Effect of fluid chemistry on the microstructure of light backfill: An X-ray CT investigation

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**Abstract**

Bentonite–sand mixtures are widely accepted as the candidate sealing materials of radioactive waste repository due to their high swelling capacity and low hydraulic conductivity. These properties of the material depend largely on its microstructure, particularly, the pore space geometry. The aim of this investigation is to explore the effect of the pore fluid (NaCl and CaCl₂) on the microstructure, pore geometry and hydraulic conductivity of a barrier material. Representative samples of light backfill (LBF) prepared with a 50–50 bentonite–sand mixture at a compacted dry density of 1.24 Mg/m³ were used. As a first attempt, X-ray computed tomography (X-ray CT) was used to study the LBF under the distilled water (DW) and two other pore fluid conditions. In order to acquire a good quality image with high resolution, X-ray source, detector, and a small LBF specimen (5.5 mm in diameter) were placed close together and scanned with Xradia Micro XCT–400. The voxel of the scanned images were (1.15 × 1.15 × 1.15) μm³, indicating that the particles with a diameter greater than 2 μm could be easily observed. The porosity value estimated at the end of the consolidation test showed significantly higher values compared to the X-ray CT analysis. The interconnected pore components and absolute permeability of distilled water or salt-solution saturated LBF samples were analyzed using Avizo software. This analysis showed that the volume of interconnected pore increased due to the presence of salt solutions and resulted higher hydraulic conductivity. The salt solution increased porosity, pore size, volume of interconnected pore and hydraulic conductivity of the LBF.

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1. Introduction

The increasing generation of radioactive waste has become a challenge for safe storage of these materials. One of the possible solutions for the safe disposal of high level radioactive waste is deep geological disposal. Researchers have been investigating various sealing materials to limit the flow of liquid around deep geological repositories. Bentonite is widely used as an attractive material in general waste disposal sites while bentonite–sand buffer material is used in radioactive waste disposal sites (Kawaragi et al., 2009). The Canadian repository concept for disposal of radioactive and chemical wastes is a multi-component clay based materials to a great extent (Siddiqua et al., 2011, 2014). This change in hydraulic behavior can be attributed to the change in the microstructure of the sealing materials.

X-ray computed tomography (X-ray CT) is a non-destructive and non-invasive technique used to investigate the microstructure of an object based on the attenuation coefficient of the electromagnetic wave such as an X-ray. This technique was first developed by Godfrey Hounsfield (1973) and he received a Nobel Prize in 1979 for the invention of the X-ray CT equipment. X-ray CT was first used in the field of geotechnical engineering a few years after the invention. For example, X-ray CT has been used to determine the bulk density of soils (Petrovic et al., 1982) and the water content and water movement through soils (Crestana et al., 1984; Hainsworth and Aylmore, 1983). The exploration of synchrotron based X-ray micro-tomography by Flannery et al. (1987), has led to the development of elemental map and dynamic flow profiles at a resolution of 2.8 μm. Thereafter, this technique was applied in different fields of geotechnical studies such as the spatial distribution of soil properties (Heeraman et al., 1997; Nunan et al., 2006; Pierret et al., 2002; Rogasik et al., 2003; Young et al., 2001), pore network structures (Al-Raoush and Willson, 2005) porosity (Grevers et al., 1989; Hejls et al., 1995), permeability (Ketcham and Carlson, 2001; Mooney, 2002), and the characterization of pore space geometry and fractures with respect to different variables such as...
density (Anderson et al., 1990; Petrovic et al., 1982) and layer detection (Lipiec and Hatano, 2003; Macedo et al., 1998).

Industrial, medical, and synchrotron X-ray systems have various applications, dependent on the differing features of each system. In the medical scanner, the low energy X-ray source (<125 kV) and high efficiency detector rotate around a static object whereas in the industrial scanner, the object rotates in a cone-shaped beam of polychromatic X-ray. The medical scanner is also used for scanning an object at a meter-scale while in the industrial X-ray system an object can be scanned from a meter-scale to a micro-scale. Similar to the industrial scanner, the synchrotron scanner can scan an object to micrometers, where high intensity and highly collimated electromagnetic radiation emitted from the magnetic field of a particle accelerator is used during beam focusing. The advantage of the synchrotron scanner over others is its rapid scan speed and acquisition of low-noise data with few artifacts, which create an excellent environment of visualizing soil structure as described by Lehmann et al., 2006.

In this study, the effect of fluid chemistry on the hydraulic behavior and microstructure of the LBF were studied with 50% Na-bentonite and 50% sand at a maximum dry density of 1.24 Mg/m³ was observed. The particle size distribution of Wyoming bentonite shows that about 90% of the particles are within the range of 2 to 15 μm. Various researchers have studied the microstructure of compacted bentonite–sand mixtures using mercury intrusion porosimetry (MIP) and scanning electron microscopy (SEM) (Delage et al., 2006; Lloret et al., 2003; Villar and Lloret, 2001). The main disadvantages of these techniques are that they require some preparation, such as preliminarily dehydration of the samples and local observation of thin samples. These disadvantages can be resolved by using micro-focus X-ray CT. The use of X-ray CT on the investigation of microstructure of compacted bentonite based clay barriers (Kawaragi et al., 2009; Kozaki et al., 2001; Saba et al., 2014; Van Geet et al., 2005) is limited and most research is focused on the observation of macro-porosity (Baveye et al., 2002; Gantzer and Anderson, 2002; Nunnan et al., 2006; Rogasik et al., 2003). Recently, Feeney et al. (2006), indicated that with advanced scanning technologies, it is possible to scan an object having a voxel less than 15 μm, which contributes to the interpretation the fraction finer than sand. However, the phase isolation of a soil mass is still a challenge as there is no standardized approach for the interpretation of a particle’s size distribution and orientation, air filler pore spaces, and water volumes (Helliwell et al., 2013). Literature regarding the application of X-ray CT for compacted bentonite–sand mixtures mainly consist of the observation of microstructure and anisotropic swelling behavior (Kozaki et al., 2001; Saba et al., 2014; Tomioka et al., 2010), and properties of cracks, such as porosity and pore networks (Gebrenewegus et al., 2011; Kawaragi et al., 2009). According to the author’s knowledge, there are no studies regarding the application of X-ray CT to detect the effect of fluid chemistry on the microstructure of bentonite–sand mixtures. Therefore, this study will be a first attempt to investigate the 3D-microstructure, porosity, pore connectivity, and permeability of compacted bentonite–sand mixtures in relation to variable pore fluid chemistry. This paper will also present a comparison of X-ray CT data with respect to the experimental results.

2. Materials and methods

2.1. Materials

Na-based Wyoming bentonite with a montmorillonite content of 76% and silica sand were used to prepare LBF samples. The particle size analysis showed that about 90% of the particles are within the range of 2 to 15 μm and the sand particles are within the range of 200 to 600 μm. Initially, three dry samples, (i) dry sand, (ii) dry bentonite and (iii) dry bentonite–sand mixture, were prepared at a dry density of 1.65, 1.5 and 1.5 Mg/m³, respectively, to understand the microstructure of the materials at dry state. Along with the three dry samples, five saturated samples were prepared (Table 1) with distilled water and two concentrations (50 g/L and 100 g/L) of two different salt solutions (NaCl and CaCl₂). This observation allowed for a comparison between the microstructure of the dry and saturated compacted bentonite–sand mixtures. Polypropylene tubes with a volume of 0.2 mL and inner diameter of 5.5 mm were used for containing the samples during scanning.

2.2. Swelling pressure and consolidation testing

In this study, swelling pressure was measured using an incremental stress method using 1D consolidation apparatus. Initially, 50–50 bentonite–sand was mixed in the dry state as this yields the highest homogeneity and repeatability for both swelling and saturated hydraulic conductivity measurements (Gebrenewegus et al., 2011). The carefully mixed bentonite–sand materials were then mixed with distilled water or salt solution. A total of five samples such as S1, S2, S3, S4 and S5 were prepared by using distilled water, 50 g/L NaCl, 100 g/L NaCl, 50 g/L CaCl₂ and 100 g/L CaCl₂, respectively (Table 1). The initial water content of all of the samples was 19%. A compacted bentonite–sand sample having a diameter of 63.5 mm and height 10 mm was prepared at a dry density of 1.24 Mg/m³ to study the hydraulic behavior of the LBF.

A computer-controlled automated oedometer (from GDS Instruments) was used to conduct both the swelling pressure and hydraulic conductivity tests. The maximum swelling pressure was determined by incremental stress condition. In this approach, for a particular load increment the specimen was first deformed and after some time it was rebound to its original height without releasing loads. When it returned to its original height the next incremental load was applied. Swelling pressure was determined in such a way that it is the maximum applied stress at which the sample could swell and rebound to its original height and beyond this stress the specimen could no longer rebound. After completing the swelling stage of the sample, the loading for the consolidation test was applied on the same specimen. The applied loads for the loading stage for the consolidation was 250, 500, 1000, 1500 kPa and for the unloading stage was 750, 250, 100, 50 and 25 kPa. The deformation for each particular load increments were recorded every 30 s and continued until it showed constant deformation.

Table 1

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>LBF samples prepared with</th>
<th>Maximum dry density (Mg/m³)</th>
<th>Maximum swelling pressure (kPa)</th>
<th>Hydraulic conductivity (m/s)</th>
<th>Porosity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Experimental</td>
</tr>
<tr>
<td>–</td>
<td>Dry sand</td>
<td>1.65</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>–</td>
<td>Dry bentonite</td>
<td>1.50</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>–</td>
<td>Dry bentonite–sand</td>
<td>1.60</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>S1</td>
<td>Distilled water</td>
<td>1.22</td>
<td>225</td>
<td>9.86 × 10⁻¹²</td>
<td>61.97</td>
</tr>
<tr>
<td>S2</td>
<td>50 g/L NaCl</td>
<td>1.23</td>
<td>175</td>
<td>2.81 × 10⁻¹¹</td>
<td>61.65</td>
</tr>
<tr>
<td>S3</td>
<td>100 g/L NaCl</td>
<td>1.27</td>
<td>150</td>
<td>7.93 × 10⁻⁹</td>
<td>60.39</td>
</tr>
<tr>
<td>S4</td>
<td>50 g/L CaCl₂</td>
<td>1.33</td>
<td>50</td>
<td>2.56 × 10⁻⁹</td>
<td>57.59</td>
</tr>
<tr>
<td>S5</td>
<td>100 g/L CaCl₂</td>
<td>1.34</td>
<td>25</td>
<td>2.19 × 10⁻⁹</td>
<td>49.59</td>
</tr>
</tbody>
</table>
2.3. X-ray CT observation

X-ray CT primarily depends on the imaging techniques rather than the paramagnetic elements of a soil (Macedo et al., 1998). The process consists of a synchrotron light or conventional X-ray tube as the source, a sample manipulation stage and a detector. The basic principle of X-ray CT is the progressive attenuation of electromagnetic waves by absorption and scattering of the X-rays emitted from the source. When the X-rays pass through the sample it creates a series of radiograph images of the sample which are acquired at an incremental angle over 360°. The attenuation coefficient is the amount of absorbed or scattered photons by an object depending on the density of materials, electron density of the voxel of interest, and the incident energy (Helliwell et al., 2013). The attenuation coefficient of radiograph images are integrated based on the mathematical filtered back propagation algorithms to generate 2D image slices, termed tomographic reconstruction (Stock, 2008; Taina et al., 2008; Wildenschild et al., 2002). Each reconstructed tomographic slice is comprised of a distinct unit known as voxels (3D pixels) which demonstrate the spatial resolution of the scans.

In the current investigation, the Xradia Micro XCT-400 X-ray tomographic microscope (Fig. 1a) located at the University of British Columbia Okanagan campus was used to acquire the 3D radiographs of both dry and saturated samples. A specially designed sample holder (Fig. 1b) was used to support the samples and to minimize the loss of water. After placing the sample holder on the rotating disk, an energy of 70 kV and current of 125 μm were used to acquire the radiographs over a rotation from −180° to 180°. Although it was challenging to acquire good quality images with a voxel size less than 2 μm, the particle size analysis of Wyoming bentonite showed that about 90% of particles were within the range of 2 to 15 μm. The spatial resolution of a CT image depends on the focal spot size, performance of detector, binning number, and the distance between the source and detector from the sample. For example, Dhondt et al., 2010, indicated that high resolution images can be acquired by placing the sample closer to the source and further from the detector, although the larger distance of detector from the sample results blur images. Higher resolution can also be attained by reducing the size of the sample. However, for a better understanding of the pore distribution and pore networks in field conditions, larger samples are required for adequate representation of the field conditions (Young et al., 2001). The application of the ‘region-of-interest’ technique has significant benefits to ensure high resolution for large soil samples (Carminati et al., 2009; Stock, 2008) but tends to blur the image quality. After several trials, it was found that good quality images could be acquired with a resolution of 1.0695 μm by placing the sample at a distance of 37 mm from the source and 8 mm from the detector. Another important parameter for acquiring good quality images is the exposure time. The exposure time (time required for taking an individual radiographs) was carefully selected as it determines the number of photons counted at the detector and affects the level of noise in the image. Xradia LE #4 source filter was selected after some trials and errors to reduce the artifact i.e. beam hardening on the CT images. Ketcham and Carlson (2001) demonstrated that when a polychromatic beam passes through an object, it loses the lower energy portion of its spectrum because the lower energy X-ray are attenuated more radially compared to the higher energy X-ray. As a result, the effective attenuation coefficient of an object diminishes and makes the edge (short ray path) of the images brighter than the interiors (long ray path). An average of 3200 radiographic projections was acquired with an exposure time of 25 s. The acquired images were then reconstructed to 8 bit gray scale 3D volume (voxels) using XM Reconstructor-Cone Beam software developed by Xradia.

In order to investigate the effect of water on the properties of bentonite–sand mixtures, the dry samples of each material and their mixture were scanned along with the distilled water (S1) and salt solution saturated LBF (S2 to S5). A polypropylene tube was used for sampling the LBF specimens after the consolidation test. After the consolidation test the LBF samples were in the swelling stage and thus the porosity of the samples showed maximum values. The two dimensional scanned images of dry bentonite–sand at a dry density of 1.60 Mg/m³ and water saturated bentonite–sand LBF at a dry density of 1.24 Mg/m³ are shown in Fig. 2. The scanned images from the dry samples showed better quality than the wet samples due to the development of high swelling pressure in the saturated sample. This swelling pressure changes the structure of bentonite particles causing them to spread out due to the formation of a strong bentonite–water gel which reduces the ability of fluid flow, according to the diffuse double layer theory.

![Fig. 1. (a) Sample mounted on the rotation stage of the Xradia Micro XCT-400; and (b) sample holder.](image-url)
In the second stage, the interconnected particles were segmented present in the LBF degrades, swells and constitutes bentonite. The segmented images were then used to observe the 3D microstructure, porosity, pore size distribution, pore connectivity and permeability of LBF samples prepared with both the distilled water and salt solutions.

3. Image analysis

3.1. Preprocessing of scanned images

Fig. 3 presents the image preprocessing steps of dry sand, bentonite, bentonite–sand mixtures and distilled water saturated LBF. Initially, scanned images were cropped to nearly square in size and the brightness of the images was adjusted to improve the visualization. The images contained speckled noise and were de-noised using a median filter with a radius of 1.35 pixels. In the first stage of segmentation, the de-noised images were segmented to classify the individual voxels with a common mean gray scale value termed the threshold value. The threshold value in the segmented images is ordered in terms of attenuation density where the densest part is represented by bright voxels and the least dense portion is represented by darker voxels. The saturated LBF samples comprised of sand, bentonite, water and air void, which have different attenuation density. The resulting CT value represents some average of their attenuation density, which is termed as partial volume effect and the resulting voxel is called mixel. Although, when an image contains only two phases voxel it can easily be minimized by using subtraction technique (Fukuda et al., 2012). However, for images that shows multiphase voxels (for examples LBF samples), it is a difficult task to subtract different voxels. Additionally, bentonite present in the LBF degrades, swells and constitutes bentonite–water gel around the sand particle when it was saturated with liquid. Therefore, the application of subtraction technique was not suitable for the LBF samples. Global thresholding, based on the histogram of the image, was used to segment materials of interest in the soil (Fig. 3b). In the second stage, the interconnected particles were segmented using the watershed algorithm presented by Vincent 1991 (Fig. 3b). This algorithm first calculates the gradient of the image and then searches the region of highest density that divides the neighboring local minima. The highest density region, local minima and their connecting edges are considered as the catchment basin, surface hole, and shed or dam. The segmentation is done on the immersion basis so that water successively fills surface holes enclosed by the shed of the catchment basin. The key advantage of this technique is that it firstly detects the main edges of the object and then computes the watershed of the identified gradient. Prior to applying the watershed algorithm, the images were processed using a Gaussian filter with a radius of 1.5 pixels to reduce the initial number of regional minima. The segmented images were then used to observe the 3D microstructure, porosity, pore size distribution, pore connectivity and permeability of LBF samples prepared with both the distilled water and salt solutions.

3.2. Analysis of porosity and pore size distribution

A 3D visualization of the microstructure improves the reflection of the actual arrangement of the particles in a bentonite–sand–water matrix and quantifies the continuous pore network and pore connectivity (Mooney, 2002). However most of the research regarding pore networks and pore connectivity focus on the macro level because of the scale limitation associated with X-ray CT imaging (Helliwell et al., 2013). The 3D microstructures of the compacted specimen were composed with a resolution of 1.2 μm³ (voxel size: 1.0695 × 1.0695 × 1.0695). An algorithm was used to determine the volume porosity of the 3D oriented samples. This algorithm is a counting program which counts all the black and white voxels from the 3D binary stack; the porosity was then determined by dividing the black voxels with the total voxels.

An algorithm was developed and coded in Matlab by the authors in order to determine the pore size distribution of the 3D oriented samples. All of the image slices were converted into binary, where ‘0’ denoted a black pixel (i.e. pore) and ‘1’ denoted a white pixel (i.e. particles). The binary images were cropped to 300 × 300 μm to reduce the simulation time. In the first stage of the algorithm, a function was developed to pick a black pixel (X) from the binary image (Fig. 4a) and then moved along the entire neighboring pixels according to Fig. 4b. When the function reached a white pixel the movement stopped and the white pixel was marked as a boundary for that pore. However if the function found any neighboring black pixels during each movement, it would send the black pixel to a type of data structure known as a queue. A queue is a sequential data collection system, in which the entities are added and removed according to the first-in-first-out (FIFO) basis and for a single queue structure all the removed data are marked with the same label number (Fig. 4c). In a similar way, ‘X’ move the three global axes to determine the total voxel occupied by the first pore and label them with the same number. The total voxel found for a particular set of labels represents the volume of that pore. After labeling the first pore, the function randomly picks another black pixel and labels it accordingly. This process of counting pore volumes continued until all of the black pixels was labeled. The volume of each pore present in a 3D pore structure was determined accordingly to make a pore volume distribution plot.
3.3. Pore connectivity

Investigation of pore space geometry and pore connectivity within the soil mass is important to characterize fluid flow. The pore connectivity depends on the size and number of pore throats surrounding a single pore. The 3D pore space geometry was visualized and connected porosity was computed using an Avizo built-in function named connected component analysis. The interconnected pores were analyzed in such a way that the volume porosity is considered as connected if the pores share at least one common voxel face as mentioned in Hemes et al. (2015). In the current investigation, the connected porosity volumes were divided into the four groups according to their volume: 1 to 10 μm³, 10 to 100 μm³, 100 to 1000 μm³ and greater than 100 μm³.

3.4. Analysis of hydraulic conductivity

The ability of a porous material to transmit a single phase fluid, known as absolute permeability (m²), was measured from the 3D microstructure of the LBF samples using Avizo-Xlab simulation Software. The absolute permeability was computed by solving a single phase flow problem for a unit drop of applied pressure in all of the axial directions of the samples (Krotkiewski et al., 2011). Darcy’s law was used to determine the permeability of porous material only on the macro-scale using Eq. (1).

\[
\nu = -\frac{K}{\mu} \frac{\Delta P}{L}
\]  

(1)

where, \( \mu \) is the dynamic viscosity of fluid (0.000891 kg/m s for water at a temperature of 25 °C), \( L \) is the length of porous media in the flow direction (m), \( \nu \) is the velocity through the porous media (m/s) and \( \Delta P \) is the pressure difference applied to the sample (Pa). However on the micro-scale, where free flow occurred, it was necessary to understand the flow through individual pores and pore throats. This problem was solved by using both Darcy’s Law and Stokes’ theorem for the incompressible and Newtonian fluid flowing in a steady and laminar manner. To make the calculation easier, a simplified Stokes' equation was used (Eq. 2).

\[
\begin{align*}
\nabla \cdot \mathbf{V} &= 0 \\
\mu \nabla^2 \mathbf{V} - \nabla P &= 0
\end{align*}
\]  

(2)

where, \( \nabla \cdot \) is the divergence operator, \( \nabla \) is the gradient operator, \( \mathbf{V} \) is the velocity of the fluid in the fluid phase of the material, \( \nabla^2 \) is the laplacian operator and \( P \) is the pressure of the fluid (Pa) in the fluid phase of the material. The boundary conditions associated with the computation of permeability are that (i) no slip occurred in the fluid–solid interface, (ii) the flow is isolated within the system, and (iii) the fluid can freely spread on the face of the sample. Permeability (K in m²) computed from both the Darcy’s and Stokes’ equations can be expressed in terms of hydraulic conductivity (k in m/s) using Eq. (3) (Bear, 1972).

\[
k = \frac{K \rho g}{\mu}
\]  

(3)
where, \( \rho \) is the density of fluid (1000 kg/m\(^3\) for water) and \( g \) is the acceleration due to gravity (9.81 m/s\(^2\)).

4. Results and discussion

4.1. Swelling pressure

The maximum swelling pressures of the compacted LBF at different pore fluid conditions obtained from the laboratory tests are presented in Table 1. The results showed that the swelling pressure decreased while the hydraulic conductivity increased in the samples prepared with the salt solution. It was observed that the variation in swelling pressure was more prominent to S2 and S3 compared to the S4 and S5. For example, samples S4 and S5 showed that the decrease in swelling pressure is about 80\% that of S1. However, samples S2 and S3 showed a decrease in swelling pressure is only 20\% of S1. Sato et al. (1992) observed the expansion of different types of bentonites, in terms of basal spacing, saturated with various types of solvents (ethylene glycol, Glycerol, Na, K and ca). They observed that Wyoming bentonite shows higher value of basal spacing than other types of bentonite because its 75\% layer charges are distributed on the octahedral sheet. In addition the basal spacing of Wyoming bentonite saturated with Na-solvent was higher than that saturated with other solvents. The swelling pressures determined from the experiments in the present study show similar trend as that found in Sato et al. (1992).

4.2. Porosity

The porosities of both the distilled water and salt-solution saturated LBF were calculated from the one dimensional (1D) consolidation test and X-ray CT image analysis (Table 1). In the 1D consolidation tests, porosity (n) was calculated from the void ratio–porosity relationships as presented in Eq. (4). While, void ratio at maximum applied stress was calculated using Eq. (5). Porosity values for the salt-solution saturated LBF from 1D consolidation test were corrected according to Siddiqua et al. (2011).

\[
n = \frac{e}{1 + e} \quad (4)
\]

\[
e = \frac{G_s \rho_w}{\rho_{dry}} - 1 \quad (5)
\]

where, \( G_s \) = specific gravity of the soil solids; \( \rho_w \) = water density; and \( \rho_{dry} \) = dry density of the soil mass. It was observed that the porosity value i.e. total pore space per unit volume found at the end of the consolidation tests showed significantly higher values compared to effective porosity that was found from the X-ray CT analysis. These higher values of experimental porosity are attributed to the swelling behavior of Wyoming bentonite by constructing a thicker diffuse double layer (DDL) after being saturated with liquid. This DDL spreads the solid particles present in the saturated LBF to a wider range by creating a bentonite–water gel around the particle’s periphery. Although, this bentonite–water gel does not allow fluid flow through it, in the calculation technique used in the experimental porosity investigation (i.e. experimental porosity is the change in volume of solids compared to the initial volume), the bentonite–water gel was considered as void space, resulting in higher porosity values. Dixon et al. (1985) also showed that the value of effective porosity is lower compared to the total pore space per unit volume due to the presence of highly viscous bound water in the pore space. In the case of image analysis, the bentonite–water gel was considered as a compound mass resulting in low porosity estimations. Moreover, the value of experimental porosity decreased with the addition of salt solution due to the reduced thickness of DDL by the action of cations present in the pore fluid. However, the estimated porosities from the image analysis tended to increase with the addition of salt solution, similar to findings of Studds et al., 1998. Therefore, the porosity values calculated from the X-ray CT investigation are effective porosity which show a better correlation lower permeability of bentonite samples. While porosity obtained from the 1D consolidation tests overestimates the porosity, therefore, it was not consistent with the permeability values.

Fig. 5. Particle size distribution of dry materials (bentonite, sand, bentonite–sand mixture) and distilled water and salt-solution saturated LBF.
4.3. Pore size distribution

Fig. 5 depicts the volumetric pore size distribution of compacted dry materials (sand, bentonite and bentonite–sand mixtures) and both the distilled water and salt-solution saturated LBF. It was observed that almost 70% of the pores present in the compacted sand are on the macro-size, whereas the dry bentonite resulted with about 50% of the pores within the 1 to 10 $\mu$m³ range. Additionally, the 50–50 bentonite–

![Fig. 6.](image)

Results of the connected component analysis of distilled water saturated LBF samples (a) 3D Microstructure of the representative samples, (b) interconnected pore with a volume ranging from 1 to 10 $\mu$m³, (c) connected pore with a volume ranging from 10 to 100 $\mu$m³, (d) interconnected pore with a volume ranging from 100 to 1000 $\mu$m³, (e) interconnected pore with a volume larger than 1000 $\mu$m³, (f) interconnected pore space showing different sizes of pore volumes.
sand mixtures reduced the pore size, where about 75% of the pores were within the range of 1 to 10 μm$^3$. This is due to the micro-sized bentonite particles filling the macro pore of the sand particles. The pore size distribution of samples S1, S2 and S3 had about 50% of the total pores in the range of 1–10 μm$^3$ and had about 90% of the total pores in the range of 10–100 μm$^3$. While, the percentage of pores found within the range of 1 to 10 μm$^3$ decreased in the LBF samples saturated with CaCl$_2$ solution from 60% to 40%.

4.4. Pore connectivity

The number of pore volumes interconnected on different faces of the pore by pore throat were analyzed to establish a relationship between the pore throats in the microstructure of the LBF and the hydraulic conductivity. Although there are different calculation techniques to determine the number and volume of connected components (Vervoort and Cattle, 2003; Vogel, 1997), in this investigation a built in function in Avizo (Avizo 9.0, 2015) ‘connected component analysis’ was used to label and compute the separate components in a binary image. The connected components were divided into four size ranges according to their volume. The percentage of connected components in each size range was determined to investigate the ability of samples to transport fluid, as the ability of fluid transfer increases with the increasing percentage of larger sized connected pores. The result of the connected component analysis for the representative samples S1 revealed a total interconnected pore volume of 3.31 × 10$^2$ μm$^3$ (Fig. 6a). While the maximum number of interconnected pores were within the range of 1 to 10 μm$^3$ and contributed to 23% of the total resolved porosity (Fig. 6b), the maximum porosity (about 50% of the total resolved volume porosity) was governed by the interconnected pores which had a volume in the range of 100 to 1000 μm$^3$ (Fig. 6c). Few interconnected pores were found within the range of 100 to 1000 μm$^3$ (Fig. 6d), which contributed to 22% of the total resolved porosity and only two pores are found having a volume larger than 1000 μm$^3$ (contributed only 5% of total resolved porosity) (Fig. 6e). The enlarged view of the interconnected pores in Fig. 6e shows how the pore bodies are connected with pore throats which creates a larger volume of pores.

The effect of pore fluid on the interconnected pore volumes in terms of number fraction and percentage of total porosity is shown in Fig. 7. It was found that the total porosity volume contributions of interconnected pores of the LBF samples were shifted, to a large extent, with the presence of salt solutions. The majority of the numbers of pore fractions were found below the volume of 100 μm$^3$ for the LBF samples. Sample S1 contained the maximum number of pore fractions within the range of 1 to 10 μm$^3$, but the percentage contribution to the total volume porosity was found within the range of 10 to 100 μm$^3$. The peak contribution to the total porosity of samples S3, S4 and S5 was found for the pore volume ranging from 100 to 1000 μm$^3$, while sample S2 showed the peak value within the range of 10 to 100 μm$^3$. Moreover, the effect of the samples S4 and S5 was more pronounced in increasing the larger volume interconnected pores. Therefore, it was concluded that the LBF samples prepared with a salt solution increased the volume of the micro-pores resulting in higher permeability within the sample.

4.5. Hydraulic conductivity

The experimental results showed that the hydraulic conductivity of the LBF samples prepared with CaCl$_2$ solution at the unloading stage increased by about 27 times compared to samples prepared with distilled water, whereas for the NaCl solution treated LBF, the hydraulic conductivity increased only about 2 times that of the samples prepared with distilled water (Fig. 8). This change in hydraulic behavior is attributed to the thinning of the DDL, which varies inversely with the square root of the concentration of pore fluid (Mitchell and Soga, 1976; Yong and Warkentin, 1975). However, to inspect the hydraulic behavior of the LBF samples through X-ray CT, a small representative sample (about 400 × 400 × 350 μm) was cropped to analyze. Though it is difficult to determine the true behavior from this small sample, the results contribute to the understanding of fluid flow through individual and connected pores and gives insight to the hydraulic conductivity of the dry materials. For example, the hydraulic conductivity of the dry sand was 3.9 × 10$^{-2}$ m/s, similar to the typical values for sand mentioned in (Das, 2013). However, the hydraulic conductivities found for both the dry bentonite (1.7 × 10$^{-4}$ m/s) and bentonite–sand mixture (1.7 × 10$^{-4}$ m/s) were significantly different from the typical values (10$^{-10}$ to 10$^{-12}$). This is because the hydraulic conductivity of saturated fine grained soil largely depends on the swelling pressure and the bentonite–water gel formation according to the DDL theory (Siddiqua et al., 2011). The hydraulic conductivity of the pore fluid treated LBF determined from the X-ray CT investigation showed similar patterns where they increased with increasing salt concentration (Fig. 8). The hydraulic conductivities determined from the scanned images of CaCl$_2$ solution treated LBF showed a better correlation with the experimental data compared to samples prepared with the NaCl solution. However, hydraulic conductivities computed from the scanned images of the distilled water treated samples showed significant variation with the experimental measures.

5. Conclusions

In the current investigation, a non-destructive X-ray CT technique was used to visualize the 3D pore space such as pore bodies, throats, and size distribution of salt-solution saturated LBF samples. A special type of sample holder was designed to hold the samples and reduced the loss of water during the scanning. In order to obtain good quality images at the expected resolution (~2 μm), the sample holder was placed 8 mm from the detector and 37 mm from the source. The reconstructed images were de-noised and segmented to separate the particles and pores present in the compound mass. Total porosity analysis of the representative segmented samples revealed that the porosity of the LBF samples increased with the salt solutions because of the thinning of the DDL. It was also observed that the thinning of the DDL was more pronounced in the presences of higher cation valances and thus the CaCl$_2$ saturated LBF sample resulted in a higher volume porosity. Similar to the total porosity, the results of the pore size distribution and interconnected pores revealed that salt solutions increased the pore sizes and number of interconnected pores of LBF, making the material more permeable compared to distilled water saturated ones. This finding was supported by the results of the absolute permeability analysis.
using Avizo XLab Simulation Software, even with the limitation of sample size. This investigation has revealed that X-ray CT is a versatile technique for the analysis of soil microstructure and hydraulic behavior.

The results showed that the hydraulic behavior between the LBF and the pore fluid present in the deep geological repositories may increase the connections among the pore bodies resulting in a higher permeability in the field, highlighting the importance of this research for designing safe radioactive waste disposal technologies.

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