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## Further insight into the microstructure of compacted bentonite-sand mixture

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26 **Abstract**

27

28 Compacted bricks of bentonite/sand mixture are planned to be used as sealing plugs in  
29 deep radioactive waste disposal repositories because of their low permeability, high  
30 swelling capacity and favourable properties with respect to radionuclide retention. The  
31 isolating capacity of compacted bentonite/sand mixture is closely related to  
32 microstructure features that have been often investigated, in particular by using  
33 scanning electron microscope (SEM or ESEM) and mercury intrusion porosimetry  
34 (MIP).

35 In this work, microfocus X-ray computed tomography ( $\mu$ CT) observations were used in  
36 parallel with MIP measurements to further investigate at larger scale the microstructure  
37 of a laboratory compacted bentonite/sand disk (65/35% in mass). Qualitative  
38 observation of  $\mu$ CT images showed that sand grains were inter-connected with some  
39 large pores between them that were clearly identified in the bimodal pore distribution  
40 obtained from MIP measurements. Due to gravitational and to frictional effects along  
41 the specimen periphery, a higher density was observed in the centre of the specimen  
42 with bentonite grains more closely compacted together. This porosity heterogeneity was  
43 qualitatively estimated by means of image analysis that also allowed the definition of  
44 the representative elementary volume. Image analysis also provided an estimation of the  
45 large porosity in good agreement with MIP measurements.

46 **Keywords:** compacted sand-bentonite mixture, sealing plug, radioactive waste disposal,  
47 microstructure, mercury intrusion porosimetry, X ray micro tomography

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## 55 **Introduction**

56 In deep radioactive waste disposal concepts, sealing plugs made of compacted  
57 bentonite/sand blocks are planned to be used to close the galleries and to limit water  
58 transfers. Compacted bentonite/sand plugs are used for their low permeability, high  
59 radionuclides retention capability and sealing/swelling abilities when infiltrated by  
60 water (e.g. Pusch, 1979; Yong et al., 1986). Once the disposal galleries are closed, plugs  
61 will be progressively infiltrated by the pore water of the host rock. They will swell and  
62 seal the so-called technical voids of the system, i.e. the voids remaining between blocks  
63 and at the interfaces with the rock. These technical voids are estimated at 14% of the  
64 total volume of the plugs by IRSN (Institut de Radioprotection et de Sûreté Nucléaire,  
65 the French expert Institution in the field of nuclear safety) in the in-situ SEALEX  
66 experiment that they carry out in their Tournemire Underground Research Laboratory.

67

68 Various investigations of the microstructure of compacted bentonites and sand bentonite  
69 mixture have been carried out by using mercury intrusion porosimetry (MIP) and  
70 scanning electron microscopy (SEM, ESEM, Komine and Ogata, 1999; Villar and  
71 Lloret, 2001; Montes-H, 2002; Cui et al., 2002; Lloret et al., 2003; Agus and Schanz,  
72 2005; Delage et al., 2006). These techniques require a cautious preliminary dehydration  
73 of the samples, most often by freeze-drying. They provide local observations on a small  
74 part of millimetric samples. These localized analytical techniques can be fruitfully  
75 complemented by use of by microfocus X-ray computed tomography ( $\mu$ CT), a high-  
76 resolution non destructive 3D observation technique. The  $\mu$ CT does not need any

77 sample pre-treatment and gives further 3D information on the whole sample (including  
78 grain size distribution as well as pore size distribution and pore inter-connectivity).  
79 Previous applications of  $\mu$ CT have been devoted to the monitoring of hydro-chemo-  
80 mechanical processes (Comina et al., 2008), to the detection of dessication cracks  
81 (Gerbrenegus et al., 2006; Mukunoki et al., 2006), to the visualisation of  
82 diffusion/hydration phenomena, to the study of fluid movements (Rolland et al., 2003;  
83 Rolland et al., 2005; Carminati et al., 2006; Koliji et al., 2006) and to the investigation  
84 of the microstructure of compacted bentonite based materials (Kozaki et al., 2001, Van  
85 Geet et al., 2005 and Kawaragi et al., 2009). In this work,  $\mu$ CT was coupled to MIP for  
86 further microscopic investigation of a compacted bentonite/sand sample.

## 87 **Material and methods**

### 88 ***Material***

89 The studied material is a compacted mixture of Wyoming MX-80 bentonite (65% in dry  
90 mass, commercial name Gelclay WH2) and sand (35%). The MX80 bentonite contains  
91 92% of montmorillonite with several other minerals including quartz, alumina, and  
92 hematite (Tang et al., 2008). The sand is quartz sand (commercial name TH1000).

93

94 The sand-bentonite mixture was provided in boxes containing a powder with an initial  
95 water content of 10.2% and a suction of 73.3 MPa measured with a chilled mirror dew  
96 point tensiometer (Decagon WP4). Samples of sand and bentonite were also provided  
97 separately. The grain size distribution of the bentonite powder obtained by dry sieving  
98 is presented in Figure 1 together with that of the deflocculated bentonite obtained by  
99 sedimentation. The grain size distribution of the sand is also plotted in Figure 1.

100

101 The grain size distribution curves show that the bentonite powder grains are larger than  
102 the sand grains with  $D_{50}$  values of 1.2 and 0.6 mm respectively. Both curves are  
103 representative of well sorted materials. The unit mass of the bentonite particles that  
104 constitute the bentonite grains was measured by using a pycnometer with water and was  
105 found equal to  $2.77 \text{ Mg/m}^3$  in agreement with published data (Madsen, 1998; Karnland  
106 et al., 2006). The unit mass of the bentonite powder was determined by using a  
107 pycnometer with a non aromatic hydrocarbon fluid (commercial name Kerdane). The  
108 bentonite grains appeared to be stable once immersed in Kerdane and a value of  
109  $2 \text{ Mg/m}^3$  was obtained. This value could suffer from some uncertainty due to possible  
110 Kerdane infiltration into the grains. A comparable unit mass value was however  
111 obtained from the cumulative pore size distribution curve presented later in Figure 2  
112 that allows determining the unit mass of the bentonite grains from the value of inter-  
113 grains porosity. The unit mass of the sand grains was found equal to  $2.65 \text{ Mg/m}^3$ .

114

115 Compacted samples were prepared by uniaxial static compaction (strain rate of 0.1  
116 mm/min) in a cylindrical mould in order to obtain a disk (diameter 50 mm, height 10  
117 mm) at the targeted dry density ( $1.8 \text{ Mg/m}^3$  obtained at a maximum compaction stress  
118 of 25.5 MPa). The sample water content was 10%, resulting in a degree of saturation of  
119 55 % and a suction of 76.3 MPa. Note that this value is close to that of the powder prior  
120 to compaction (73.3 MPa). The slightly higher value could be due to some effects of  
121 evaporation during the process of compaction. In any case, these small changes in  
122 suction after compaction are consistent with the observations of Li (1995), Gens et al.  
123 (1995) and Tarantino and De Col (2008) on soils compacted on the dry side of Proctor

124 optimum. It confirms that suction is governed by aggregates that are not much affected  
125 by the compaction. As a consequence, there is little dependency of the water retention  
126 properties on the sample density in compacted bentonite based materials.

### 127 ***Mercury intrusion porosimetry***

128 The pore size distribution of the compacted samples was measured on freeze dried  
129 samples by using an Autopore IV 9500 mercury intrusion porosimeter (Micromeritics)  
130 working to a maximum pressure of 230 MPa. Instantaneous freezing was carried out by  
131 plunging small samples (volume 0.40 cm<sup>3</sup>) into slush nitrogen (liquid nitrogen at cooled  
132 down from -195°C to -210°C by vacuum application, Delage et al., 2006). In such  
133 conditions, there is no nitrogen boiling around the samples when plunging them into  
134 nitrogen, resulting in an optimized quick freezing and good microstructure preservation.  
135 The intruded porosity was determined from the total volume of mercury intruded into  
136 the sample and the pore size distribution was obtained, in a standard fashion, assuming  
137 parallel, cylindrical nonintersecting pores of different radii, using the Autopore IV 9500  
138 V1.09 standard software package. The intruded porosity was systematically compared  
139 to the total porosity obtained by standard methods so as to detect the smaller porosity  
140 (entrance equivalent diameter smaller than 5.5 nm) not intruded by mercury at the  
141 highest applied pressure (200 MPa).

### 142 ***Microfocus X-ray tomography and image analysis***

143 Microfocus x-ray computed tomography ( $\mu$ CT) was used to examine in three  
144 dimensions the internal microstructure of the compacted bentonite/sand mixtures.  $\mu$ CT  
145 is a non-destructive observation technique that has proven to be useful in the  
146 investigation of various geological porous media including compacted bentonite

147 (Kozaki et al., 2001), bentonite pellet/powder mixture (Van Geet et al., 2005) and  
148 compacted bentonite/quartz mixture (Kawaragi et al., 2009).  $\mu$ CT consists firstly of  
149 recording a set of two-dimensional X-ray radiographs of an object at several angles  
150 (typically at  $180^\circ$  or  $360^\circ$ ) and secondly in reconstructing the 3D slices from the  
151 radiographs using a mathematical algorithm. The final 3D image of the internal  
152 structure is obtained by stacking the slices. The final measurement is the attenuation  
153 coefficient to x-ray which depends on the mass density and the atomic number of the  
154 object (Ketcham and Carlson, 2001; Van Geet et al., 2005).

155

156 The  $\mu$ CT scans presented here were carried out with the device of the Navier laboratory  
157 (Ecole des Ponts ParisTech), an “Ultratom” device specifically designed and  
158 manufactured by RXsolutions (France). Images were reconstructed using the software  
159 Xact-reconstruction developed by RXsolutions. The system is a dual-head and dual-  
160 imager scanner; two sources [a nano-focus xs-160hpnf/GE-Phoenix (160 kV, 15W, 0.9  
161  $\mu$ m min spot size) and a micro-focus xs-225d/GE-Phoenix (225 kV, 320W, 5  $\mu$ m min  
162 spot size)], two interchangeable imagers: [HD camera PhotonicScience VHR  
163 (4008x2672 pixels, 9 $\mu$ m pixel size) and a flat-panel-CsI scintillator-Varian  
164 2520V/Paxscan (14 bit, 1920x1526 pixels, 127 $\mu$ m pixel size)].

165

166 A micro-focus source was used together with a flat panel. The X-ray source parameters  
167 were a voltage of 140 kV with an intensity of 210  $\mu$ A, a source power resulting in a spot  
168 size below or equal to the spatial resolution, here a voxel size of 30  $\mu$ m (a voxel  
169 describes a Volume Pixel which is the smallest distinguishable box-shaped portion of a  
170 three-dimensional image). The sample was scanned using 1440 projections on  $360^\circ$



171 with an exposure time of 0.2 s. Each projection was integrated on 4 s (average of 20  
172 frames) in order to obtain a good signal-to-noise ratio. The total scanning time was  
173 about an hour and a half. The final 3D image is a 16 bit type with a size of  
174 1840x1840x386 voxels. The image analysis and treatment was then carried out by using  
175 ImageJ, a public domain Java image processing program (Rasband, 1997-2012). Note  
176 that the image was first converted to 8 bit for size reasons.

## 177 **Results**

### 178 ***Pore size distribution***

179 The pore size distribution determined at a dry density of  $1.8 \text{ Mg/m}^3$ , a water content of  
180 10% and a suction of 76.6 MPa is presented in terms of cumulative and density  
181 functions curves in Figure 2. The bimodal curve obtained is comparable to that of  
182 samples compacted dry of optimum (Ahmed et al., 1974; Delage et al., 1996; Romero et  
183 al., 1999 among others) and also with the results of Cui et al. (2002), Lloret et al.  
184 (2003), Agus and Schanz, (2005) on compacted bentonite materials. The PSD curve  
185 indicates that the smaller pores population (micropores) has a mean entrance diameter  
186 of  $0.19 \mu\text{m}$  and the larger pore population (macro-pores) has a mean entrance diameter  
187 of  $22 \mu\text{m}$ . Note that the intrusion of mercury into the specimen starts at quite a low  
188 pressure, evidencing a largest diameter of  $360 \mu\text{m}$ , the upper limit of the method. The  
189 total porosity (0.337) calculated in a standard fashion by macroscopic volume  
190 measurements is also plotted in Figure 2. The smaller value of the porosity intruded by  
191 mercury (0.273) confirms that pores with an entrance diameter smaller than  $5.5 \text{ nm}$  (the  
192 lower limit of MIP) could not be intruded by mercury. This feature is typical of

193 compacted bentonites and strongly depends on the water content (the higher the water  
194 content, the higher the smallest porosity, see Delage et al., 2006).

195

## 196 ***Microfocus X-ray computed tomography ( $\mu$ CT)***

### 197 **Image Observation**

198 The reconstructed 3D volume of a compacted sample of bentonite/sand mixture at a dry  
199 density of  $1.8 \text{ Mg/m}^3$  and a water content of 10% is presented in Figure 3. The position  
200 of the plane where the cross section has been done is indicated. The horizontal cross  
201 section at 4 mm from the top of the sample is provided in Figure 4.

202

203 The  $\mu$ CT technique is able to distinguish the various components according to their  
204 density and atomic composition (the grey level range goes from white representing high  
205 attenuating material to black representing void). A clear distinction can be made  
206 between the clearer sand grains and the less clear larger bentonite powder grains  
207 (hydrated at a gravimetric water content of 10.6%), even though their respective  
208 densities are not so different (around  $2 \text{ Mg/m}^3$  for the bentonite grains and  $2.65 \text{ Mg/m}^3$   
209 for sand grains). This is due to the atomic composition difference. One can also observe  
210 in Figure 4 that the sample density is not strictly homogeneous with more voids  
211 observed around the sample and a larger density with less voids in the centre. This  
212 shows that, unsurprisingly, compaction was less effective around the sample along the  
213 ring against which friction effects resulted in looser compaction. Detailed observation  
214 made on a zoom taken close to the sample perimeter also shows that the bentonite  
215 powder grains remain clearly apparent around the sample and that they are apparently

216 more frequent than in the centre. In the centre, powder grains appear to be more  
217 compacted one against another, resulting in a denser structure.

218

219 Inspection of Figure 4 also shows that the bentonite/sand mixture is not really  
220 homogeneous and that sand grains and powder grains appear to be somewhat  
221 segregated. As indicated in the Figure, aggregations of powder grains are observed in  
222 some locations and sand grain packings with inter-grains pores in other locations. This  
223 segregation, probably resulting from the difference in density between the bentonite  
224 powder and the sand grains, is not compatible with the common statement that, in a 35-  
225 65% sand/bentonite mixture, sand grains should be homogeneously scattered among a  
226 (supposedly finer) clay matrix. Conversely, it seems that, due to segregation, some  
227 continuity between the sand grains is observed. As a consequence, there could be some  
228 continuity and interconnection of the pores located between sand grains.

229

230 Note that this segregation is probably depending on the process adopted here of pouring  
231 the dry mixture into the ring. It would hence be interesting to investigate the effects of  
232 two possible options for preparing more homogeneous sand-bentonite mixture: i) by  
233 previously moisturizing the mixture so as to make the clay grains stick to sand particles,  
234 resulting in less segregation than in the dry mixture, provided the wet mixture is  
235 thoroughly mixed prior to being poured into the ring and ii) by grinding bentonite grains  
236 to smaller sizes. Obviously, the heterogeneity observed here is typical of the preparation  
237 procedure of pouring a dry mixture into the compaction ring.

238

239 A more detailed examination of the voids configuration shows that many voids are  
240 located between sand grains. Around the sample, some voids are also located between  
241 powder grains but this is less true in the denser centre in which bentonite grains appear  
242 to be aggregated together. This means that the maximum compression stress, in spite of  
243 being as high as 25.5 MPa, is not high enough to allow the intrusion of clay particles  
244 (hydrated at a water content of 14.5 % with a 76.6 MPa suction) into the pores located  
245 between the sand grains. This high compression stress only allows some aggregation of  
246 bentonite grains as observed in the centre (around 8 grains of various diameters are  
247 aggregated in the circle indicated). This slight deformability of dry grains under high  
248 stresses is compatible with the observation made above about the small suction  
249 variations observed when compacting samples in dry conditions: compaction affects the  
250 assembly of bentonite grains but not significantly the bentonite grains themselves, the  
251 microstructure of which is governing suction.

252

253 Visual observation of two cross sections close to the bottom and to the top of the sample  
254 also showed that the pore distribution was not homogeneous along the axial direction,  
255 with more large pores and large particles observed in the bottom of the sample. This is  
256 related to segregation effects during sample preparation. When pouring the powder into  
257 the compaction mould, the largest grains first tumble resulting in more macropores at  
258 the bottom. Indeed, this problem could be solved by using wet mixture with bentonite  
259 grains sticking along sand grains and then reducing segregation.

260

261 Figure 4 provides further information about the interpretation of the PSD curves  
262 provided in Figure 2, in which one observes that mercury intrusion started in

263 macropores with an entrance diameter as large as 330  $\mu\text{m}$  and an average diameter of the  
264 corresponding pore population equal to 22  $\mu\text{m}$ . This size is compatible with the inter-  
265 grain pores observed in  $\mu\text{CT}$  in Figure 3. Given that the sample tested by MIP was  
266 extracted from the specimen centre where bentonite grains are more closely compacted  
267 together, this pore population is mainly related to the pores located between the sand  
268 grains. The clear presence of the pore inter-grains population in the PSD curve confirms  
269 the interconnection of inter-grains pores along the grain skeleton that was suspected  
270 from the  $\mu\text{CT}$  image and commented above. Hence, the bimodal porosity that is  
271 generally related to inter-aggregate and intra-aggregate pores in soils compacted dry of  
272 optimum (Ahmed et al 1974, Delage et al. 1996, Romero et al. 1999) is due here to the  
273 nature of the mutual arrangement of sand and bentonite powder grains. Indeed, the  
274 inter-aggregate average entrance pore diameter observed by Delage et al. (1996) in a  
275 compacted low plasticity Jossigny silt is 8  $\mu\text{m}$  whereas Romero et al. (1999) detected  
276 inter-aggregates pores close to 2  $\mu\text{m}$  in compacted Boom clay. The significantly larger  
277 average entrance pore diameter detected here is too large to be representative of inter-  
278 aggregate pores as in the case of standard compacted soils. It is linked to the connected  
279 inter-grains pores observed along the sand skeleton in  $\mu\text{CT}$ .

## 280 Image analysis

281 Further investigation of the microstructure was carried out by using the ImageJ image  
282 analysis software. The first step was to reduce noise by applying a 3D median filter with  
283 a 1 pixel radius. Then, a segmentation of the image is needed in order to isolate pores  
284 from the other existing phases. The choice of the threshold value is made by using the  
285 “mixture modelling” plug in. The mixture modelling technique is a histogram based  
286 technique that assumes that the histogram distribution is represented by two Gaussian

287 curves. The histogram is then separated in two classes using a Gaussian model and a  
288 threshold value of 73 is then calculated as the intersection of the two Gaussians. Figure  
289 5 shows the histogram of the image on a linear (black) and logarithmic (grey) scale. The  
290 linear graph doesn't exhibit two clearly distinct peaks because the number of pore's  
291 voxels is too small compared to that of the grains. However the logarithmic graph  
292 evidences a non-symmetrical shape that indicates the presence of the smaller pore  
293 population. The two Gaussians are then fitted using the "mixture modelling" plug in as  
294 seen in the Figure 5 represented by the white (continuous and dashed) curves.

295

296 Figure 6 shows an image at different stages before segmentation: a) initial state; b) after  
297 applying a 3D median filter; c) after applying a 73 threshold value (Figure 5) and d) the  
298 segmented image. An investigation on the effect of the size of the studied volume on the  
299 calculated porosity was done. To do so, the porosity was calculated by using the  
300 histogram of the segmented image and by dividing the number of black voxels by the  
301 total number of voxels. Starting from the middle horizontal cross section of the sample  
302 at different positions (see Figure 7a), cubic volumes were selected with sides starting  
303 from 21 to 271 pixels. The side on the  $z$  direction is limited by the height of the sample  
304 (10 mm) that corresponds to a maximum thickness of 271 pixels.  $x$  and  $y$  were  
305 afterwards increased until 700 resulting in a parallelepiped volume. The porosity values  
306 calculated with respect to the considered volumes are plotted in Figure 7. The curves  
307 start with some irregularities and then stabilise. Some curves continuously and slowly  
308 increase (bottom and right in Figure 7a). These increases suggest a spatial heterogeneity  
309 of the porosity. Indeed, when the studied volume increases, porosity increases because  
310 some zones located on the side of the specimen with higher porosity are more and more

311 included into the volume. Stabilization is observed for volumes greater than  $20 \times 10^6$   
312 voxels. This volume corresponds to a cube with a side approximately equal to the height  
313 specimen i.e. 271 pixels. It can hence be considered as a Representative Elementary  
314 Volume (REV). This side size corresponds to 9 adjacent grains ( $D_{50}$  mixture = 0.9 mm  
315 = 30 voxels) in accordance with previous studies (e.g. Razavi et al., 2007).

316

317 The spatial distribution of the porosity is now investigated at different locations along  
318 the  $x$  and  $y$  directions by using volumes with heights equal to 271 pixels. The locations  
319 of the volumes sections investigated are presented in Figure 8. Figure 9 shows the  
320 variation of porosity with respect to  $x$  and  $y$  (volumes 1 to 12 in Figure 8). A clear  
321 difference is observed in the porosity values between the border ( $x, y = 160$  and  $1515$ )  
322 and the centre. A significant difference in porosities is also noticed for  $x$  (or  $y$ ) varying  
323 between 1244 and 1515 while  $y$  (or  $x$ ) equals 925. It corresponds to cubes 5 and 6, 11  
324 and 12 in Figure 8. The porosity variation at these points is not smooth like in the others  
325 but the slope is steep showing a sudden change in porosity. It confirms the slow  
326 increase observed in the REV study (Figure 7b), since it corresponds to the same  
327 positions (right for cubes 5 and 6 and bottom for cubes 11 and 12).

328

329 The same study is now made on concentric rings having a height of 271 pixels and a  
330 width of 167 pixels starting from the centre (radius from 0 to 167) to the contour (radius  
331 from 501 to 835, see Figure 10).

332

333 The porosities obtained with respect to the radius are presented in Figure 11. The  
334 smallest detail that can be identified in the  $\mu$ CT images is defined by the size of the

335 voxel (30  $\mu\text{m}$ ). The porosity estimated by image analysis only concerns pores with a  
336 diameter larger than 3 voxels (partial volume effect). The value of the cumulated  
337 porosity of pores larger than 90  $\mu\text{m}$  from the PSD curves (Figure 2) is 0.0108. This  
338 value is in the same range as that obtained by image analysis in the central part of the  
339 sample where MIP was performed (0.0135 porosity for radius smaller or equal to 334  
340 voxels). This confirms that, in spite of some limitation related to the voxel size,  $\mu\text{CT}$  is  
341 an interesting complementary method to MIP for estimating the macropores and giving  
342 additional information on the spatial distribution of the porosity.

## 343 **Discussion**

344 Most of the investigations carried out up to now on compacted soils in laboratory have  
345 been obtained from the combined use of MIP and SEM on freeze dried samples. It is  
346 commonly accepted that the bimodal pore distribution observed in fine-grained soils  
347 compacted dry of optimum is due to the aggregate microstructure of compacted soils,  
348 with macro-pores being inter-aggregate pores and micropores being intra-aggregate  
349 pores (Ahmed et al. 1974, Delage et al. 1996). In such soils, it has also been  
350 demonstrated that compaction has little effect on the sample's suction (Li et al., 1995,  
351 Gens et al., 1995, Tarantino and De Col, 2009), given that suction is governed by intra-  
352 aggregate phenomena (mainly the adsorption of water in the clay fraction, Romero et  
353 al., 1999) whereas the change in density are due to the compression of larger inter-  
354 aggregate pores with little effect on the aggregates (Delage, 2009).

355

356 Compacted bentonites and sand-bentonite mixtures are a special case of compacted  
357 soils. They are obtained by compacting powder grains that may have, as observed here,



358 a size significantly larger than the aggregates in laboratory compacted soils. Compacted  
359 bentonites and sand-bentonite mixtures also have quite large values of suction  
360 (76.6 MPa here). In the mixture studied here, the powder grains of bentonite (1.2 mm  
361 average diameter) are larger than the sand grains (0.6 mm average diameter). As in  
362 standard compacted soils, suction appeared here not to change significantly before and  
363 after compaction (73.3 MPa in the powder before and 76.6 MPa after compaction),  
364 confirming a well known trend in compacted soils.

365

366 Compared to MIP and SEM,  $\mu$ CT provided more information on the larger scale  
367 structure, going from the size of the grains (sand and bentonite, 1 mm order of  
368 magnitude) to the size of the specimen (50 mm), i.e. on a scale much larger and more  
369 representative than both MIP and SEM. Interestingly,  $\mu$ CT has been able to distinguish  
370 clearly enough the bentonite from the sand grains within the compacted mixture, in  
371 spite of small difference in density. Thus,  $\mu$ CT provided interesting complements to  
372 MIP and SEM observations, including granules connectivity and distribution through  
373 the sample.

374

375 An important point is that  $\mu$ CT observation confirmed that the bentonite grains seem to  
376 have not been crushed during compression. They still have an average size compatible  
377 with macroscopic grain size measurements and an angular shape with no clear  
378 appearance of local grain breakage. This finding is in agreement with the small variation  
379 of suction before and after compaction, as suspected from studies in laboratory  
380 compacted soils. Suction didn't change because the grains have not been significantly  
381 affected by compression, even under 25.5 MPa.  $\mu$ CT also provides clear information

382 about the mutual arrangement of sand and bentonite grains within the mixture, showing  
383 that it is not really homogeneous with some continuity observed between sand grains  
384 with the bentonite grains (65% in mass) located within the sand grains assembly (35%  
385 in mass). The common hypothesis of having sand grains isolated within a clay matrix is  
386 not confirmed and some continuity of the inter-grains porosity is suspected. In the  
387 looser zone around the sample, some large pores with a size comparable to that of the  
388 inter-grains pores have also been observed between the bentonite grains. These pores,  
389 that are likely to exist in less densely compacted sand-bentonite samples, are no longer  
390 observed in the denser central zone in which the compaction stress appears to be high  
391 enough to aggregate the bentonite grains together.

392

393 In spite of the precautions taken during sample preparation,  $\mu$ CT clearly evidenced that  
394 the compacted sand-bentonite mixture was heterogeneous with looser zones all around  
395 the sample close to the ring in which compaction was made. This difference could have  
396 been better estimated by running MIP tests at different distances from the centre to  
397 compare them with the data of Figure 2 obtained in the denser central area. Some  
398 segregation probably resulting from the difference in density between the bentonite and  
399 the sand grains was also detected by  $\mu$ CT. Again, the use of a wetter mixture as  
400 commented above could have helped achieving better heterogeneity with a mixture less  
401 sensitive to gravity segregation effects during pouring. The heterogeneity observed is  
402 certainly linked to the dry mixing preparation procedure used here and this conclusion  
403 should not apply to wetter mixture. Note however that bentonite grains were at  
404 equilibrium with ambient atmosphere and that this simple condition is likely to be  
405 prevailing when preparing mixtures in real disposal system. Indeed, the process of

406 mixing a wetter bentonite mixture could add some complexity to the process of  
407 preparing compacted mixtures.

408 The hydro-mechanical response of the sample should be somewhat influenced by this  
409 heterogeneity, with mechanical parameters affected by the stronger central area and the  
410 hydraulic conductivity affected by the smaller permeability around the sample.

411

## 412 **Conclusion**

413 The  $\mu$ CT investigation of the microstructure of compacted bentonite/sand mixture  
414 samples (65/35% respectively) provided interesting complementary features at larger  
415 scale that could not have been identified by using MIP.  $\mu$ CT allowed further  
416 observation of the mutual arrangement of bentonite and sand grains. The commonly  
417 reported assumption that sand grains are present as isolated particles regularly scattered  
418 within a clay matrix has not been observed. The results of this study showed that some  
419 continuity could be observed between the sand grains with interconnected inter-grains  
420 large pores that were also detected by MIP. Note that the large pore population  
421 evidenced by MIP comprises pores located between sand grains and also between  
422 bentonite grains as well, indicating some connectivity in this pore population.

423

424 A study on the effect of the size of the REV demonstrated that it was necessary to  
425 consider a cubic volume with a size of 271 voxels (around 9 grains). The sample  
426 heterogeneity due to compaction with a larger porosity around the sample was  
427 quantified by carrying a 3D estimation of porosity by using image analysis.

428

429 The interconnection of inter-grains pores evidenced here could have some effects during  
430 the hydration of the bentonite/sand mixture, at least in the vapour phase, since it could  
431 facilitate the propagation of vapour within the mixture through connected inter-grains  
432 pores. The question as to whether this porosity remains interconnected and not clogged  
433 when bentonite grains start being hydrated remains. It could be examined by conducting  
434 a similar investigation at various degrees of hydration. This complementary study could  
435 also allow further understanding about achieving better heterogeneity by using wetter  
436 sand bentonite mixture less prone to gravity segregation thanks to some possible  
437 sticking of bentonite grains to the sand grains.

438 **References**

- 439
- 440 Agus S.S. and Schanz T. (2005). Effect of shrinking and swelling on microstructures  
441 and fabric of a compacted bentonite-sand mixture. *Proceedings of the International*  
442 *Conference on Problematic Soils*, Cyprus, 32, 543 – 550.
- 443 Ahmed S., Lovell C. W. and Diamond S. (1974). Pore sizes and strength of compacted  
444 clay. *J. Geotech. Eng. Div. Am. Soc. Civ. Eng.*, GT4, 407– 425.
- 445 Carminati A., Kaestner A., Hassanein R. and Koliji A. (2006). Hydraulic properties of  
446 aggregate-aggregate contacts. In: *Desrues J, Viggiani G, Bésuelle P (eds.) Advances*  
447 *in X ray tomography for geomaterials*. ISTE Ltd, London, UK, 325–331
- 448 Comina C., Foti S., Musso G. and Romero E. (2008). EIT oedometer: an advanced cell  
449 to monitor spatial and time variability in soil. *ASTM Geotechnical Testing Journal* 31  
450 (5), 404–412.
- 451 Cui Y.J., Loiseau C. and Delage P. (2002). Microstructure changes of a confined  
452 swelling soil due to suction controlled hydration. In *Unsaturated soils: proceedings of*  
453 *the Third International Conference on Unsaturated Soils, UNSAT 2002*, 10-13 March  
454 2002, Recife, Brazil, volume 2, 593-598.
- 455 Delage P. (2009). Compaction behaviour of clay: discussion. *Géotechnique* 59 (1), 75-  
456 76.
- 457 Delage P., Marcial D., Cui Y.J. and Ruiz X. (2006). Ageing effects in a compacted  
458 bentonite: a microstructure approach, *Géotechnique* 56(5):291–304.
- 459 Delage P., Audiguier M., Cui Y.J., and Howat M. (1996). The microstructure of a  
460 compacted silt. *Canadian Geotechnical Journal*, 33 (1): 150-158.
- 461 Gebrenegus T., Tuller M. and Muhuthan B. (2006). The application of X-ray computed  
462 tomography for characterisation of surface crack networks in bentonite-sand mixtures.  
463 In: *Desrues J, Viggiani G, Bésuelle P (eds) Advances in X-ray tomography for*  
464 *geomaterials*. ISTE Ltd, London, UK: 207–212
- 465 Gens A., Alonso E.E., Surlol J. and Lloret A. (1995). Effect of structure on the  
466 volumetric behaviour of a compacted soil. *Proc. 1st Int. Conf on Unsaturated Soils*  
467 *UNSAT' 95* 1, 83-88.
- 468 Karnland O., Olsson S. and Nilsson U. (2006). Mineralogy and sealing properties of  
469 various bentonites and smectite-rich clay material. *SKB report*.
- 470 Kawaragi C., Yoneda T., Sato T. and Kaneko K. (2009). Microstructure of saturated  
471 bentonites characterized by X-ray CT observations. *Engineering Geology* 106, 51–57
- 472 Ketcham R. A. and Carlson W. D. (2001). Acquisition, optimization and interpretation  
473 of X-ray computed tomographic imagery : applications to the geosciences. *Computers*  
474 *& Geosciences* Vol. 27, 381-400.
- 475 Koliji A., Carminati A., Kaestner A., Vulliet L., Laloui L., Fluehler H., Vontobel P. and  
476 Hassanein R. (2006). Experimental study of flow and deformation in aggregated soils  
477 using neutron tomography. In: *Desrues J, Viggiani G, Bésuelle P (eds). Advances in*  
478 *X-ray tomography for geomaterials*. ISTE Ltd, London, UK: 341–348
- 479 Komine H. and Ogata N. (1999) Experimental study on swelling characteristics of sand-  
480 bentonite mixture for nuclear waste disposal. *Soils and Foundations* 39 (2): 83-97.

- 481 Kozaki T., Suzuki S., Kozai N., Sato S. and Ohashi H. (2001). Observation of  
482 microstructures of compacted bentonite by microfocuss X-ray computerized  
483 tomography (Micro-CT). *Journal of Nuclear Science Technology* 38 (8), pp. 697–699
- 484 Li Z.M. (1995). Compressibility and collapsibility of compacted unsaturated loessial  
485 soils. *Proceedings of the 1<sup>st</sup> International Conference on Unsaturated Soils UNSAT'*  
486 95 1, 139-144, Paris, Balkema, Rotterdam.
- 487 Lloret A., Villar M.V., Sanchez M., Gens A., Pintado X. & Alonso E.E. 2003.  
488 mechanical behaviour of heavily compacted bentonite under high suction changes.  
489 *Géotechnique* 53 (1), 27-40.
- 490 Madsen FT. (1998). Clay mineralogical investigations related to nuclear waste disposal.  
491 *Clay Minerals*, 33(1):109–129.
- 492 Montes-H G. (2002) Etude expérimentale de la sorption d'eau et du gonflement des  
493 argiles par microscopie électronique à balayage environnementale (ESEM) et analyse  
494 digitale d'images. *PhD Thesis*. Université Louis Pasteur, Strasbourg, France.
- 495 Mukunoki T., Otani J., Maekawa A., Camp S., Gourc J.P. (2006). Investigation of crack  
496 behaviour on cover soils at landfill using X-ray CT. In: *Desrues J, Viggiani G,*  
497 *Bésuelle P(eds) Advances in X-ray tomography for geomaterials*. ISTE Ltd, London,  
498 UK: 213–219.
- 499 Pusch, R. (1979). Highly compacted sodium bentonite for isolating rock-deposited  
500 radioactive waste products. *Nucl. Technol*, United States. 45(2):153-157.
- 501 Rasband W.S. (1997-2012). ImageJ, U. S. National Institutes of Health, Bethesda,  
502 Maryland, USA, <http://imagej.nih.gov/ij/>.
- 503 Razavi M.R., Muhunthan B. and Al Hattamleh O. (2007) Representative elementary  
504 volume analysis using X-ray computed tomography, *Geotechnical Testing Journal* 30  
505 (3), 212-219.
- 506 Rolland S., Stemmelen D., Moyne C. and Masrouri F. (2003). Transfert hydrique dans  
507 un sol argileux gonflant non saturé: influence du confinement. *Revue Française de*  
508 *Géotechnique* 104, 21–35
- 509 Rolland S., Stemmelen D., Moyne C. and Masrouri F. (2005). Experimental hydraulic  
510 measurements in an unsaturated swelling soil using the dual-energy gamma-ray  
511 technique. *Proceedings of International Symposium on Advanced Experimental*  
512 *Unsaturated Soil Mechanics, Trento, Italy*. In: Tarantino A, Romero E, Cui YJ (eds)  
513 *Advanced experimental unsaturated soil mechanics*. Taylor, Francis Group, London,  
514 305–310.
- 515 Romero E., Gens A., and Lloret A. (1999). Water permeability, water retention and  
516 microstructure of unsaturated compacted Boom clay. *Engineering Geology*, 54 (1-2):  
517 117-127.
- 518 Tang A.M., Cui Y.J. and Le T.T. (2008). A study on the thermal conductivity of  
519 compacted bentonites. *Applied Clay Science*; 41 (3-4):181-189.
- 520 Tarantino A. and De Col E. (2008). Compaction behaviour of clay. *Géotechnique* 58  
521 (3): 199-213.
- 522 Van Geet M., Volckaert G. and Roels S. (2005). The use of microfocuss x-ray computed  
523 tomography in characterising the hydration of a clay pellet/powder mixture. *Applied*  
524 *Clay Science*, 29 (2):73–87, 2005.

- 525 Villar M.V. and Lloret A. (2001). Variation in the intrinsic permeability of expansive  
526 clays upon saturation. *In: Adachi K., Fukue M. (eds) Clay Science for Engineering.*  
527 *A.A. Balkema, Rotterdam, 259-266.*
- 528 Yong R.N., Boonsinsuk P. and Wong G. (1986). Formulation of backfill material for a  
529 nuclear fuel waste disposal vault. *Canadian Geotechnical Journal*, 23(2):216–228.
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- 531

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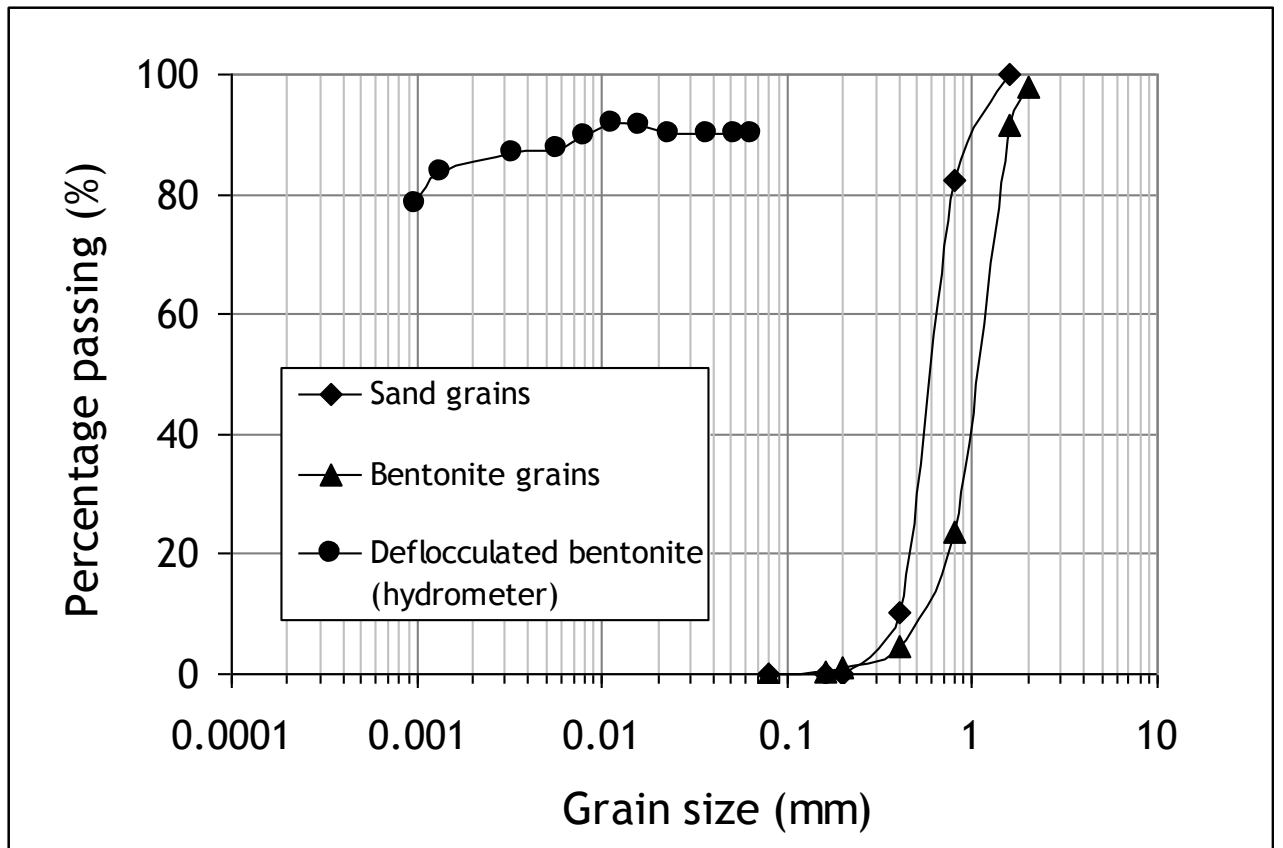


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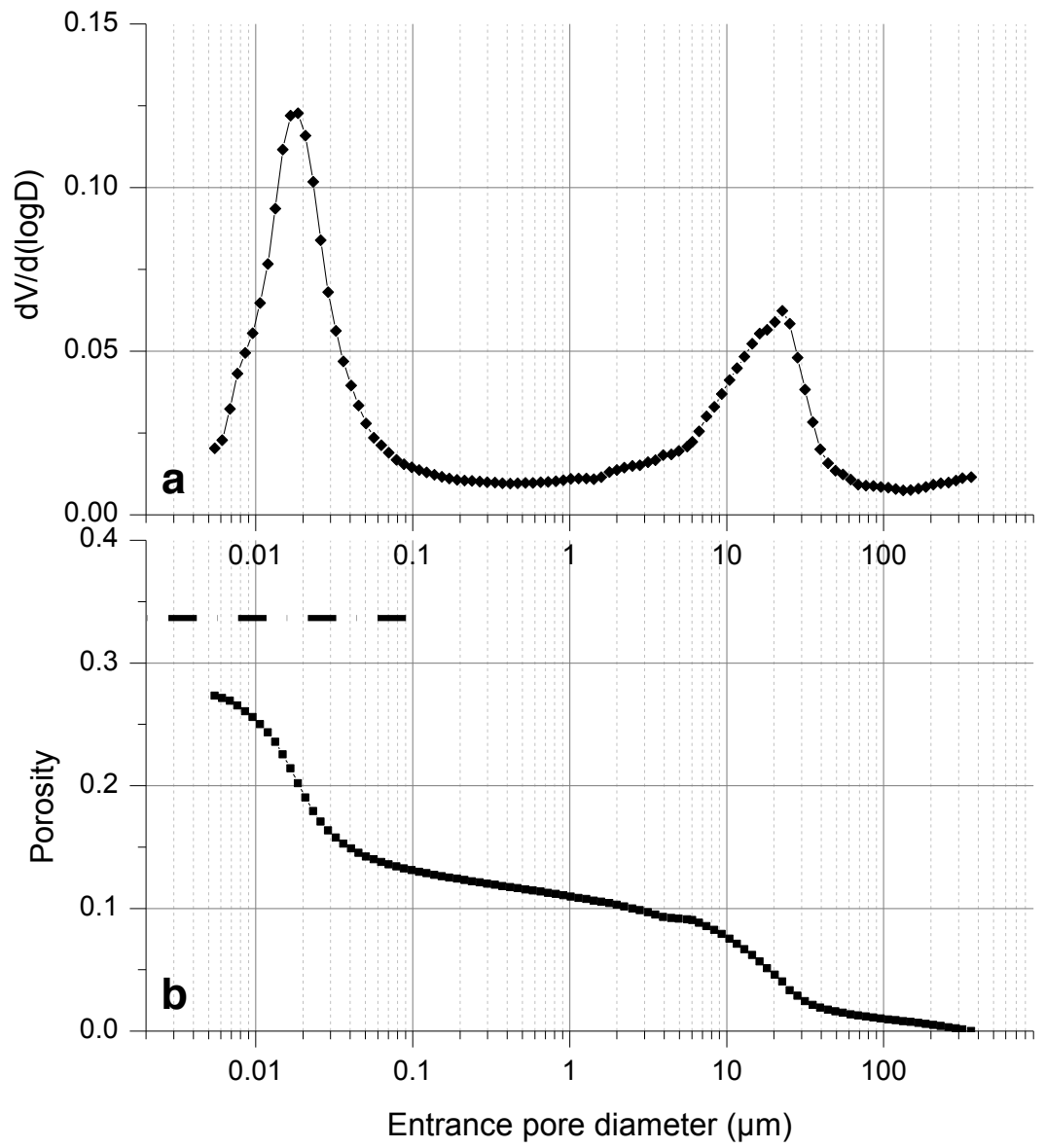


Figure 2. (a) Pore size distribution curve and (b) Cumulative porosity curve.

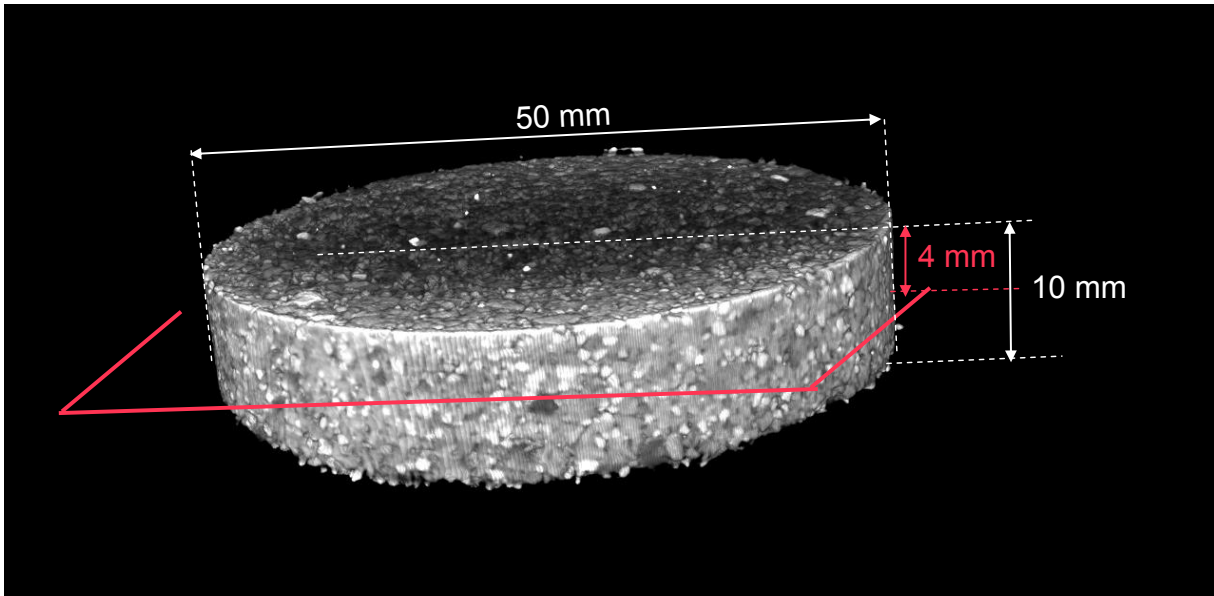


Figure 3. The 3D reconstructed image of the sample with the position of the cross section considered.

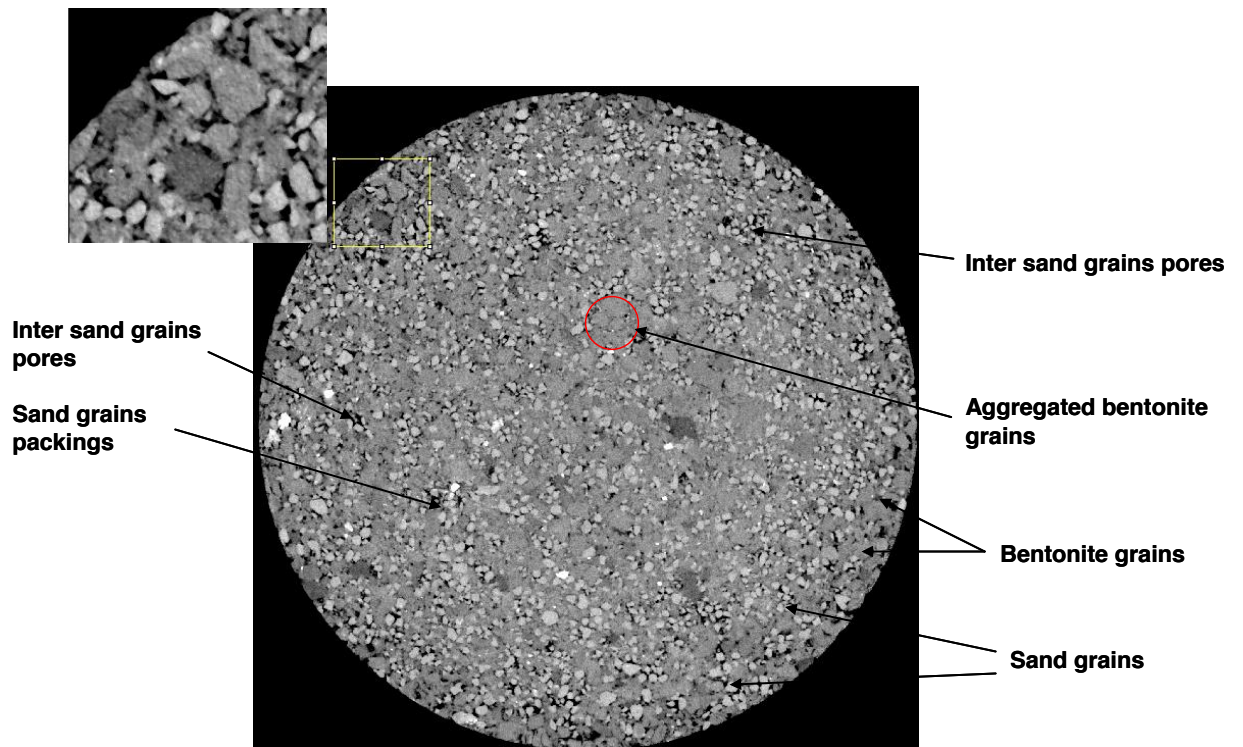


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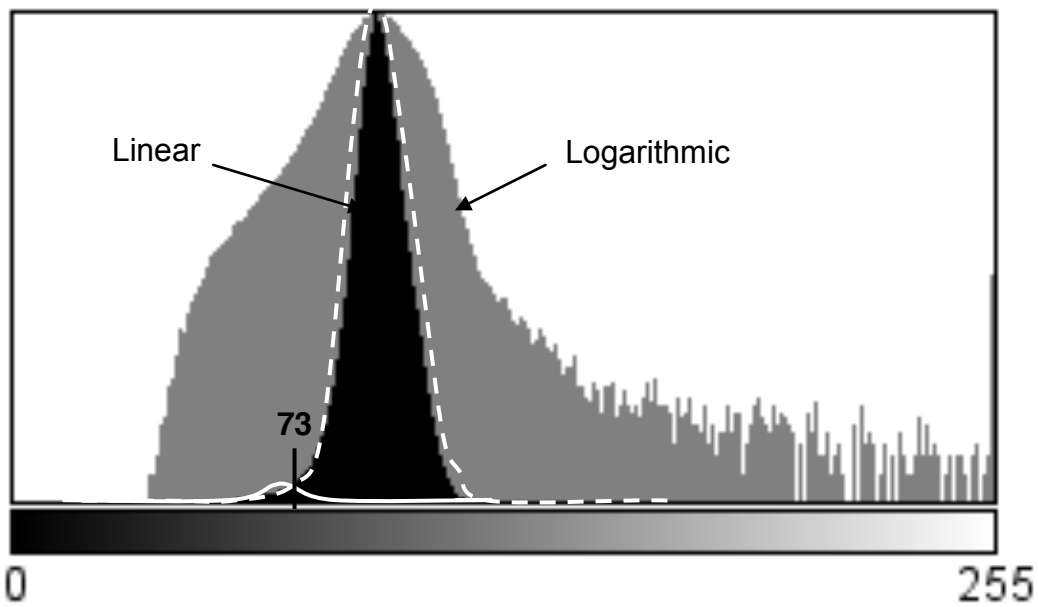


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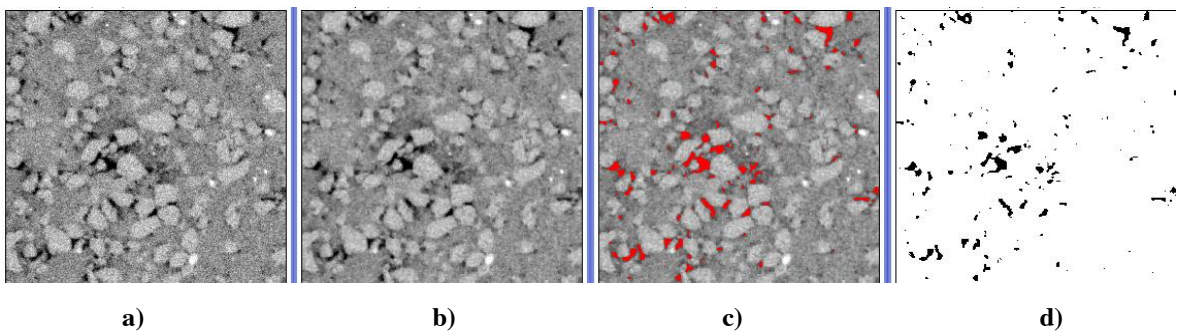


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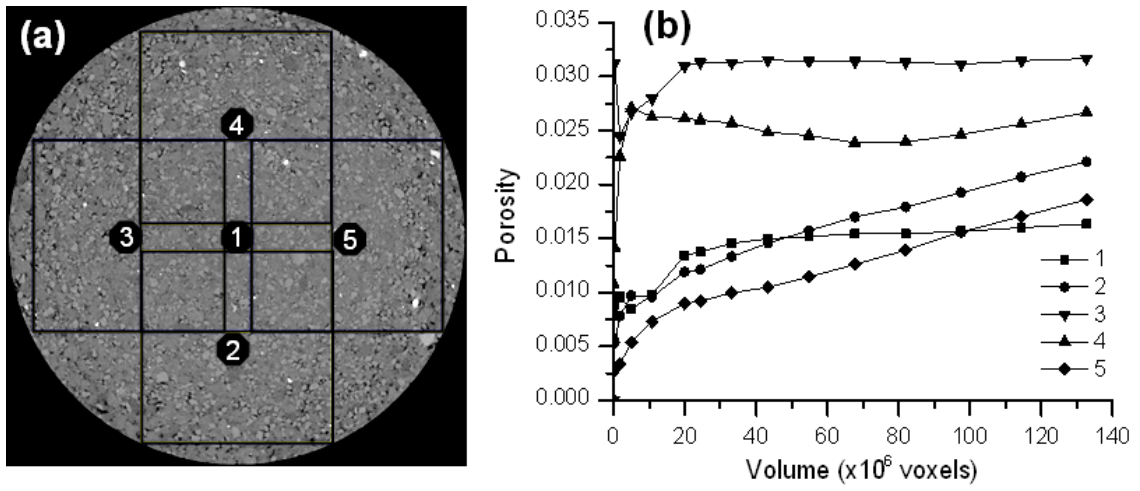


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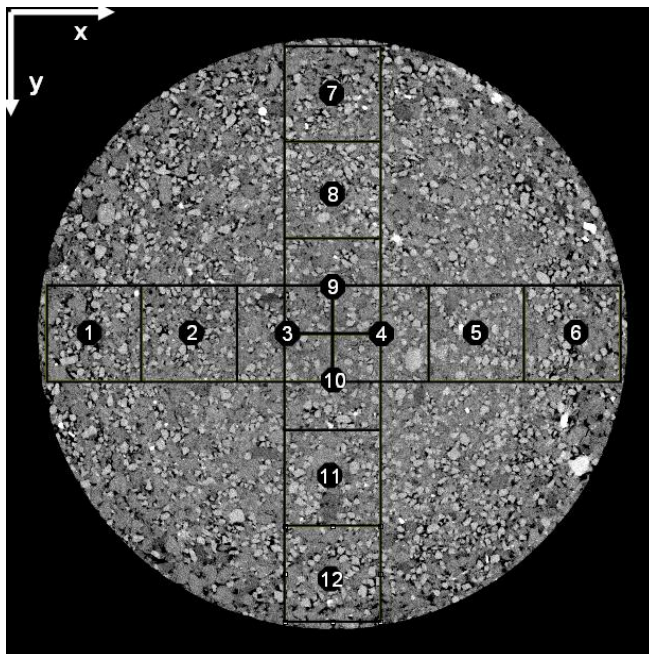


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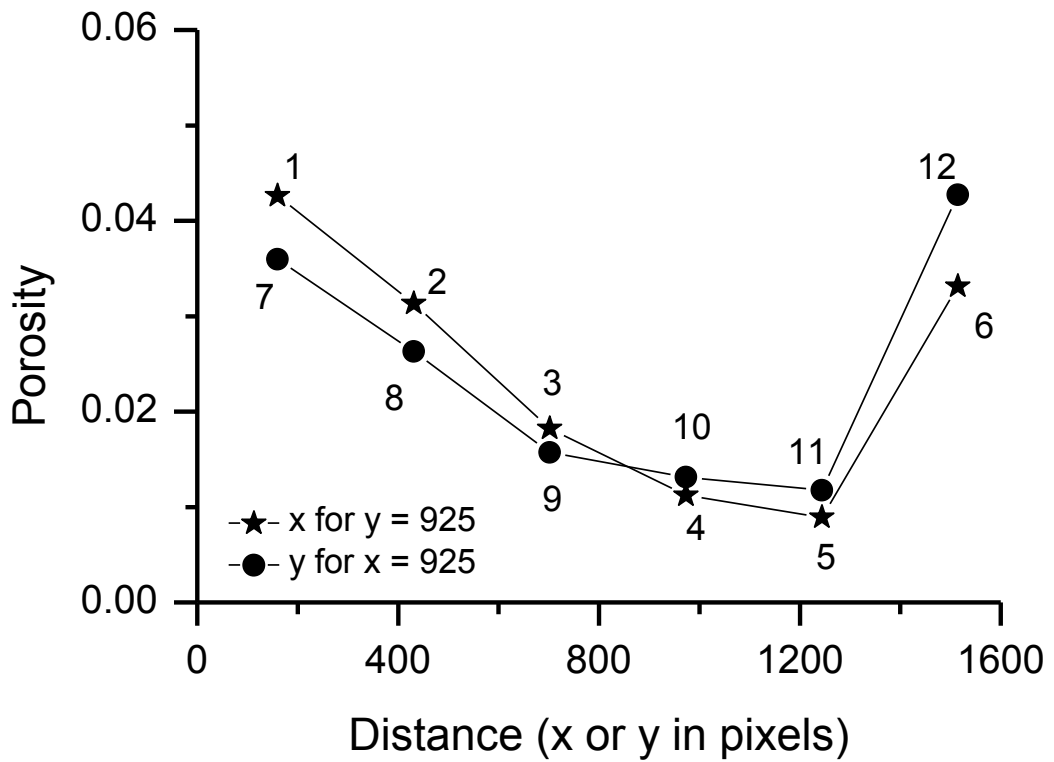


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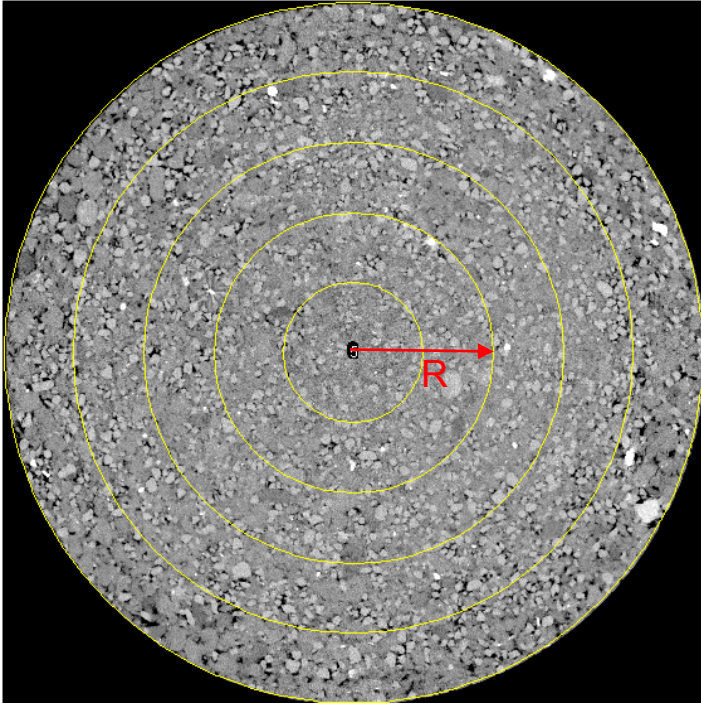


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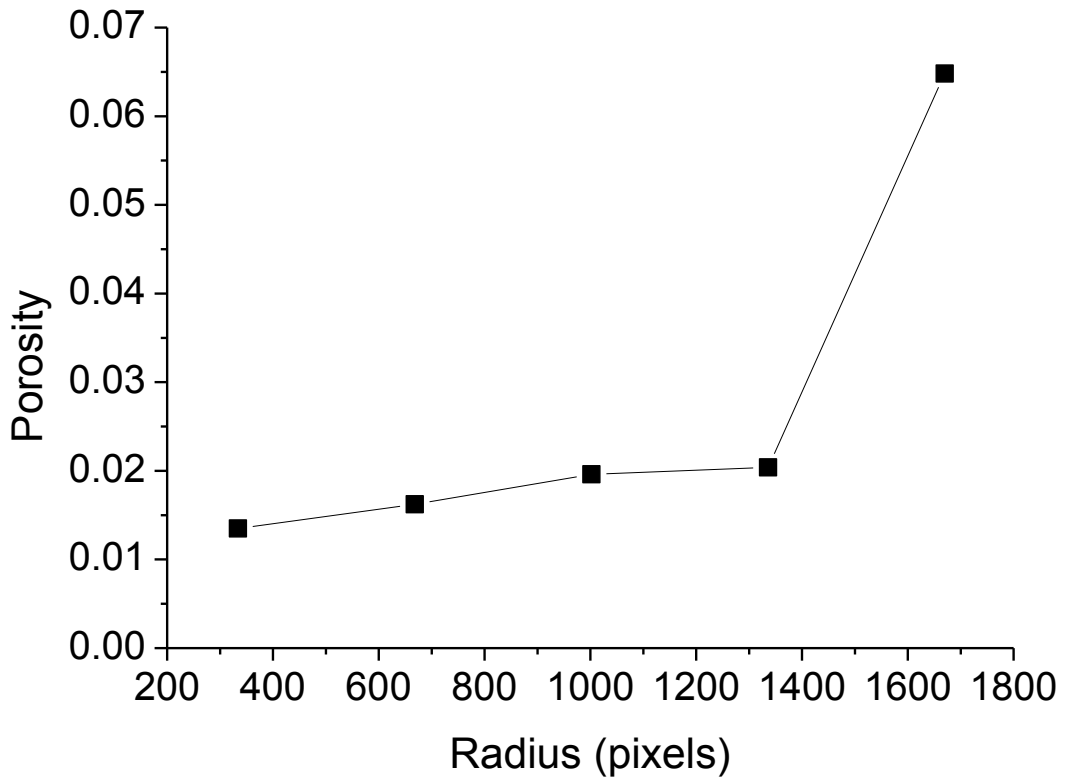


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