone. For more details, this sample has been used for the  
279 micro-scale mineralogical and microstructure investigations, to achieve more evidence for  
280 this special feature.

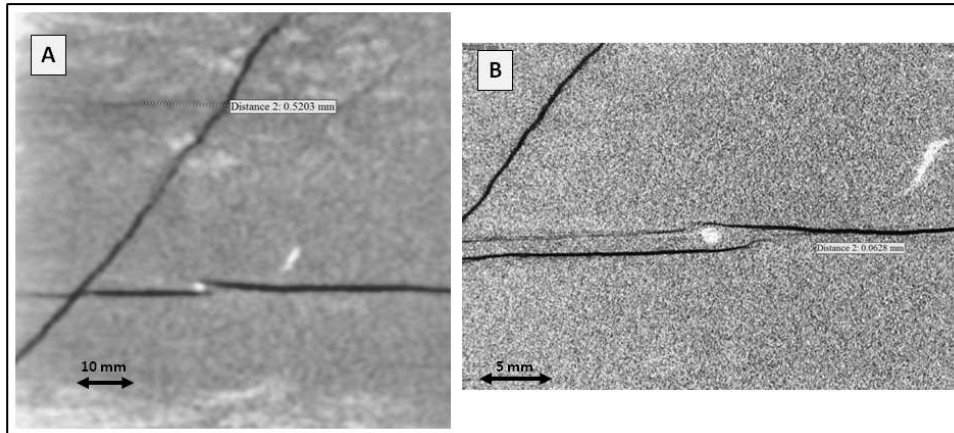


281

282 **Figure 8:** For the high resolution data set, many small cracking features can be observed. The  
283 zoomed in area marked by the red box is of special interest, since indications for a mylonitic zone can  
284 be found, where the disk-like and shear fracture intersect.

#### 285 **Multi-scale comparison of CT results**

286 For the investigation of the OPA material, a consequent top-down approach has been used.  
287 Due to the different 3D imaging scales, quantification of sample features (here: the cracks  
288 and fractures) is challenging and may lead to different results. Table 1 highlights the number  
289 of detected cracks and fractures as well as the average aperture of the two main fractures in  
290 relationship to the sample size and to the derived imaging resolution. Whereas the coarse  
291 resolution scans show good results for a first mineralogical and textural sample  
292 characterization, especially details on the fracture development cannot be revealed (Figure  
293 9).



294

295 **Figure 9:** Direct comparison of both “coarse” resolution scans with different techniques. Either  
 296 enhanced phase contrast or spatial resolution can be derived. Hence, both techniques should be used  
 297 complementary.

298 Accordingly, sub-sampling the OPA material stepwise greatly increases the information of  
 299 the fracture network. Hence, the total number of cracks detected increased by a factor of  
 300 almost 36. If the result is up-scaled to the large core size, this would be a factor as high as  
 301 100 to 150. Additionally, the existence of smaller diskings as well as of smaller shear fractures  
 302 has been showcased in the previous sections. As a matter of fact, the evaluation of the two  
 303 main fractures on different scales leads to very different results. For the large core, fracture  
 304 apertures are greatly over-estimated (3 to 6 times, related to scanning resolution) due to  
 305 partial volume effects and – of course – the effective segmentation resolution. Results of the  
 306 small and micro samples are almost equal for the main crack evaluation. Nevertheless, the  
 307 number of detected cracks still increases by a factor of about 2. Features such as the  
 308 observed mylonitic zone can be indicated from the coarse scan data and evaluated in detail  
 309 by the high resolution image data.

310 **Table 1:** Comparison of sample size and image resolution related fracture detection and geometrical  
 311 fracture analysis results for the two main cracks observed.

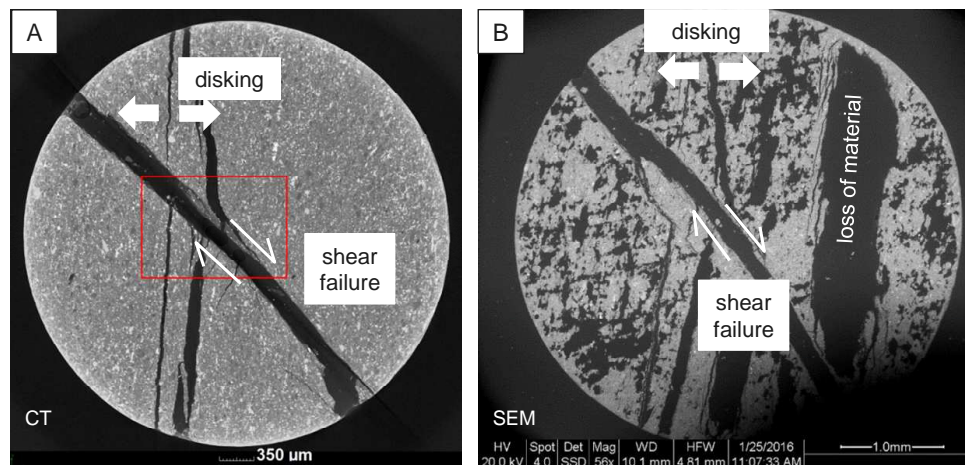
core size [mm]	voxel resolution [μm]	# of cracks detected	average crack aperture [μm]	
			fracture A	fracture B
100	312,5	3	990	1300
100	57,5	15	393	364
30	17,8	47	185	237
3	2,8	107	182	228

312



### 313 **Mircostructural Investigation**

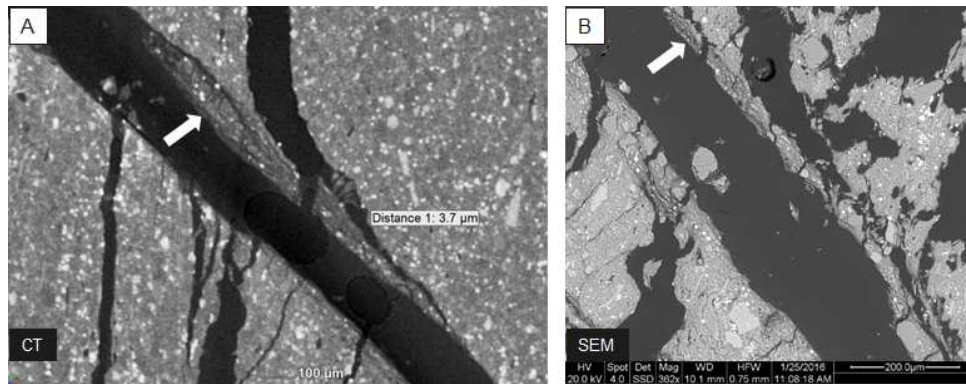
314 The microstructural investigation was carried out on micro plugs. The plugs were first  
315 scanned with the high resolution X-ray CT. For the scanning electron microscope  
316 investigations (low vacuum) the samples were embedded in resin and the surface was  
317 polished. CT-investigation provided complete 3D information of the samples. One section of  
318 the 3D scan is shown in Figure 10-A. The SEM image, of course, only represents the  
319 polished surface of the plug (Figure 10-B). Dark areas represent cracks filled with air (CT) or  
320 the resin (SEM). Much more dark areas were observed by SEM which resulted from artifacts  
321 caused by sawing and polishing. Ideally an even surface is produced by polishing but the  
322 preparation of even surfaces of claystones is difficult. Depending on sample pretreatment  
323 (drying, wetting etc.) claystones may at least partly disintegrate resulting in a loss of material  
324 upon sawing and polishing. This explains why more dark areas were observed by SEM.  
325 Nevertheless, the main features to be investigated were observed by both techniques.



326

327 **Figure 10:** Sample overview: A) X-ray CT image of a single layer, B) SEM-image of the polished  
328 surface.

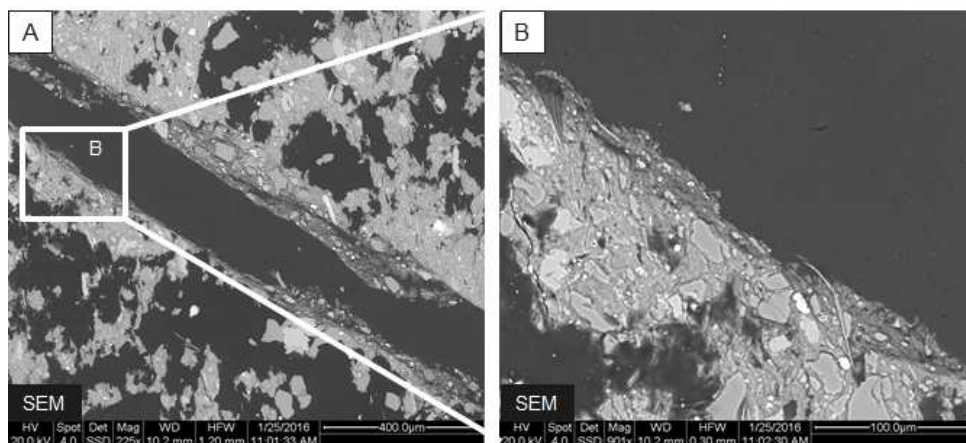
329 In both figures, shear failure and diskings could be observed. Disking is assumed to be a relief  
330 failure in the bedding plane. It was already observed before the mechanical test was  
331 performed. The shear failure is overprinting diskings and a material offset is clearly visible in  
332 both images. A closer look at the shear failure reveals more details (Figure 11). In the CT-  
333 image the shear failure and smaller micro-cracks parallel to bedding planes could be  
334 observed. Different minerals can be recognized, but a clear mineral boundary is not visible.  
335 Nevertheless on the top of the shear zone a darker zone is identifiable, which is a result of  
336 particle reduction.



337

338 **Figure 11:** Zoomed in view of the shear failure from the X-ray CT image (A) and of the SEM image  
339 (B).

340 The SEM-image (Figure 11-B) has a higher resolution compared to the CT-image. In  
341 principle, identical microstructure features were found with both methods. In combination with  
342 the 2D SEM inspection, more indications can be observed in order to find out what happened  
343 in the mylonitic zone.



344

345 **Figure 12:** Very high resolution close up of the mylonitic zone located in the shear failure area. It can  
346 be clearly observed, how the particles have been pulled apart from the original matrix material and re-  
347 arranged near the surface.

348 Close up SEM images (Figure 12) prove that the claystone did not simply break as one  
349 would expect from broken glass. Either before or throughout breaking a rearrangement of the  
350 particles and hence a destruction of the microstructure occurred. Platy particles as micas  
351 rearranged (Figure 12-B) which indicates plastic deformation. As a result a “micro mylonitic”  
352 seam at both sides of the crack was observed. This phenomenon was already observed by  
353 Laurich et al. (2014) for naturally deformed OPA. They explain the occurrence of this  
354 mylonitic zone as a gouge zone. It is not clear whether the mylonitic zone formed just before



355 breaking or if it formed by the relative movement of both sides of the crack. Nevertheless, it  
356 is a key finding that such a zone exists in artificially deformed OPA, and that this zone has  
357 been observed both, in 2D and 3D data sets.

## 358 **CONCLUSIONS**

359 For the long term safety analysis of repositories for radioactive waste it is necessary to  
360 predict the mechanical behavior of the host rock. The understanding of mechanical  
361 processes in argillaceous rocks is considerably less developed than that of other materials  
362 like salt rocks. Hence the investigations presented above, for microstructure analysis in  
363 various scales regarding mechanical failure, is important to develop our understanding of  
364 mechanical behavior of clay stones.

365 The OPA material has been intensively studied by a variety of multiple scale and non-  
366 destructive 3D X-ray CT investigations, following a consequent top-down approach to identify  
367 specific regions of interest. According to the mechanical experiment, it has been observed  
368 that the shear failure is located in a clay-rich area. Within the intersecting area of the two  
369 main fractures, a so called mylonitic zone with a particle reduction was observed on the open  
370 shear failure using CT and SEM techniques. But it is not known, until now, when and how  
371 this zone was developed. As far as the authors are aware, this is the first time that  
372 experimental deformation shows such a mylonitic zone.

373 Therefore it is necessary to investigate further mechanical loaded specimens under different  
374 conditions (water content and strain). These mechanical investigations should be monitored  
375 with non-destructive X-ray CT investigations and in further step accompanied with sub  
376 sampling and small-scale image investigations. Then we have the possibility to get more  
377 information about the petrophysical processes behind the mylonitic zone. All these  
378 investigation can help us to develop our understanding of mechanical behavior which is an  
379 important part in the long term safety analysis of potential hazardous waste disposal places.

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467 **Appendix**468 **Table 2:** Geochemical and mineralogical composition of the bulk sample.

mineral composition			XRF	
quartz	++		SiO <sub>2</sub>	[mass-%] 49,8
calcite	++		TiO <sub>2</sub>	[mass-%] 0,7
Mg-kutnohorite	+		Al <sub>2</sub> O <sub>3</sub>	[mass-%] 10,7
muscovite/illite (ML)	+-		Fe <sub>2</sub> O <sub>3</sub>	[mass-%] 5,1
kaolinite	+-		MnO	[mass-%] 0,1
feldspar	+-		MgO	[mass-%] 2,2
pyrite	+-		CaO	[mass-%] 12,2
			Na <sub>2</sub> O	[mass-%] 0,4
			K <sub>2</sub> O	[mass-%] 1,9
			P <sub>2</sub> O <sub>5</sub>	[mass-%] 0,3
			SO <sub>3</sub>	[mass-%] 1,2
			LOI	[mass-%] 15,4
<b>LECO</b>				
C <sub>total</sub>	[mass-%]	3,6		
C <sub>org</sub>	[mass-%]	0,6		
C <sub>inorg</sub>	[mass-%]	3,1	Sum	[mass-%] 99,9
S <sub>total</sub>	[mass-%]	0,9		
<b>CEC</b>				
CEC	[meq/100g]	7		

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