Observation of autogenous sealing of bentonite clay with X-ray computerized tomography

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ABSTRACT
Cracking in clay is frequently encountered in geoenvironmental and geotechnical engineering, sometimes causing failures in foundations, landfill liners and road and railway embankments. At the same time, desiccated clay materials are known to spontaneously heal their surface cracks when rehydrated. The autogenous healing capacity of clay has been typically inferred from changes in its permeability and mechanical strength. However, very little direct observation of crack sealing in real time has been reported in the literature. In addition, knowledge of the mechanisms driving self-healing in clay, and the effects of different variables on these mechanisms remain poor. In this paper, micro X-ray computerized tomography (µ-XCT scanning) is used to observe and quantify changes of crack volume in a bentonite clay as a result of swelling driven by hydration. A vertical crack is artificially introduced into an intact cylindrical sample of consolidated bentonite and the crack closure observed. The cracked zone is segmented and quantified by image processing. The effects on the pace and extent of sealing of sealing time, consolidation pressure and boundary constraints, are considered. Results from these experiments, bearing in mind the specific limitations of X-ray imaging, can contribute to a possible coupled chemo-hydro-mechanical interpretation of autogenous sealing of clay.

Keywords: crack sealing, swelling behaviors, crack volume change, µ-XCT scanning

1 INTRODUCTION
Cracking in clay is frequently encountered in nature and can cause failures in geotechnical and geoenvironmental engineering. The initiation and propagation of cracks in different clay materials have been extensively investigated by experimental observation and numerical modelling (Eigenbrod, 2003; Lu et al., 2015; Lu and Liu, 2017; Tang et al., 2011).

On the other hand, clays have a capacity to self-heal and self-seal their cracks which can be very useful in countering hazards caused by cracks (Mohammadi and Choobbasti, 2018). According to Bernier and Bastiaens (2004) and Horseman (2001), self-sealing is a hydromechanical process through which clay spontaneously recovers some or all of its intact hydraulic and mechanical properties. Self-healing can be regarded as self-sealing with complete recovery of properties.

Most studies of clay self-healing infer the process by measuring changes in specific properties (e.g., decreased permeability or increased strength). Very few studies have attempted to observe the sealing process directly. Furthermore, no theory of clay self-healing, based in experimental observation, has yet been proposed.

This study uses X-ray tomography to visualize self-sealing of an artificially introduced crack in a bentonite specimen, while assessing the effects of sample stiffness, hydration time and boundary constraints on the process.

2 MATERIALS AND SAMPLE PREPARATION
2.1 Materials
The clay mineral investigated in this research is a commercial sodium-bentonite produced in Queensland, Australia, named ActiveGel 150. This bentonite clay is a grey-yellow powder with an initial gravimetric water content of 12.7%. The basic geotechnical characteristics of the bentonite are presented in Table 1. These properties were either measured by the authors according to Australian Standards or taken from Shannon et al. (2010). It is worth noting that the liquid limit, has been measured with a cone penetrometer as 270%, compared to a manufacturer-supplied value of over 500%. SiO₂ and Al₂O₃ are the two typical chemical components occupying 56% and 16% of the material by weight, respectively. The pH value of bentonite suspension is 10. Figure 1 shows grain distribution of the Na-bentonite obtained from a hydrometer analysis.

2.2 Sample preparation
Bentonite powder was first dried in an oven under 105°C for at least 24 hours. After a few preliminary trials, distilled water was mixed with the dry bentonite powder at a 4:1 water-clay mass ratio to obtain slurries while avoiding soil leakage during the subsequent consolidation process. The mixtures were manually stirred with a spatula for at least 20 minutes, then sealed
and left to soak for at least 2 days. The sample was then poured into a consolidation cell of 38 mm diameter, then loaded in steps. The consolidation process of each sample took at least 2 weeks because of bentonite’s low permeability.

Table 1. Bentonite geotechnical properties.

| Parameter          | Type                  | Value | Unit | Source of data          |
|--------------------|-----------------------|-------|------|-------------------------|
| 𝑆𝑝                 | Specific gravity      | 2.69  | /    | Shannon et al. (2010)   |
| LL                 | Liquid limit          | 270   | %    | Measured by authors of this paper using a cone penetrometer |
| PL                 | Plastic limit         | 34.6  | %    | Measured by authors of this paper using a rolling device |
| PI                 | Plasticity index      | 235.4 | %    | /                       |
| LS                 | Linear shrinkage      | 46.38 | %    | Measured by authors of this paper using a shrinkage dish |
| 𝜌𝑑, max            | Maximum dry density   | 1.23  | g/cm³| Shannon et al. (2010)   |
| 𝑤𝑜𝑝𝑡              | Optimum water content | 20.2  | %    | Shannon et al. (2010)   |
| CEC                | Cation exchange capacity | 75  | meq | Shannon et al. (2010)   |
| Swelling volume    |                       | 35    | mls  | Shannon et al. (2010)   |
|                    |                       |       | /2g#| Shannon et al. (2010)   |

# meq/100g=milliequivalent per 100 grams
mls/2g=milliliter per 2 grams

Fig. 1. Particle size distribution.

After consolidation, specimens of 22 mm diameter and 12.5 mm height, were cut from the consolidated sample and inserted into a Perspex container of 22 mm inner diameter and 30 mm height. Next, a vertical crack of approximately 2.05 mm diameter was artificially introduced into the intact sample by pushing a needle through the centre of the sample and pulling it out at the other end. Care was taken to make the crack as uniform and vertical as possible. The samples were placed on a porous stone and allowed to hydrate from the bottom in a sealed container. They were then placed in a CT scanner for X-ray visualization at different times starting from t=0 when the crack was first introduced. The sample had to be disconnected from its bottom water source during scanning, but hydration was resumed after each scan, and while waiting for the next one.

2.3 μ-XCT scanning

A SkyScan 1172 micro-CT scanner at the Australian Centre for Microscopy and Microanalysis (ACMM), University of Sydney was used (Fig. 2) to visualize, monitor and quantify the self-sealing process during hydration. The scanner employs X-ray computerized tomography to capture images of the sample. A sample sits on a rotating table in the chamber and the X-ray beams can penetrate through the sample during scanning to generate shadow projections (Ponomarev et al., 2016).

The projections can be reconstructed by a software to provide a replica of the 3D structure of the sample. This structure is composed from a sequence of horizontal cross-sectional image slices where each slice represents a given thickness determined by the scanning resolution. In this study, the resolution was a 29.66μm pixel. The detailed instrument settings used in the study are shown in Table 2. Most samples have been allowed to swell upwards during hydration (no clamping) and hence need to be oversize-scanned, to obtain projections for the whole sample. 1565 and 2400 projections were generated in 80 min and 120-min estimated scanning durations, for normal and oversize scans, respectively.

Table 2. SkyScan 1172 setting.

| Scan setting               | Value               |
|----------------------------|---------------------|
| Source voltage & current   | 100kV and 100μA     |
| Image pixel size (resolution) | 29.66μm            |
| Camera pixel size          | 11.75μm            |
| Filter type                | Std High Al+Cu      |
| Exposure                   | 474ms               |
| Rotation step (normal)     | 0.23 degree         |
| Rotation step (oversize)   | 0.3 degree          |
| Frame averaging            | 3                   |
| Random movement            | 10                  |
| Use 360° rotation          | Yes                 |

The different testing scenarios are listed in Table 3. The aim was to quantify the effects on the pace and extent of clay sealing of three different variables, namely hydration time, boundary constraints and sample stiffness. The first two tests (CT#1 and CT#2) were identical and designed to assess the repeatability of the
In these two tests, the sample was consolidated under 157kPa allowed to swell freely at the top surface when hydrated (no clamping).

To test the effect of the boundary condition (free or constrained swelling), compaction was introduced in test CT#3 to prevent vertical swelling. Swelling was achieved by screwing together two steel caps at the bottom and top of the sample (see Figure 3). Finally, tests CT#4 and CT#5 were similar to CT#1 and CT#2 except that higher pressures were applied during the consolidation process, hence producing denser samples.

Each sample was X-rayed by the CT scanner immediately after the vertical crack was introduced (time \( t=0 \)), and then scanned again at regular intervals afterwards. Hence, scans were obtained at \( t=0, 4, 44, 92 \) and 138 hours, for most samples (where \( t \) is the time at the beginning of the scan). However, the scanner was not always available and some samples could not be scanned at some points in time (e.g., \( t=92 \) for CT#1). Each scan took between 80 and 120 minutes to complete, depending on the sample.

### RESULTS AND DISCUSSION

#### 2.4 Image processing

X-ray images of the sample were analyzed by ImageJ software (Hemes et al., 2015). The cross-sectional X-ray slices were segmented into binary images to distinguish between bentonite-water mixture and air. In the segmentation, the pixels of grey values from 58 to 255 were regarded as air (white, grey value of 255), and the pixels of grey values from 0 to 58 were treated as bentonite-water mixtures (black, grey value of 0). ImageJ software was used to calculate the area of air, which stood for the existing crack in the cross-sectional slices. Bentonite minerals such as quartz were known to attenuate X-ray beams (Kawaragi et al., 2009). Thus, shining dots appeared in the reconstructed images, which were regarded as large voids when segmenting into binary images, and removed as noise after segmentation.

Since each image represented a 29.66 mm-thick slice, several hundred data points of cracked areas were generated along the height from each scan (approximate total height was 12.5 mm before vertical swelling). In order to produce smoother curves, the sample height was divided into 10 segments and the crack area averaged over each section separately, hence generating an average crack area at the center of each segment.

Sealed sample area at a given elevation along the sample height was found by subtracting a) cracked area at a given time from b) cracked area at initial time for the same elevation. Hence, a negative value of sealed surface indicated crack widening rather than narrowing.

For samples hydrated without clamping, upward swelling of the sample raised a question about how to incorporate the total change in height of the sample (a point \( P \) at elevation \( x \) at a given time, will no longer be at the same elevation at a later time). To deal with this issue in the absence of data to keep track in time of each point in the sample, we proceeded as follows. We assumed that the sample swelled proportionally in the vertical direction during hydration. Hence, at each point in time, we converted the elevation scale (e.g., 0 to 12.5 mm) into a dimensionless scale from 0 to 1, with 0 representing the bottom of the sample and 1 its top after swelling at that point in time. Extent of sealing was then obtained by comparing cracked areas at the same dimensionless elevation and different points in time. Finally, overall sealing effectiveness (OSE) for the whole sample at a given point in time was calculated as:

\[
OSE = 1 - \frac{\text{present crack volume}}{\text{initial crack volume}}
\]

### 3 RESULTS AND DISCUSSION

#### 3.1 Overall self-sealing patterns

3D visualizations of samples were created by CTvox software (Cherobim et al., 2019), used for volume rendering of reconstructed image slices. Figures 4 and 5 present the cut-away views of the crack at different times in the hydration process, for non-clamped and clamped samples, respectively. The darker area is a rough,
qualitative sketch of the crack (quantified crack volumes are discussed later). In Figure 4, after 4 hours of hydration, the sample without clamping swelled upwards and the crack started to close from the bottom. However, towards the top, the crack became a little wider. After 44 and 138 hours of hydration, the sample showed significant vertical swelling and complete crack closure in the lower third of the vertically elongated sample. In addition, the crack in the middle and top thirds of the sample became narrower and presented as a conical shape. In Figure 5, after 92 hours of hydration, the sample prevented from swelling vertically did not completely seal cracks in any sections. The bentonite clay swelled inwards and the crack became narrower but more so toward the bottom.

3.2 Repeatability of experiments

To assess repeatability of experimental results, Figure 6 shows the size of the initial crack area at different elevations of two similar samples (CT#1 and CT#2). Both samples were consolidated under 157kPa pressure and hydrated without clamping. The figure shows that, despite our effort in producing identical cracks, some discrepancy between the crack sizes in the two samples existed (8% to 15% difference depending on elevation).

Figure 7 shows the sealed area at different points in time (crack area at initial time minus crack area at time t) for the two samples. At elevations 0.05 and 0.25, there is good repeatability at all three points in time under consideration. On the other hand, significant discrepancies seem to exist elsewhere to different degrees. This can be due to several factors including 1) differences in initial crack sizes and shapes between the two samples; 2) local material heterogeneities in each sample; 3) local heterogeneities in water distributions driving swelling in each sample; hysteresis in sealing and cracking as a result of interruption of hydration during the scanning process.

More encouragingly, good repeatability is observed at the scale of the whole sample. Table 1 shows the final water content, height and OSE of the samples at different points in time. Comparing results for samples CT#1 and CT#2, it is clear from the table that good repeatability of experiments is obtained when comparing vertical swelling and OSE of the two samples.

3.3 Sealing in time

Figure 8 shows change in cracked area with height for CT#1 and CT#2. Overall, the crack becomes smaller with time. However, the process is not spatially uniform. In the lower 10% of height, the crack seals quickly, as a result of swelling, driven by osmotic flow from the reservoir of water at the bottom of the sample into the soil. Towards the top, on the other hand, the crack becomes slightly larger in the first 4 hours of hydration then begins to narrow. After 138 hours of hydration, the crack has closed, completely in the lower part, but only partially elsewhere.

3.4 Effects of clamping

Sample CT#3 was prevented from swelling vertically as described earlier. Figure 9 shows the crack area at different elevations and different points in time. As discussed in section 3.1, the figure shows that clamping has a significant impact on the sealing pattern. After 92 hours of hydration, the crack was reduced to 33% and
62% of its original size, towards the bottom and the top, respectively, compared to about 75% towards the middle. This is compared to, in the case of no clamping (CT#1), a crack size of 0%, 61% and 78% of its initial value, in the lower, middle and upper regions of the sample after 138 hours of hydration. The difference in spatial distribution of sealing can be clearly seen in figure 10.

The OSE of this sample was 12.7%, 42.9% and 63.5% after 4, 44 and 92 hours of hydration, respectively. By contrast, in CT#1, the OSE was 30.4%, 36.6% and 51.1%, after 4, 44 and 138, respectively. Hence, sealing was found to be more extensive and more uniformly distributed when swelling was prevented in the direction normal to the crack.

3.5 Effects of consolidation pressure

Figure 11 presents sealed crack areas at different elevations and points in time for three samples consolidated under different pressures (CT#1, CT#4 and CT#5). After 4 hours of hydration (Figure 11a), the OSE was 30.4%, 30.0% and 46.6% for samples under 157kPa, 236kPa and 314kPa, respectively. After 138 hours of hydration (Figure 11c), the OSE of the three samples was 51.1%, 40.8% and 65.5%, respectively. Hence, the sample consolidated under 314kPa presented the greatest extent of self-sealing. However, the results did not reveal a clear relationship between sample stiffness and self-sealing capacity, since the sample consolidated under 236kPa has sealed less than the one consolidated under 157kPa pressure. Repetition of these tests is required to confirm these findings and additional tests are needed to further explore this relationship.

4 CONCLUSIONS

The healing capacity of clay materials is usually inferred from measuring changes in mechanical strength and permeability. Very few studies have reported direct observation of crack self-sealing in real time. Thus, this study employed X-ray computerized tomography to study the effects of different hydration times, boundary constraints and consolidation pressures on self-sealing of a bentonite. The following conclusions can be drawn:

1. The µ-CT scan does provide an effective means of visualizing and quantifying crack self-sealing
in bentonite, with good repeatability at the sample scale.

2. The vertical crack has narrowed with time. For the sample without clamping, self-sealing has been found to be spatially non-uniform, with sealing found to be complete towards the bottom of the sample and only partial elsewhere.

When the sample is prevented from swelling vertically, sealing is found to be more extensive and more spatially uniform.

3. The study has not yielded conclusive results about the effects of sample consolidation pressure on sealing. However, the stiffest soil has been found to experience more sealing. This finding still needs to be confirmed with further experiments that are underway.

The experiments conducted in this paper provide a set of experimental data that can help in developing a theory of clay self-healing. A starting point is an effort to capture the observed patterns of sealing through chemo-hydro-mechanical models that simulate osmotic swelling of montmorillonite.

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Fig. 11. Effect of consolidation pressure (a) after 4-hour hydration (b) after 44-hour hydration (c) after 138-hour hydration.