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## In-depth characterisation of a mixture composed of powder/pellets MX80 bentonite

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Abstract: Mixtures made up of bentonite powder and pellets are a possible candidate for 9 making sealing plugs used in deep radioactive waste disposal due to their low permeability, 10 high swelling capacity, favourable properties with respect to radionuclide retention and 11 operational advantages in terms of placement in situ, that is much easier than that of pre-12 compacted bricks of bentonite/sand mixture. It is therefore essential to better understand their 13 hydro-mechanical behaviour to optimize the design of the repository. In this context, the 14 French Institute for Radiation Protection and Nuclear Safety (IRSN) has launched the 15 16 SEALEX project (SEALing performance EXperiments) in which this work has been conducted. 17

Once the initially heterogeneous unsaturated powder/pellet (80/20) MX80 bentonite mixture 18 is put in place, these sealing materials will be subject to coupled hydro-mechanical loadings: 19 hydration due to the infiltration of pore water from the natural barrier and mechanical 20 confinement resulting from the engineered barriers. The present work focuses on the different 21 scales of the material: at the macroscopic scale, it is characterized by a heterogeneous 22 23 distribution of pellets and powder of bentonite; at the microscopic scale, it is studied by several techniques (MIP, µ-CT observations and SEM). From MIP results, a typical bimodal 24 distribution was found for both pellet and powder. From µ-CT and SEM observations, a 25 heterogeneity was revealed in the internal structure of the pellet: heterogeneous density 26 distribution of the clay minerals and presence of several high density elements. 27

Keywords: heterogeneous pellet/powder bentonite mixture; sealing plug; radioactive waste
 disposal; microstructure; mercury intrusion porosimetry; X-ray computed microtomography;
 scanning electron microscopy.

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#### **1. Introduction**

In the French concept of deep geological disposal for High Level and Intermediate Level Long Lived radioactive Wastes (HLW & ILLLW), the wastes are emplaced within large diameter boreholes (HLW) and galleries (ILLLW) excavated at great depths in a lowpermeability host-rock (Callovo-Oxfordian argillite, ANDRA, 2005). Sealing of theses underground works to prevent potential pathways for water, gas and radionuclides migration is one of the key points to ensure the long-term safety of the repository.

Mixtures made up of bentonite powder and pellets are a candidate sealing material for deep 44 45 underground radioactive waste repositories. In addition to their low permeability, high swelling capacity and high radionuclide migration retardation properties, powder/pellet 46 mixtures have obvious operational advantages in terms of emplacement and reduction of the 47 gaps between the rock and the seal. Once installed in the repository, powder/pellet mixtures 48 will be submitted to coupled hydro-mechanical loadings comprising hydration due to the 49 50 infiltration of pore water from the host rock and mechanical confinement resulting from the constrained volume condition imposed in the galleries. It is therefore essential to better 51 understand their hydro-mechanical behaviour when assessing the overall repository safety. 52

In this context, and as part of the overall IRSN (French Institute for Radiological Protection and Nuclear Safety) R&D program that provides scientific background for its expertise on disposal safety, the SEALEX (SEALing performance Experiments) project was launched to specifically focus on long-term performance of sealing systems. The SEALEX project is dedicated (i) to test the long-term hydraulic performance of sealing systems in normal conditions for different core compositions (mixtures composed of bentonite pellets with bentonite powder or of sand with bentonite powder) and for different configurations (pre-

compacted bentonite blocks or bentonite compacted in situ), (ii) to quantify the impact of 60 61 construction joints (also called technological voids) on the hydraulic properties of the sealing system, and (iii) investigate the concept of robustness by considering altered scenarios, such 62 as an incidental decrease of the swelling pressure (for instance originating from the failure of 63 the confining plugs). This project relies on a series of *in situ* experiments emplaced in IRSN's 64 Underground Research Laboratory (URL) at Tournemire located in a Mesozoic sedimentary 65 66 basin on the western edge of the French Causses (South France) (Mokni & Barnichon., 2016). To install the experiments, horizontal boreholes  $(0 \pm 2^{\circ})$ , with a 60 cm diameter and 540 cm 67 long have been excavated from recent drifts (2008). Each experiment consists of a bentonite-68 69 based core mechanically confined at both ends corresponding to a generic seal mock-up except in terms of saturation that will be an artificial one carried out by water injection 70 through porous filters installed at both ends of the core (Barnichon et al., 2012; Mokni & 71 72 Barnichon., 2016). Various materials are being considered as seals in the SEALEX project. The present work focuses on a mixture made up of MX80 bentonite powder and pellets with a 73 proportion of 20/80 in dry mass. 74

The use of high-density bentonite pellets combined with bentonite powder has also been proposed in Salo & Kukkola (1989), Dereeper et al. (2001), Ab (2002). A characterisation of the fundamental properties of the MX80 bentonite pellets used in the Prototype Repository test is reported in Sugita et al. (2005). At a larger scale, a series of infiltration tests on a 50/50% FoCa bentonite pellet/powder mixture compacted at different dry densities were performed by Imbert & Villar (2006) to investigate the swelling capacity of the material.

It is well documented that the macroscopic behaviour of expansive soils is related to its microstructure (e.g. Alonso et al., 2011; Gens & Alonso, 1992). For this reason, several investigations have been carried out on the microstructure of bentonite-based materials by using several methods including Mercury Intrusion Porosimetry (MIP) and/or Scanning Electron Microscopy (SEM) (Wang et al., 2012, 2013, 2014; Saba et al., 2014; Romero et al., 2011). There is less available data on pellets and powder-pellets mixtures. Hoffmann (2005) carried out MIP tests on samples of pellets of FEBEX bentonite under different conditions and observed a bimodal distribution on a single pellet with a dry unit mass  $\rho_d = 1.95 \text{ Mg/m}^3$ . For a non-compacted pellet mixture, a trimodal distribution was identified with two populations of pores corresponding to the intra and inter-aggregate pores and the third one related to the inter-pellet pores.

The microstructure investigation techniques require a preliminary dehydration of the samples, 92 often carried out by freeze-drying. Due to the size of the sample investigated, these techniques 93 94 provide local observations of a part of millimetric samples. These results may fruitfully be complemented by the use of X-ray computed microtomography (µ-CT), a non-destructive 3D 95 technique that provides high-resolution observations of samples at a larger scale without any 96 pre-treatment. The changes in microfabric during hydration under constant volume conditions 97 of a 50/50 pellet/powder mixture of FoCa clay at a dry unit weight of 1.36 Mg/m<sup>3</sup> were 98 observed by Van Geet et al. (2005) using µ-CT. The significant changes of the pellet/powder 99 mixture during hydration could be observed but the technique did not allow investigating the 100 microstructure of an elementary pellet itself because the resolution of the images obtained was 101 not enough in order to study the material at this scale. 102

103 This paper deals with an experimental program aiming to study the microstructural features of 104 a pellet/powder mixture at various scales by using  $\mu$ -CT observations coupled to MIP and 105 SEM investigations. This mixture will afterwards be used to develop a mock-up test in the 106 laboratory, to simulate SEALEX *in situ* tests at reduced scale (1/10) by considering a column 107 of 60 mm in diameter and 120 mm in height.

#### **2. Materials and methods**

#### 109 **2.1. Materials**

The materials investigated are on the one hand the basic pellet made of MX80 bentonite and 110 on the other hand a mixture of MX80 bentonite powder and pellets at a proportion of 20/80 in 111 dry mass (Figure 1). The bentonite used comes from Wyoming, USA, and was provided by 112 the Laviosa-MPC company under the commercial name Expangel (Expangel SP7 for pellets 113 and Expangel SP32 for the powder). To make things clear, the name MX80 will however be 114 used here. The MX80 bentonite has a high smectite content (80%) with some inclusions of 115 non-clayey minerals (see Table 1) and a cation exchange capacity (CEC) of 98 meq/100g. The 116 major exchangeable cation is Na<sup>+</sup>, with a value of 52 meq/100g (Table 2). The liquid limit is 117 560%, the plastic limit is 53% and the unit mass is of 2.77 Mg/m<sup>3</sup> (Saba et al., 2014, Table 3). 118 Pellets were industrially produced in Laviosa-MPC company by instantaneously compacting a 119 120 powder of MX80 bentonite in a mould of 7 mm of diameter and 7 mm of height (Laviosa Minerals). The fabrication was done at water content w = 5% - 7% and at dry unit mass  $\rho_d =$ 121 1.998 Mg/m<sup>3</sup> - 2.12 Mg/m<sup>3</sup>. Pellets were received in packages of 25 kg and stored in the 122 laboratory at 20°C. The pellet initial suction was measured in the laboratory with a chilled 123 mirror dew point tensiometer (Decagon WP4), providing a value s = 132.4 MPa at initial 124 water content w = 7.25% (determined after drying in the oven at 105°C during 24h). More 125 details about the initial properties of the pellets and the powder are presented in Table 3. 126

Figure 2 shows a typical pellet with a radius of 7.14 mm and a height of 7.27 mm. It appears that the pellet has a quasi-cylindrical shape with two spherical poles on top and bottom. The lengths measured in the laboratory were larger than those just after the fabrication (7 mm), indicating swelling due to hydration in the course of storage. Indeed, higher water contents (7.25% - 6.69%) were measured on pellets located at the top of the bucket stored in the laboratory, compared to that located at the bottom (6.06% - 6.00%). The MX80 bentonite powder was produced by crushing pellets. The characteristics at initial state is presented in Table 3. Compared to the fabrication value of water content (between 5% and 7%), a value of 3.17% was found in the laboratory (after drying at 105°C for 24h) corresponding to an initial suction s = 190.9 MPa, measured with a chilled mirror dew point tensiometer (Decagon WP4). The grain size distribution of the powder, obtained by dry sieving is presented in Figure 3 together with the size of a pellet. The average diameter is  $D_{50} = 0.65$  mm.

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#### 141 **2.2. Testing procedures**

#### 142 Mercury intrusion porosimetry (MIP)

The pore size distribution of both pellet and powder of MX80 bentonite was obtained on 143 freeze dried samples using an Autopore IV 9500 mercury intrusion porosimeter 144 (Micromeritics) that operates at a maximum pressure of 230 MPa. Instantaneous freezing was 145 carried out by plunging small samples (a pellet of bentonite) in slush nitrogen (-210°C) 146 147 obtained by previously submitting it to vacuum (Delage et al., 1996). In such conditions, there 148 is no nitrogen boiling around the samples when plunging them into nitrogen, resulting in an optimized quick freezing and good microstructure preservation. The pore size distribution was 149 obtained assuming parallel, cylindrical nonintersecting pores of different radii, using the 150 Autopore IV 9500 V1.09 standard software package. The total void ratio was obtained by 151 standard methods and compared to the total volume of mercury intruded into the sample at a 152 pressure of 230 MPa. 153

154 *X-ray computed microtomography* ( $\mu$ -*CT*)

The μ-CT scans were carried out using an "Ultratom" microtomograph (RX Solutions,
France). Images were reconstructed using the software Xact (RX Solutions). The source is a

microfocus X-ray tube Hamamatsu L10801 and the imager is a Paxscan Varian 2520V
(1960x1536 square pixels 127μm in size).

Several µ-CT observations were carried out on a pellet of bentonite at initial state and on the 159 160 powder/pellet MX80 bentonite mixture with a proportion of 80/20 in dry mass. For a single pellet of bentonite, the X-ray source parameters were 80 kV and 40 µA; the voxel size was 161 4.4 µm. The samples were scanned using 1440 projections equally spread on 360°. After 162 reconstruction, 1292 horizontal slices were calculated (16bit images; 1644x1292 pixels). For 163 the powder/pellet mixture, the X-ray source parameters were 100 kV and 25  $\mu$ A; the voxel 164 size was 59 µm. The mixture was scanned using 2880 projections in helical mode and after 165 reconstruction 2472 horizontal slices were obtained (16 bit images, 1735x1735 pixels). 166

167 Scanning electron microscopy (SEM) and Energy-Dispersive X-ray spectroscopy (EDS)

SEM combined to EDS was performed in order to carry out a chemical characterization of a 168 pellet of bentonite at initial state. Instantaneous freezing was carried out by plunging the 169 samples in slush nitrogen (-210°C). After freeze-drying, the pellet of bentonite was carefully 170 cut at mid-height, separating the upper and the lower poles. Once cut, the specimens were 171 immerged into a resin and polished with abrasive papers with decreasing fineness from grade 172 800 to 4000 in order to obtain a smooth surface. Finally, the samples were metallized under 173 vacuum with gold and palladium. Observations in conventional SEM require a high vacuum 174 condition to allow a precise focusing of the incident electrons on the sample and to prevent 175 the emitted electrons from interacting with the atmosphere. 176

#### 177 **2.3.** The pellet/bentonite mixture

Two SEALEX performance tests with core made of MX80 bentonite powder/pellets (20/80) have been installed in Tournemire URL, in February and October 2013, to investigate the impact of core composition and conditioning on the long term performance of sealing

systems. To ensure the feasibility of injecting within a large diameter and compacting *in situ* a 181 seal made of pellet/powder mixture so as to obtain a reasonably homogeneous core with the 182 target dry unit mass of 1.49 Mg/m<sup>3</sup>, a series of preliminary full scale mock up tests were 183 carried out using a plastic tube with transparent windows to allow the observation of the 184 distribution of the mixture within the emplacement borehole. The bentonite was introduced by 185 means of an auger conveyor measuring 5 m in length and 60 mm in outer diameter. The auger 186 measures 50 mm in diameter and is driven by a 1.5 kW geared motor. The tube features a 187 loading hoper and has a flanged connection in its midpoint, so that despite its length it can be 188 easily mounted in the gallery with its back end already inserted in the borehole, as the auger 189 190 inside is flexible.

191 It is however important to prepare an homogeneous powder/pellet MX80 bentonite mixture at 192 the required target dry unit mass  $(1.49 \text{ Mg/m}^3)$  because the dry unit mass governs both the 193 saturated permeability  $(k_w)$  and the swelling pressure of the mixture (Saba et al., 2014; 194 Hoffmann et al., 2007). In this study, the challenge was to obtain a reasonably homogeneous 195 powder/pellet mixture to be placed in a reduced scale mock-up test of the SEALEX *in situ* 196 tests. This mock-up test is planned to be carried out at a scale of 1/10 of the SEALEX tests, 197 corresponding to a cylinder of 60 mm in diameter and 120 mm in height.

Three protocols were considered aiming at identifying the one that provides the best homogeneity. The first protocol consisted in filling the cell by packets corresponding to one layer of pellets spread over the base of the 60 mm diameter cylinder and by adding the corresponding amount of powder (80% pellets and 20% powder). The second protocol was comparable but involved two layers of pellets. Finally, the third protocol consisted in filling a quantity of pellets corresponding to three layers, but that has been previously mixed with the powder.

#### 206 **3. Results**

#### **3.1. X-ray computed microtomography**

208 *Case of Pellet* 

Further information about the microstructure of a pellet of bentonite at initial state (suction *s* = 132.4 MPa, water content w = 7.25% and dry unit mass  $\rho_d = 2.12 \text{ Mg/m}^3$ ) was obtained using  $\mu$ -CT observations.

Figure 4 and Figure 5 show horizontal and vertical slices, respectively, taken at different 212 positions as indicated in the photo of the pellets presented on top left in the figures. Grey 213 levels depend on the density and atomic number of the components, with darker grey levels 214 typically corresponding to lower densities and black zones to voids. Several higher density 215 elements (light grey levels) are observed in both cases. Several cracks are identified in the 216 sections located in the upper part of the pellet (horizontal sections I and II, and vertical 217 218 sections I, II and III). It can be deduced from Figure 4 that they coincide with the upper spherical part, close to the contact with the cylindrical section. Such fissures are not observed 219 on the same zone at the bottom of the pellet. 220

Further information about the fissure network is obtained by a 3D rendering of the surfaces defined by the cracks within a region of interest of the pellet at initial state presented in Figure 6. Cracks are defined as the voxels with a grey level below some threshold. It is observed that crack networks are located on both the upper and bottom part of the pellet; however, as it was revealed by horizontal and vertical slices, there are more cracks in the upper part. The 3D rendering was performed by means of the Avizo image analysis software.

Further investigation on a single pellet and on the respective distribution of pellets and powder within the mixture was done by means of image analysis. In a first step, a region of interest (ROI) in the sample was selected (Figure 7). Then, a segmentation of the image was

made to separate the voids from the other phases (i.e. the pellets and the powder grains). This 230 231 step was completed using the ImageJ image analysis software. This method is a variation of the IsoData algorithm (Ridler & Calvard, 1978), an iterative procedure that divides the image 232 into an object and a background by adopting an initial threshold. Then the averages of the 233 pixels at or below the threshold and pixels above are computed, the threshold is incremented 234 and the process is repeated until the threshold is larger than the composite average. Figure 7 235 236 shows the initial state with the selected ROI (a) and the segmented image (b). It is noted that such a procedure does not give access to an accurate evaluation of the actual total porosity of 237 the sample, as the result strongly depends on the selected threshold, is strongly sensitive to 238 partial volume effects and other image artefacts, and does not allow to detect the smaller 239 pores. It provides however a way to characterize, at least qualitatively, the spatial distribution 240 of the larger voids within the sample. 241

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An investigation of the porosity was conducted on a single pellet of bentonite with this 243 244 approach. The porosity was calculated for each horizontal slice obtained by µ-CT observations by dividing the number of black voxels (below some threshold, corresponding to 245 voids) by the total number of voxels within the pellet. Figure 8 and Figure 9 present the 246 calculated values of void ratio for horizontal and vertical sections respectively. It is confirmed 247 from Figure 8 that the value of void ratio increases at the upper part of the pellet (for a level 248 higher than 5 mm) because of the presence of the fissures. In Figure 9 (calculated void ratio 249 for each vertical section), the highest values of void ratio are located at both edges and in the 250 centre of the pellet. Note that this porosity corresponds to that detected by image analysis with 251 a resolution of 4.4 µm/voxel and for a particular choice of grey level threshold; hence, it 252 provides a first evaluation of the porosity associated with the cracks (see for instance the 3D 253 representation of the fissure network of Figure 6). 254

#### 256 *Case of Pellet-powder mixture*

The homogeneity of the specimens obtained by using each of the three protocols was 257 examined by using X-ray computed microtomography, the results of which are presented in 258 Figure 10. Observation of Figure 10a shows that the first protocol (1 layer) provided a 259 reasonably good homogeneity of the mixture, with regular scattering of the powder grains 260 within the pores located between the pellets. Several voids between the pellets are however 261 observed at the top of the sample. Inspection of Figure 10b shows that the homogeneity is not 262 ensured with the second protocol (2 layers) with much more inter-pellets pores that are not 263 filled with powder, particularly in the peripheral part of the specimen. Note also the presence 264 of large inter-pellets voids completely filed with powder, in the middle of the sample at 1/3265 of the height. The same problem is observed with the third protocol (Figure 10c). The target 266 dry unit mass ( $\rho_d = 1.49 \text{ Mg/m}^3$ ) was obtained with the first protocol only, while a unit mass 267 of  $\rho_d = 1.41 \text{ Mg/m}^3$  and 1.43 Mg/m<sup>3</sup> is found for the second and third protocols respectively. 268

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An investigation of the porosity was conducted on the three protocols in the same way as 270 detailed previously for a single pellet of bentonite. To this end, the porosity was estimated for 271 each horizontal slice obtained by µ-CT observations, using the same threshold for all slices. 272 Figure 11 presents the evolution of the obtained porosity along the height of the sample for 273 the three protocols. One can observe an oscillation of the value of porosity for all three 274 protocols. In the first one, the minimum and maximum void ratios are 0.18 and 0.63 275 respectively. Oscillations are more remarkable and they correspond to one layer of pellets. In 276 the second protocol, the minimum and maximum void ratio values are 0.15 and 0.33 277 respectively, oscillating around a value of 0.2. Finally, for the third protocol, void ratio goes 278 from 0.08 to 0.55, and the oscillations correspond to three layers of pellets. 279

Taking into account the target dry density, the homogeneity observed in Figure 10 and the 281 data of image analysis, the first protocol should be selected in order to prepare the 282 pellet/powder mixture samples of MX80 bentonite. Note however that the best result 283 corresponds to the second protocol according to the image analysis, because the values of 284 void ratio oscillate around a value of 0.2 for all the sections. This low fluctuation along the 285 vertical direction of the apparent average porosity within a section does however not reflect 286 the strong heterogeneity of the pore distribution within a section, which is observed in the 287 vertical slice represented in Figure 12b. Furthermore, the target dry density (1.49 Mg/m<sup>3</sup>) was 288 only obtained with the first protocol. 289

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#### **3.2.** Mercury intrusion porosimetry

Figure 12 shows the results of MIP tests carried out on a pellet (w = 6.7%, s = 138.4 MPa) and on the bentonite powder (w = 3.1%, s = 190.9 MPa). The cumulative curve (Figure 12a) shows that the final value of intruded mercury void ratio of the pellet (e = 0.23) is lower than its total void ratio ( $e_T = 0.34$ ), indicating a significant porosity with an average diameter smaller than 5.5 nm, corresponding to the maximum mercury pressure applied (230 MPa). This is well known for compacted smectite (Lloret et al., 2003; Delage et al., 2006; Ridley et al., 2010).

A typical bimodal porosity distribution is observed for the bentonite powder, with an intruded void ratio of 0.9 at maximum mercury pressure (230 MPa). Actually, this void ratio is conditioned by the isotropic compression exerted by the mercury prior to penetrating the powder. The density function curve indicates that this penetration occurs at an average entrance diameter of 207 µm between powder grains with an average diameter of 0.65 mm.

This configuration is not far from the standard ratio between the diameter of spherical grains 304 305 and the inter-grains pores of their dense assembly, known to be close to 1/3. The second pore population is defined by an average entrance of 16.6 nm, to relate to a suction of 190.9 MPa 306 307 of the powder.

The PSD curve of the pellet has a population of micro-pores quite comparable with an 308 average entrance diameter of 13.3 nm, recalling that the powder was obtained from crushed 309 pellets. Another less clearly defined porosity is observed with an average entrance diameter of 310 5 µm, that will be described in more details later on. The intruded void ratio represents 67.6% 311 of the total void ratio. The void ratio of the population of large pores represents around 5.6% 312 of the total void ratio. 313

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#### **3.3. EDS characterisation**

315 Figure 13 presents SEM photos along with the corresponding EDS chemical analysis of the inclusions of different minerals observed within the pellet of bentonite. These inclusions 316 correspond to the high density elements found by  $\mu$ -CT observations and are included in the 317 mineralogical composition of the MX80 bentonite given in Table 1. These minerals are 318 identified based on the chemical composition detected by EDS: a high concentration of silicon 319 indicate an inclusion of quartz (see the two inclusions observed in Figure 13a and b); a 320 concentration in sulphur/iron indicate a pyrite inclusion (see Figure 13c). Note that the 321 dimensions of these inclusions are significant (137.3 µm and 70.4 µm for the quartz 322 inclusions) compared to the size of the pellet. An inclusion of calcite is also observed in 323 Figure 14, which was not identified in the X-ray diffractometer (XRD) analyses of MX80 324 bentonite (Table 1). 325

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#### 4. Discussion 327

Most of the investigations carried out up to now on bentonite-based materials have been based on the use of MIP and SEM (e.g. Saba et al., 2014; Wang et al., 2013, 2014; Romero et al., 2011). These techniques, that require preliminary dehydration of the samples and that concern specimen volumes smaller than about 1 cm<sup>3</sup> can be fruitfully complemented by more recent techniques like X-ray computed microtomography (Van Geet et al., 2005) that investigate specimens at larger scale and do not require any preliminary treatment.

In this paper, the microstructure of a pellet of bentonite at its initial state was studied by the 334 pore size distribution curve, obtained by MIP technique. An expected bimodal distribution 335 was obtained, where, theoretically, micro-pores correspond to voids inside an aggregate of 336 337 bentonite and macro-pores are related to pores between aggregates. The difference between the final value of intruded mercury and the total void ratio in the cumulative curves (Figure 338 12) suggests that there are several void sizes which cannot be filled by mercury. These voids 339 correspond to pores with a diameter smaller than 5.5 nm and large fissures identified by µ-CT 340 observations. Consequently, it would be convenient to complete this MIP results with 341 342 nitrogen gas adsorption (BET) technique in future investigations. The PSD curve of the powder of bentonite at its initial state was also obtained. The micro-pores of a single pellet 343 and that of the powder are similar. For the macro-pores, the mean entrance is much larger 344 than the obtained value for a pellet of bentonite. This value corresponds to the voids between 345 grains of bentonite. 346

347 X-ray computed microtomography ( $\mu$ -CT) observations revealed fissures at the upper part of 348 the pellet of bentonite (sections I and II presented in Figure 4 and sections I, II and III in 349 Figure 5) as well as non-swelling elements which could influence the swelling potential of the 350 material. For the section I in Figure 4, located at the upper spherical pole of the pellet, there is 351 a predominant grey level with some deviations. At the border of the section an element of high density (white) is distinguished. Furthermore, several fissures are observed, confirmingthat the pellet had already swollen during storage at the laboratory.

For section II (Figure 4), located at the upper part of the pellet, more fissures are observed at the edge of the pellet, as well as elements of high density (white pixels observed everywhere). Based on those observations, the following remarks could be made: (i) pellets swell from the border to the centre (layers in contact with the atmosphere will swell, inducing fissures) and (ii) fissures observed at section I (Figure 4), located at the upper spherical pole of the pellet, could be due to the fabrication process combined to swelling.

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For sections III and IV (Figure 4), located at the medium level and the lower spherical pole respectively, no more fissures are observed, but several elements of high density can still be identified. This suggests that the internal structure of a pellet of bentonite is not homogeneous. In addition to the existence of high density elements, different grey levels in the clayey part of the pellet are observed. The same conclusions can be extracted from the vertical sections I, II and III (Figure 5).

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#### **5.** Conclusion

The  $\mu$ -CT investigation of the microstructure of a pellet of bentonite provided interesting complementary features that could not be identified by MIP and SEM observations. Heterogeneous features were observed in the internal structure of the pellet, consisting in a heterogeneous density distribution of the clay minerals and in the presence of several high density elements. Furthermore, several fissures were observed at the upper pole of the pellet. This heterogeneity of a single pellet was confirmed by SEM and EDS observations, where several inclusions of non-swelling elements were observed (quartz, pyrite and calcite). The dimensions of these inclusions are not negligible, indicating that this could have an influenceon the swelling potential of the pellet.

The MIP tests carried out on a pellet and powder of MX80 bentonite revealed some similarity 378 379 between both materials, which is not surprising since the powder was fabricated by crushing pellets of bentonite. This similarity consisted on a double porosity and a comparable value of 380 the mean size diameter of the micro-pores. The complementary information provided by µ-381 382 CT observations evidenced that macro-pores found by MIP results are due to voids between fissures observed at the upper pole of the pellet and cracks observed at the top and the bottom. 383 A difference is observed when comparing the final value intruded by mercury and the total 384 385 void ratio in the cumulative curves, which corresponds to micro-pores with a diameter smaller than 5.5 nm and large fissures observed by  $\mu$ -CT observations on the upper part of the pellet. 386 Thus, in order to have a good understanding of the microstructure of bentonite-based 387 materials, it is important to carry out these complementary observations together with MIP, 388 SEM + EDS results. The u-CT observations obtained from a pellet/powder bentonite mixture 389 revealed a heterogeneous initial distribution of both materials within the sample. Thus, it can 390 be concluded that this mixture is characterized by a double heterogeneity: at a microscopic 391 level, a heterogeneous structure inside a pellet of bentonite, and at a macroscopic level, a 392 heterogeneous distribution of pellets and powder. The investigation of this initial 393 heterogeneity is important to understand the long-term evolution of the sealing for larger scale 394 systems. Initially, the system is characterized by a heterogeneous distribution of dry density, 395 so an anisotropic swelling will be attended. Future investigations should be conducted in 396 order to study the heterogeneity of the material at the final state. 397

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Table 1. Results from the X-ray diffractometer (XRD) of the MX80 bentonite. The results are
 expressed as a percent of the total mass (Laviosa Minerals SpA)

Mineral	% of the total mass
Pyrite	<1%
Smectite	80%
Albite	2%
Anorthite	8%
Quartz	4%
Muscovite	4%

<sup>510</sup> 

### Table 2. Cation exchange capacity (CEC) and original exchangeable cations of the MX80 bentonite (Laviosa Minerals SpA)

Property				
CEC Capacity	98 meq/100g			
Na conversion	52 meq/100g			
K conversion	1.2 meq/100g			
Mg conversion	10 meq/100g			
Ca conversion	37 meq/100g			

513

514

#### Table 3. Initial properties and conditions of pellets and powder of bentonite

	Pellet	Powder
Initial suction, $s_0$	132.4 MPa	190.9 MPa
Initial water content, $w_0$	7.25%	3.17%
Initial dry density, $ ho_0$	2.12 Mg/m <sup>3</sup>	
Initial porosity, $\varphi_0$	0.25	
Unit mass of bentonite particles, $\rho_s^*$	2.77 1	Mg/m <sup>3</sup>
*(C 1 ( 1 0014)		

515 \*(Saba et al., 2014)516

#### 517

#### Table 4. Properties of powder of MX80 bentonite

Atterberg limits			
Liquid limit, LL	560%		
Plasticity limit, PL	62%		
Shrinkage index, SI	498%		

518



Figure 1. Mixture of MX80 bentonite powder and pellets with a proportion of 20/80 in dry mass.



Figure 2. Pellet of bentonite at initial state. Dry unit mass  $\rho_d = 2.12 \text{ Mg/m}^3$ 



Figure 3. Grain size distribution of the MX80 bentonite powder.





Figure 4. X-ray computed microtomography (µ-CT) observations - horizontal sections - of a 







Figure 5. X-ray computed microtomography (µ-CT) observations - vertical sections - of a pellet of bentonite at initial state.



Figure 6. 3D rendering of a pellet of bentonite at initial state (a) and of the crack network in a region of interest (b).



Figure 7. Horizontal slice with the selected ROI (a) and segmented image (b).



vertical sections).



Figure 10. Vertical cross section of X-ray computed microtomography images of a mixture of
powder/pellets of bentonite with a proportion of 20/80 in dry mass obtained by three different
protocols. (a) First protocol of mixture, (b) Second protocol and (c) third protocol.



Figure 11. Evolution of the void ratio along height of a pellet/powder MX80 bentonite mixture (80/20) fabricated by three protocols.





Figure 13. SEM pictures taken on a pellet of bentonite at its initial state (132.38 MPa of suction) + EDS results. Mineral inclusions of quartz (a, b) and pyrite (c). Dry unit mass  $\rho_d = 2.12 \text{ Mg/m}^3$ 



Figure 14. SEM picture taken on a pellet of bentonite at its initial state (132.38 MPa of suction) + EDS results. Mineral inclusion of calcite. Dry unit mass  $\rho_d = 2.12 \text{ Mg/m}^3$