The swelling pressure of bentonite and sand mixtures

Sandra Sánchez González

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Abstract

The compacted bentonites are used as buffer and backfill materials for engineering barriers for high-level nuclear waste repositories located underground. For this purpose, it is very important to evaluate the swelling characteristics of this clay.

The swelling capacity is one of the most important properties of the bentonite clay. The swelling behaviour is due two mechanisms, the crystalline swelling and the osmotic swelling. These mechanisms produce an increase in the distance between the layers of montmorillonite which is one component of bentonite.

The result of the swelling capacity is the swelling pressure. It has been studied in several investigations. The results of experimental tests have been collected and compared in this thesis, considering only the Na-dominant bentonite and sand and distilled water as test solution.

The experimental tests show that there is only an unique relation between different bentonites in the Na-dominant bentonite and sand mixture swollen depending on its final dry density. Also, the relation between the swelling pressure and the clay void ratio shows the mechanism of the swelling pressure.

On the other hand, a mechanistic model is used to predict the swelling pressure of fully saturated bentonite and sand mixture in distilled water. Firstly, it has been compared with the results of experimental tests and it should be pointed out that the model gives good predictions.

In addition, the model has been used to make sensitivity analysis with different parameters of bentonite. The most important conclusions in this section show that the swelling pressure mainly depends on the distance among particles. Also, the sensitivity analyses indicate which parameters should be fitted more carefully for future studies to validate this model with different bentonites.

Keywords: Swelling pressure; bentonite and sand mixture; crystalline swelling; osmotic swelling; mechanistic model; montmorillonite.

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1. Introduction

1.1. Background

Nowadays, preservation of the environment is one of the most important matters in our society. One of the most challenging factors to deal with the industrial world is to find better techniques for disposing radioactive wastes from nuclear power stations. The main principle of these wastes management is the containment and isolation for the radioactive wastes, knowing that the radiological hazard of the wastes is not permanent. This hazard diminishes over time as a result of the radioactive decay but it takes a long time to occur [4, 21].

Until the 80s, many countries threw wastes into the sea in order to remove them but in 1994, this practice was banned permanently [6]. Nowadays, this waste is stored in the land. At present, the total disposal of high-level radioactive wastes has not been reached yet but there are two considered options: a superficial or an underground repository. The best proposed solution for the high-level activity wastes is storing the radioactive wastes underground where isolation is ensured from a series of engineered barriers as well as properties of the host rock.

Finland, France and Sweden are the most advanced countries in the line of high level disposal waste [28]. *Posiva Oy* is the organization accountable for the final disposal in Finland [25], *Andra* in France and *SKB* in Sweden [30]. They are not operating with this kind of repositories yet, but in a few years they will.

The clay as engineered barrier is used in order to curb the uncontrolled movement of the contaminants in the soil. In most cases, compacted bentonite-based materials are chosen as a sealing/buffer materials.

The bentonite is a type of clay that is mixed with sand and it becomes an effective component of artificial barriers. This type of clay is better than others because of its properties. The swelling capacity of compacted bentonite and sand mixture is one of the most important researched properties. This characteristic is very important for the design and for the construction of the disposal facilities of nuclear wastes.

[10, 12, 13, 14, 15, 16, 23, 24, 32, 33, 38, 40]

1.2. Problem description

The crucial question is whether it can guarantee the security of the storage facilities for thousands of years, doing it without producing cracks which are formed in the protection barriers causing water seep.

To answer this question, there are many factors to keep in mind. We must know the characteristics of every barrier protecting the underground waste [24]. The engineering barriers must be researched and experimented to justify that there will not exist any risk of escaping from high level wastes. In this thesis we will study one property of the compacted bentonite, the behaviour of swelling.

1.3. Aims of the project

The present study is to observe from different studies the behaviour of the mixture of bentonite-sand under vertical pressure changes. In order to know the behaviour of one of their most important properties, many studies have been investigated to know about the effect of the swelling pressure, using different methods. The studies are from Karnland, [8]; Marcial, [22]; Mollins, [23]; Studds, [32]; Sun, [33]; Tripathy, [34]; and Wang, [38].

There are two different ways that we can observe the behaviour of the mixture. On the one hand, one of these ways is comparing different experimental data of different researches. On the other hand, we can analyse the behaviour of the swelling pressure when the values of the properties are changed, using a mechanistic model.

1.4. Organization and content of the thesis

The thesis is composed of 6 more chapters. In chapter 2, we will define the basic concepts about the wastes and the protective barriers from the underground repositories, focusing on the mixture of bentonite-sand. Also, this chapter presents a detailed definition of bentonite and the basic properties of the most important component of bentonite, montmorillonite.

Chapter 3 describes the mechanism of the bentonite swelling. It is divided into several parts. Firstly, it carries out a description of the swelling process. Secondly, the mechanisms on the swelling behaviour are defined, being the mechanisms the crystalline and the osmotic swelling.

Chapter 4 shows the experimental work from different studies [8, 22, 23, 32, 33, 34, 38]. It shows the material used in the investigations aforementioned and the different properties of the bentonite and sand. Furthermore, it describes the experimental methods.

The mechanistic model will be described briefly in chapter 5.

To finish, in chapter 6 we will study and compare the results of the experiments about the swelling pressure in the bentonite from the different researches aforementioned. Also, using the mechanistic model we will do a sensitivity analysis by varying some properties of bentonite. In the end, it presents some conclusions and future research in chapter 7.

2. Review

2.1. Radioactive wastes

The radioactive waste can be defined as any material which contains or it is contaminated with radionuclides in concentrations or activity levels higher than those established by the laws [7]. There are many types of radioactive wastes depending on their characteristics or management but generally, they can be classified into two groups: the low- and intermediate-level wastes and the short life wastes, and the high-level wastes [7]. Almost all the wastes that are produced belong to the first group. However, the wastes from the second group are the most dangerous [21, 39].

The high-level waste (HLW) and the spent fuel (SF) contain fission products and transuranic elements generated in the reactor core which are highly radioactive. They are also very hot due to radioactive decay [4, 39].

2.2. Final disposal

For the high-level wastes, the most often used choice is the deep storage. As shown in Fig. 1, the method of the final disposal is based on three protective barriers to avoid or delay contacts between the waste and the environment.

The first barrier is the metal canister. The most common metal used is copper. This is because copper is a good heat conductor and it is stable against heat and radiation.

The compacted bentonite is the second barrier. It is used as a buffer surrounding the capsules and also as a back-filler consisting of a mixture of bentonite and rock to seal the tunnel where the repository is located. Its functions are waterproofing and filtering.

Finally, the canister is placed into the bedrock at a depth of approximately 500 meters [34].

[1, 13, 14, 15, 25, 30, 36]

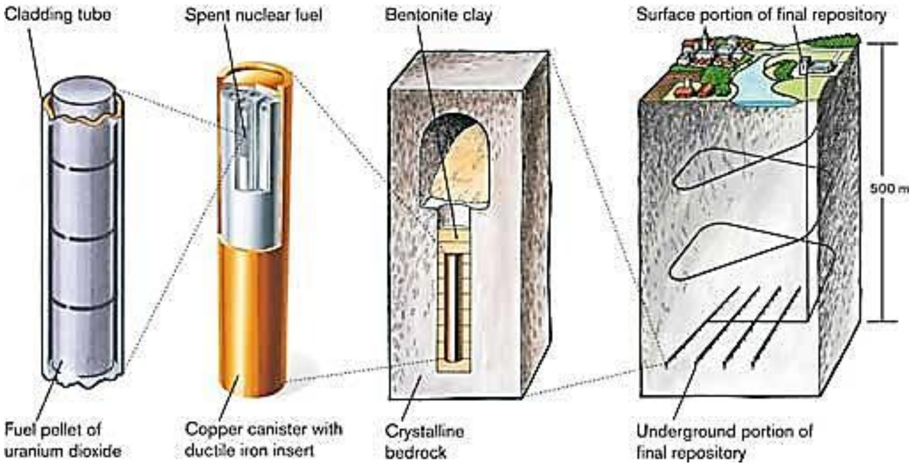

Figure 1 Graphic summary about final disposal for high level waste from SKB [30]

2.3. Bentonite

Natural bentonite is a type of clay that it is composed mainly of montmorillonite. This component is a swelling clay mineral of the smectite group [18]. Bentonite also contains nonswelling minerals such as quartz, feldspars, micas and carbonate, void and sand. The properties of bentonite are affected depending on the type of cation which is in the intermediate layer. The common interlayer cation is Na- and Ca-bentonite [20].

The choice to use bentonite instead of other clays is due to its properties [1, 37]:

- The low permeability ($\approx 3.10^{-14}$ m/s) to minimize the arrival of water
- The high thermal conductivity (0.6 to 1.4 W /mK) to dissipate heat generated from waste and from gas of the corrosion of the containers
- The high swelling capacity (7 MPa) to seal the cracks and fractures to prevent the access to other barriers
- The large specific surface (725 m^2 /g) to retain radionuclide as to keep the water chemistry conservation
- The high plasticity (liquid limit 102%, plastic limit 53%) for mechanical protection to ensure mechanical integrity of the capsule
- The high suction capacity to sealing the repository

Depending on where the clay is, there are different kinds of bentonite and each type of bentonite has got different properties. For example, from USA the bentonite is called Wyoming bentonite, Kunigel clay from Japan, FEBEX from Spain or FoCa7 from

France [27, 37]. Although different types of bentonite exist the most important part of those is the montmorillonite.

The montmorillonite participate in the most important properties of bentonite so it is advisable to know more about this component.

The structure of montmorillonite can be described in the Fig. 2 as a tetrahedral-octahedral-tetrahedral (T-O-T) unit layer. It consists of an alumina octahedral sheet sandwiched between two silica tetrahedral sheets [35]. The TOT layers have dimensions on the ‘a’ and ‘b’ directions and stack together, one above the other, in the ‘c’ direction to form particles.

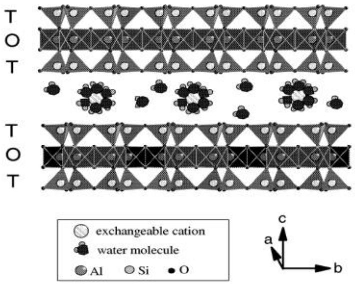

Figure 2 Representation of montmorillonite structure [18]

Moreover, the unit layer is shown specifically in Fig. 3. On the one hand, the octahedral sheet is composed of aluminium or magnesium with oxygen atoms or hydroxyl groups. On the other hand, the tetrahedral structure is composed of a silica sheet and three of the four oxygen atoms in each tetrahedron to form a hexagonal net [2, 8, 9, 12, 18].

Among the montmorillonite layers, the repulsive and attractive forces act. These forces are responsible for the changes of montmorillonite as the swelling pressure and the swelling deformation of the montmorillonite minerals [12].

The unit layer has on the faces a permanent negative charge of the octahedral and tetrahedral sheet because of isomorphous substitutions [18]. However, there are negative and positive charges on the edge. Hence, the charge is compensated because the cations are adsorbed from the edge layer to the sheet face [2, 24].

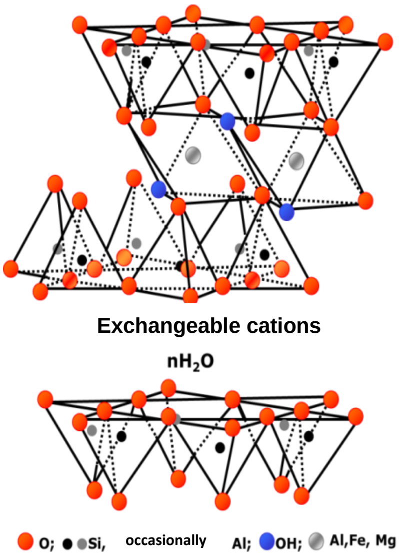

Figure 3 Montmorillonite structure [3]

Water and other polar molecules can enter among the unit layers, causing expansion in the interlayer swelling. There are small amounts of different exchangeable cations such as sodium, potassium, magnesium or calcium. The common exchangeable cations are sodium and calcium. Most studies assume that all charge compensating cations are sodium [8].

Sposito [31] said that the exchangeable cations and the method for the determination of the particle composition are two factors to predict the number of unit layer forming particles of montmorillonite, and the number varies from one to 16 [40]. Also, he explains the difference of the unit layer particles between the Na-montmorillonite and the Ca-montmorillonite in the homoionic form.

The difference between Ca or Na –montmorillonite is simple. The sodium montmorillonite characteristically swells into water many times its dry volume. However, the calcium- and the hydrous micas in general, do not increase too much in volume on hydration. Besides, the chemical composition with respect to the kind and degree of the isomorphous replacements in their structure, and to the amount and nature of their associated exchangeable cations may be the cause about differences in the swelling of the clays [5, 38].

3. Mechanism of the bentonite swelling

3.1. Swelling process

The behaviour of the swelling pressure of compacted bentonite can be described into three steps: before, during and after water uptake.

As it is shown in the Fig. 4, before water uptake, bentonite is composed of a mixture of montmorillonite, void, other nonswelling minerals and sand particles. Through this step, the voids are occupied by air and free water.

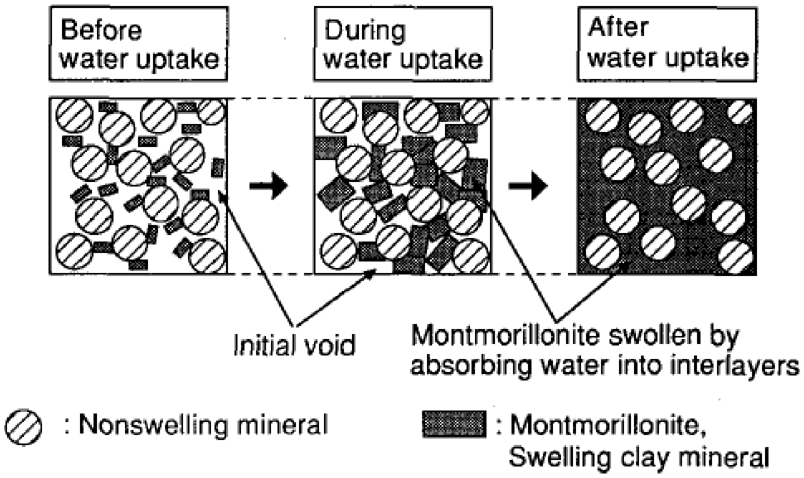

Figure 4 Mechanism on the swelling pressure of compacted bentonite from Komine and Ogata [12]

During the water uptake, montmorillonite absorbs water and into interlayers and swells occupying the void in the bentonite. Therefore, the volume of montmorillonite increases and the swelling pressure occurs. As it will be shown in the following section, the interlayer and the double-layer forces are responsible for the swelling behaviour of bentonite.

Finally, after water uptake, there is not any void to absorb hence the volume of montmorillonite cannot increase more. At this point, the swelling pressure of compacted bentonite can be measured.
[12]

3.2. Swelling behaviour

The swelling behaviour of compacted bentonite and sand mixtures is produced due to two mechanisms defined as crystalline swelling and osmotic swelling [20]. These mechanisms depend on the degree of hydration and on the cation in the interlayers. Besides, they act within the pore space of the clay. Both mechanisms are influenced by the breakup of montmorillonite particles and by the demixing of cation [17, 19].

Yong [40] stated that the difference between the crystalline swelling and the swelling due to osmotic activities of cations is because of the hydration structure of the water.

3.2.1. Crystalline swelling

The crystalline swelling is the first mechanism that appears when the montmorillonite absorbs water. This mechanism produces an enlarged in the distance in steps between the unit layers of montmorillonite. As a result, an increase of the volume of the montmorillonite is obtained and consequently, the swelling pressure appears.

Several forces act when the water comes into contact with the clay and these forces control the crystalline swelling. The dominant force is the hydration of the interlayer cations and the clay surface. Also, there are other forces as the van der Waals attraction or the Born repulsion. The hydration due to the interlayer swelling is assigned to the attraction between water molecules and the polar surface groups and also to the charge sites and exchangeable cations. It should be pointed out that the Van der Waals interaction participates in the energy of attraction.

On the one hand, in the dry state, the montmorillonite layers are almost in contact. The exchangeable cations located between the layers can be situated in two different places, on the surface of the layers or in the hexagonal holes of the tetrahedral sheets [20].

The interlayer cations and the van der Waals attraction are responsible for the strongly union among the negatively charged layers.

On the other hand, when the water is taken up by the exchangeable cations, they order themselves on a plane halfway between the clay layers [20].

It is not proven but it is supposed that firstly, the clay surface is covered fully by a layer of water molecules. After that, the second molecular layer will be distributed. To sum up, the water molecules are distributed layer after layer. In the case of the sodium montmorillonite, the water molecules are distributed from zero to four layers. It is a discrete process because interlayers cannot have a fractional number of layers of water molecules [17].

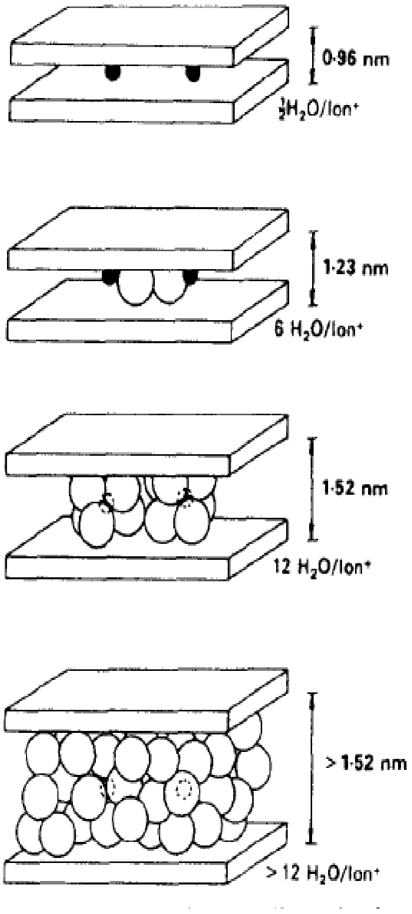

Figure 5 Process of water absorption from Madsen and Müller-Vonmoos [20]

As it is shown in the Fig. 6, during the water uptake, the negative dipoles of water molecules are disposed towards the cation. Hence, the electrostatic interaction between the layers and the interlayer cations decrease [20].

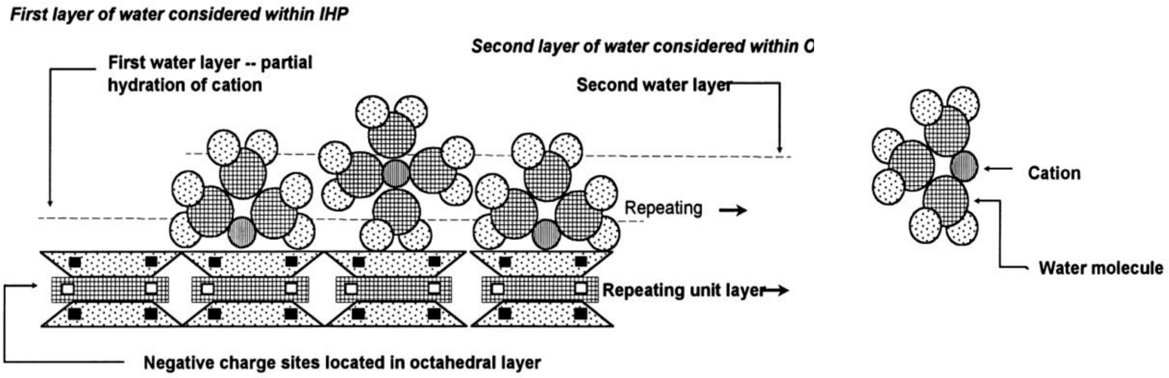

Figure 6 Arrangement of water on the clay layer from Yong [41]

The results are an increase in the distance between layers, a higher hydration energy than the bond between the clay layers and an increase of the volume of montmorillonite. Accordingly, the swelling pressure is resulting. The swelling process can completely reach the separation of layers because of the forces aforementioned.

Moreover, if there are organic compounds, the distance among the montmorillonite layers can be reduced because the organic compound makes the surface hydrophobic [20].

[16, 19, 20, 35, 40, 41]

3.2.2. Osmotic swelling

The osmotic swelling is the second mechanism that appears when montmorillonite absorbs water. This mechanism produces an increase of the distance continuously between the layers of montmorillonite.

The osmotic swelling is attributed to the difference in concentration between ions close to the surface of the layers of montmorillonite and in the pore water. This mechanism is produced due to the interactions of the diffuse double layers and the van der Waals attraction.

The clay layers are negatively charged and it leads to a repulsive force between layers. However, this negative charge is equilibrated by cations which are near to the surface of the layer. The exchangeable cations are not too strong by the external surfaces. Therefore, they tend to diffuse from regions of high concentration to the surface towards regions of low concentration in the bulk solution. The result is a diffuse cluster of ions around a clay particle. This system is called diffuse electric double layer.

Figure 7 shows the diffuse double layer. It is defined as a group of negative charge cluster on an external surface of clay and a cloud of positive ions positioned to the external surface.

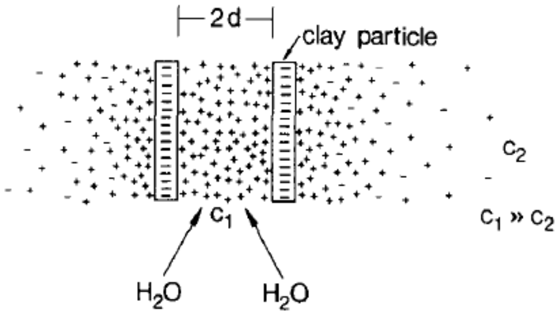

Figure 7 Diffuse double layer from Madsen and Müller-Vonmoos [20]

To form the diffuse double layer, the cations near to the surface balances the negative charge and then, the exchangeable cations tend to diffuse. As a result, a repulsive force among the overlapping double layers of the clay particles appears. This repulsive force varies with the balance and the radius of the counterions in the double layer and the concentration of electrolytes in the pore water [19].

[8, 16, 19, 20, 35, 40, 41]

4. Experimental work from literature

Before starting to explain this chapter, it should be pointed out that the experimental work is from the literature and any experimental test has not been done in this study. Therefore, the results of the experimental test from different studies have been used to compare their results with each other and also, they have been used to validate the predictions of our model.

4.1. Materials

The material used in the studies is natural bentonite, which is composed of around 65-75% montmorillonite, 10-14% quartz, 5-9% feldspar, 3-5% carbonate, 2-4% mica and chlorite, as well as 1-3% heavy minerals [18, 26].

The test solution used in the tests can be distilled water or saline solutions as Na, K, Cs, Mg, Ca and Al chloride solutions. The analytical grade reagents and the distilled water are used for preparing saline solutions [32]. This study is focused only the tests with distilled water.

The studies used to compare the results from experimental tests are from Karnland, [8]; Marcial, [22]; Mollins, [23]; Studds, [32]; Sun, [33]; Tripathy, [34]; and Wang, [38].

For the sensitivity analysis that will be shown in the chapter 6, the properties of bentonite and sand used in the different studies, are summarized in the next table. It can observe that the bentonite in each study is different and also the studies use different types of bentonites. Karnland, [8]; Marcial, [22]; Mollins, [23] and Studds, [32] work with Wyoming bentonite while Sun [33] works with Kunigel bentonite. In addition, pure bentonite is used in the study of Karnland [8]. Throughout all investigations studied in this analysis, the Na- montmorillonite is predominant.

Table 1 Profile of the bentonite and the sand used

Physical properties of bentonite and sand	Unit	Karnland	Marcial	Mollins	Sun	Studds
Bentonite content α	%	100	100	5,10, 20,100	10,17,20, 30,50,100	10,20, 100
Density of bentonite ρb	kg/m^3	2820	2650	2760	2790	2751
Montmorillonite content β	%	96	85	80	48	90
σ0	C/ m2	-0,111	-0,094	-0,118	-0,103	-0,118
CEC	eq/kg	0,86	0,68	0,95	0,732	0,95
EXCNa	eq/kg	0.809	0.60	0.534	0.405	0.534
EXCK	eq/kg	0	0.00	0.015	0.009	0.015
EXCMg	eq/kg	0.034	0.03	0.125	0.03	0.125
EXCCa	eq/kg	0.017	0.05	0.276	0.287	0.276

The sample of the mixture of compacted bentonite and sand used in each test is considered to be completely saturated at the end of the process of the water uptake because after each experiment, the degree of saturation of the specimen was 100% or higher [13, 33].

4.2. Test procedure

There are different methods to obtain the swelling pressure and the swelling deformation of compacted bentonites. Depending on the test used, the results will be different. Some of the tests are the swelling pressure test, the swelling deformation test, the zero swell method or the swell consolidation method. The most common are the swelling pressure test and the swelling deformation test. Therefore, they will be described below.

There is not difference between the result of swelling between the swelling pressure test and the swelling deformation test. However, the swelling deformation test is more complex and it gives more information.

4.2.1. Swelling pressure test

It is also known as the constant-volume test*.* This method measures the swelling pressure of bentonite and sand mixture. The most important characteristic of this test is that maintaining the volume constant is involved.

We can observe in Fig. 8 the constant-volume cell. It mainly consists of three parts: the bottom part, the middle cell and the top part. The first mentioned is composed of a drainage system for the solution circulation, and a porous stone. The middle cell has got two or more air outlets. Also, it is the part where the sample is established. At the top, there is a total pressure sensor for monitoring the swelling pressure.

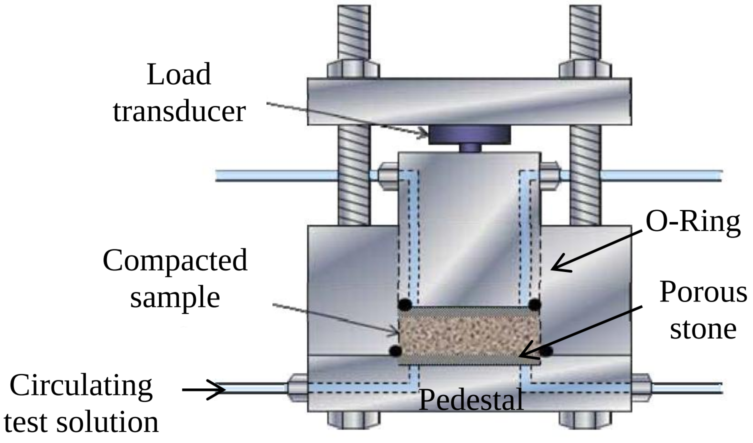

Figure 8 Schematic illustration of a constant-volume cell from Kiviranta and Kumpulainen [12]

The steps followed in this method are:

- Firstly, the sample of a mixture of bentonite and sand is compacted at the established density on the pedestal.
- An initial vertical pressure is applied to the sample and the distilled water is supplied from the bottom while the specimen is confined. Distilled water is used for saturation of the specimen.
- The samples swells
- After flooding the soil sample and the reaching stable pressure, the swelling pressure can be measured by the load transducer.

During the test procedure, the volume change is considered to be negligible.

[8, 10, 13, 18, 38]

Figure 9 Image of a constant-volume cell from Kiviranta and Kumpulainen [11]

4.2.2. Swelling deformation test

This method measures the vertical swelling deformation of the sand-bentonite mixture. Also, the swelling deformation test is used to study the effect of pre-existing technical voids on the swelling pressure. The most important characteristic of this test is that maintaining the pressure constant is carried out. This means that the sample can swell freely. The swelling deformation test apparatus is shown in Fig. 10. It shows the main parts of the apparatus.

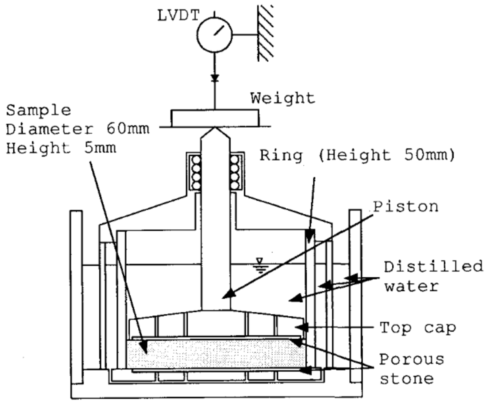

Figure 10 Schematic illustration of a swelling deformation apparatus from Komine and Ogata [12]

The process of this method can be described into the following steps:

- The sample of a mixture of bentonite and sand is compacted and the compacted samples are set up on the consolidation ring.
- After that, an initial vertical pressure is applied and hence, the compacted sample is pushed out from the ring. While this happens, the distilled water goes into contact to the sample.
- The sample can be freely swollen in the axial direction.
- Finally, the swelling deformation can be measured.

The result of this test is the relationship between the axial swelling deformation and the time required from the start of water supply.

Pre- swelling test is a combination of the swelling deformation method. In this test, the procedure is very similar to the method described above. In that case, the sample swells in the axial direction until an established value. Then, the piston is fixed and because of that, the swelling pressure is generated. [8, 13, 16,18, 33, 37]

5. Model development

5.1. Basic concepts

In order to understand some relevant parameters, Fig. 11 is shown. The behaviour of the mixture of compacted bentonite and sand, as well as the comparison of the swelling pressure, is defined. Here, it is appreciated the composition of the mixture and the notation.

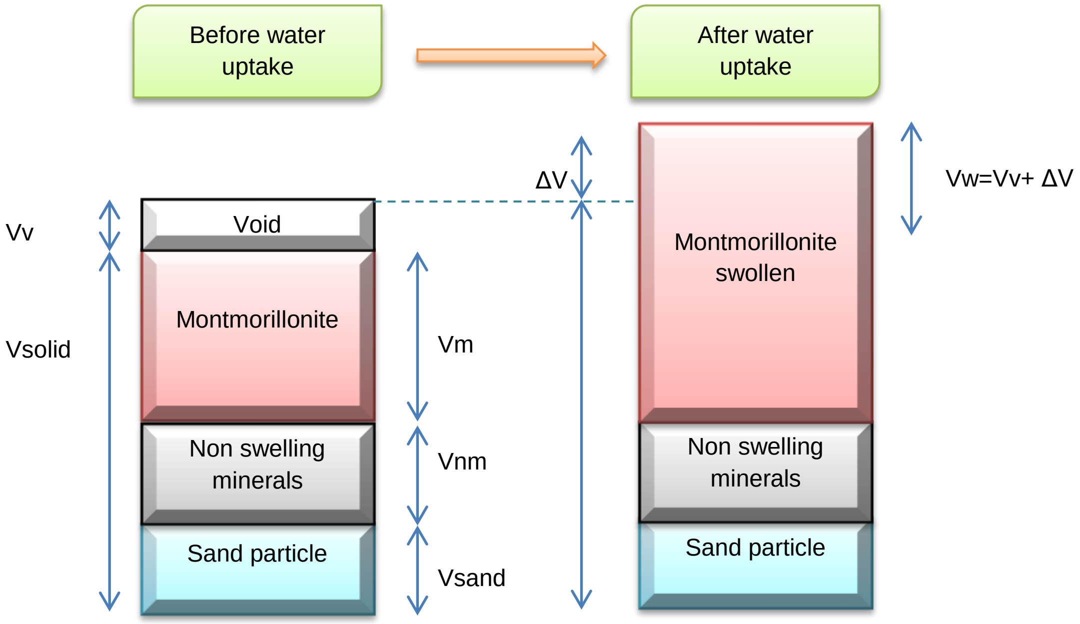

Figure 11 Conceptualization of the composition of the mixture of bentonite and sand particles from Liu [19]

As shown in the figure above and in Eq. 1, it is observed that before water uptake, the volume of solid, V_{solid} ,is the total of the sum of montmorillonite volume V_m, non-swelling minerals volume V_{nm} and sand particles volume V_{sand}.

$$\mathbf{V}_{solid} = \mathbf{V}_m + \mathbf{V}_{nm} + \mathbf{V}_{sand} \quad (1)$$

After water uptake, the water volume at saturation V_w is taken into account and the void volume disappears. It is important to mention this fact because by knowing the value of this volume the void ratio of montmorillonite can be defined. It is shown in Eq. 2 as

$$\mathbf{V} = \mathbf{V}_{\boldsymbol{solid}} + \mathbf{V}_{\boldsymbol{w}} \tag{2}$$

where V is the total volume of the material, after water uptake.

The void ratio of montmorillonite, e_m, is used to describe and to compare the behaviour of bentonite and sand mixture. It can be defined as we can see in Eq. 2

$$\boldsymbol{e_m = \frac{V_w}{V_m}} \tag{3}$$

Also, the void ratio of montmorillonite can be calculated from other equations as shown in Eq. 3 and Eq. 4, using different parameters about bentonite and sand that we should know.

$$\boldsymbol{e_m = e \cdot \frac{\rho_m}{\rho_s \cdot \alpha \cdot C_m}} \tag{4}$$

$$\boldsymbol{e_m = \frac{\rho_m}{\rho_{dry,m}} - 1} \tag{5}$$

where e is the final void ratio, ρ_m and ρ_s are the density of montmorillonite and solid in mixture, respectively. Cm and α are the content, of montmorillonite and of bentonite, respectively. The dry density of montmorillonite is illustrated as $\rho_{dry,m}$.

Another way to describe and to compare the behaviour of bentonite and sand mixture is using the clay void ratio, ec. It is defined in Eq. 5 as

$$\boldsymbol{e_c = e_m \cdot C_m \cdot \frac{\rho_{clay}}{\rho_m}} \tag{6}$$

where ρ_{clay} is the density of sand particles.

Also, knowing the dry density of bentonite $\rho_{dry,c}$, it is described the system of the mixture as the one that involves only the interaction between montmorillonite and water.

$$\boldsymbol{\rho_{dry,c} = \frac{\rho_{clay}}{e_c + 1}} \tag{7}$$

$$\boldsymbol{\rho_{dry,c} = \frac{1}{\frac{C_m}{\rho_{dry,m}} + \frac{1 - C_m}{\rho_{nm}}}} \tag{8}$$

The non-swelling density is known as ρ_{nm}

These equations have been used in this thesis, but in other cases it could not be known all the parameters required. Therefore, there are other equations using other parameters and it shows in Liu, [18, 19].

5.2. Description of the model

In this section, I briefly present the model that was initially developed by Liu [19] to predict the swelling pressures of different types of bentonite in dilute solutions [19].

Based on the thermodynamic relation and on the DLVO theory [16, 17,19, 24, 29, 34], by also assuming that the montmorillonite particles are uniformly distributed throughout the bentonite aggregate water system, the swelling pressure can generally be given as

$$\boldsymbol{p = p_{CCS} + p_{DDL} - p_{vdW}} \tag{9}$$

In Eq. 8, p_{CCS} is referred to the swelling pressure exerted in crystalline swelling mechanism and it is based on the thermodynamic relation. In this case, the swelling pressure depends only on the separation distance between the particles, h, referred to the water uptake. This is because, as it is said before in chapter 3, crystalline swelling is due mainly to hydration of the interlayer cations and the clay surface. Other forces are slightly manifested.

Particularly, the swelling pressure can be defined as

$$\boldsymbol{p_{CCS} = k \cdot e^{-h/l}} \tag{10}$$

where k and l are two constants, the pre-exponential factor and the decay length, respectively.

On the other hand, p_{DDL} and p_{vdW} are, respectively, referred to the osmotic swelling pressure due to the diffuse double layer and the van der Waals attraction respectively. They are based on the DLVO theory.

The diffuse double layer swelling pressure depends on the separation distance between particles h, on the ionic concentration of the pore solution c and on the valence v of exchangeable cations.

$$\boldsymbol{p_{DDL} = 2 \cdot c \cdot R \cdot T \cdot (\cosh y^m - 1)} \tag{11}$$

In Eq. 10, R is the gas constant; T is the absolute temperature and y^m is the scaled potential at the mid-point among the plate-like montmorillonite particles.

Moreover, the van der Waals attraction depends only on the separation distance between particles, h.

$$\boldsymbol{p_{vdW}} = \frac{\boldsymbol{A_H}}{\boldsymbol{6\pi}}\left\{\frac{\boldsymbol{1}}{\boldsymbol{h^3}} - \frac{\boldsymbol{2}}{(\boldsymbol{h}+\boldsymbol{D_p})^3} + \frac{\boldsymbol{1}}{(\boldsymbol{h}+\boldsymbol{2}\cdot\boldsymbol{D_p})^3}\right\} \tag{12}$$

where A_H is the constant of Hamaker and D_p is the thickness of montmorillonite particles.

Having said that, we can proceed to demonstrate and discuss in the following chapter for the predicted swelling pressure of different types of bentonites and bentonite-aggregate mixtures in distilled water based on the model presented above.

6. Results and discussion

6.1. Results from experimental test

In this work, for the purpose of comparisons, experimental results from literatures [8, 22, 31, 32, 33, 37] are used. The swelling pressures are presented with respect to the dry density, the void ratio of the bentonite or void ratio of the montmorillonite from different experimental studies. Those swelling pressure of different bentonite are also compared with each other since, as it shown previously in the table 1, the bentonite has got different properties from different studies.

The meaning of the names from the legend in Fig. 12-14 is described below in the table 2:

Table 2 Legend from Fig. 12, Fig. 13 and Fig. 14

Name from legend	Study	Bentonite content (%)	Reference
W70	Wang	70	[38]
M5, M10, M20, M100	Mollins	5, 10, 20, 100	[23]
S10, S20, S100	Studds	10, 20, 100	[32]
T100	Tripathy	100	[34]
D10, D17, D20, D30, D50, D100	Sun	10, 17, 20 ,30 50, 100	[33]
[K(1), K(2), K(3), K(4)]100	Karland	100	[8]

In the following figure, i.e., Fig. 12, it can be observed that there is a linear relationship between the void ratio of the montmorillonite and the swelling pressure for each type of bentonite.

It is also shown that, during the process of water uptake, the swelling pressure increases and the montmorillonite void ratio decreases. This is because the montmorillonite swells and occupies the void of bentonite as what is shown before in chapter 3.

In addition, the linear relationship between the swelling pressure and the void ratio of the montmorillonite should be noted with a great importance. This is because that the linear relationship can be conveniently adopted for calculations in engineering applications to predict the swelling pressure of Na- montmorillonite.

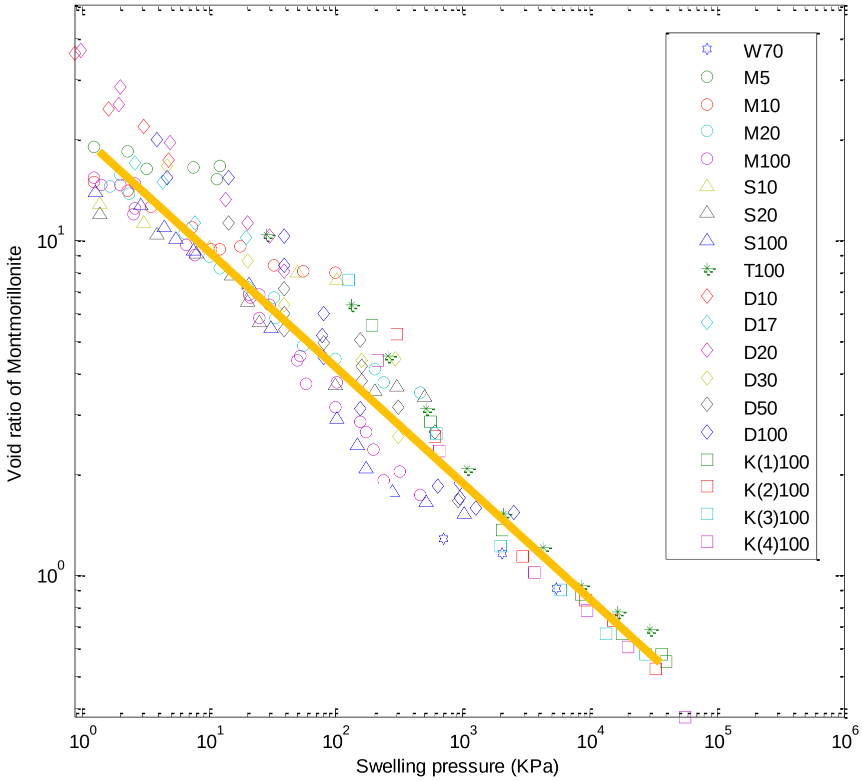

Figure 12 Comparison between the swelling pressure and the void ratio of montmorillonite

Figure 13 displays the swelling pressure with respect to the clay void ratio in a liner scale. As what has been indicated in Fig. 12 for the case of the montmorillonite, the clay void of bentonite decreases while the swelling pressure increases.

It should, however, be stressed that the scale of the clay void of bentonite, in this figure, is linear as what is different from that of the void ratio of montmorillonite in Fig. 12. In Fig. 13, it is observed that the swelling pressures experience two patterns depending on the clay-void ratio of bentonite, as what are indicated by the auxiliary lines. In particularly, the bentonite swelling behaviour in region 1 suggests to the crystalline swelling. In this region the swelling pressure is very strong. On the other hand, region 2 corresponds to the osmotic swelling. In this case, the swelling is weaker than in the crystalline swelling. The reason is that since there is always a void of bentonite to be occupied, and consequently the dominant mechanism at the beginning of the swelling process is the crystalline swelling. This has been observed in the experimental studies [8, 23, 32, 33, 34, 38], and it has been also reported that the osmotic swelling starts while montmorillonite takes up water and continue to occupy the voids. At the end of the process, osmotic swelling is the dominant method but swelling pressure is less strong. This can be understood that the diffuse double layer repulsion, as one mechanism of the osmotic swelling, is now decreasing to a large extent since the diffuse double layers do not interact with each other when the interlayer separation is very large at the end of the swelling process. In addition, at the middle of the process, both mechanisms are involved.

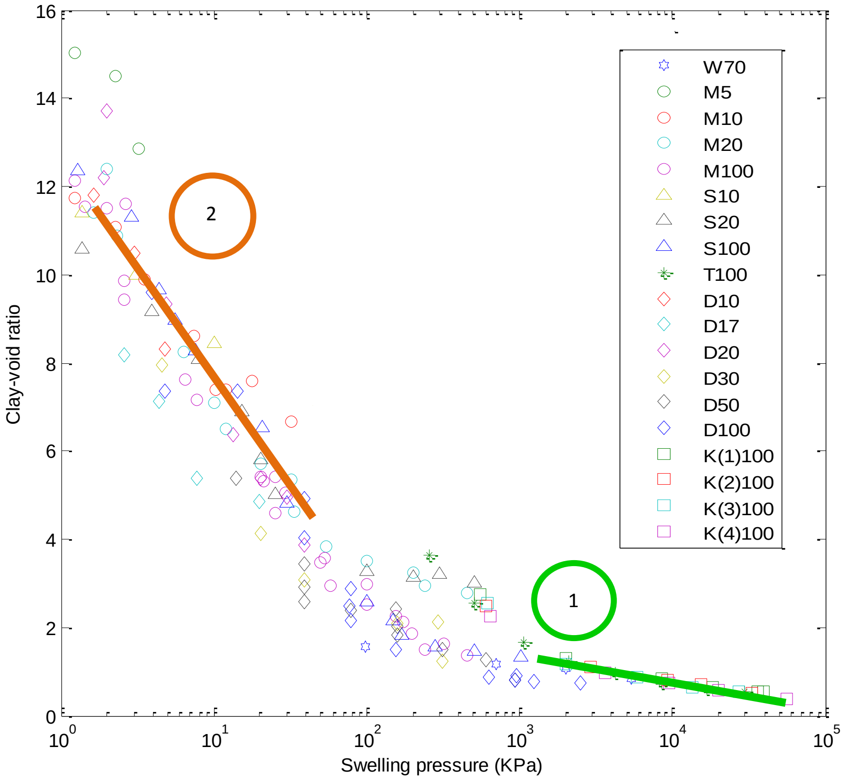

Figure 13 Comparison between the swelling pressure and the clay void ratio

The last comparison from experimental test results is the relationship between the swelling pressure and the final dry density. In Fig. 14, it is observed that the increase of the swelling pressure is proportional to the increase of the final dry density.

The importance of this graphic shows the reality of the studies. Most of the studies used to disregard the early stage of the swelling pressure in their experimental process, as what are shown in the region between 200 and 300 KPa approximately of the final dry density of the bentonite, and therefore they obtained a linear relationship to predict the swelling pressure.

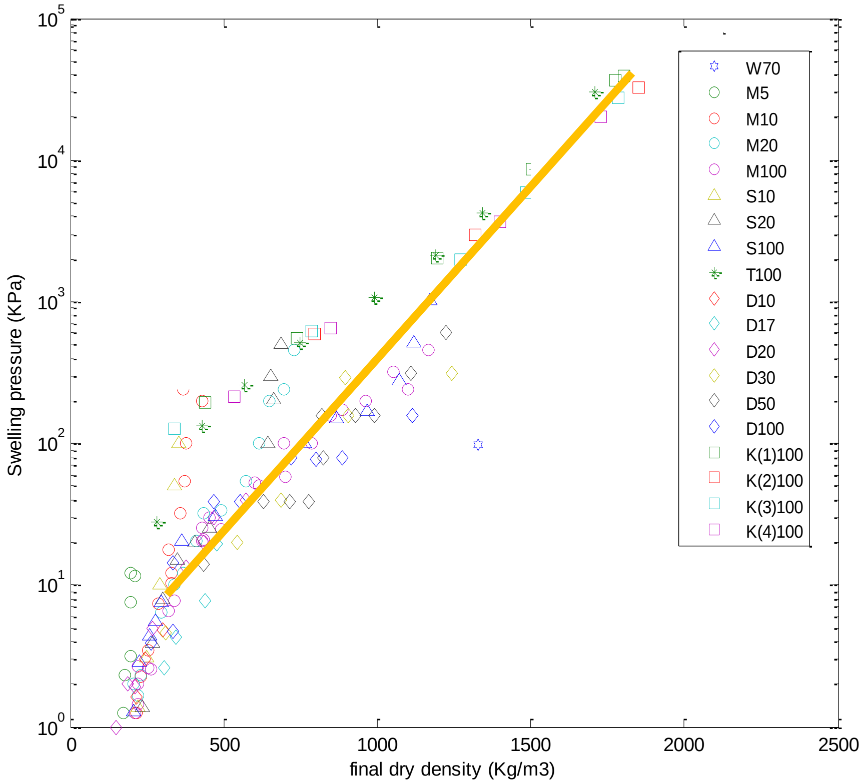

Figure 14 Comparison between the swelling pressure and the final dry density of bentonite

6.2. Validation of the model

Using the model previously described in chapter 5 and comparing it with the results of experimental tests, it is observed in Fig. 15 that the predictions of the model give the same or very similar results as the test results.

The findings in Fig. 15 shows that the model works well in each type of bentonite because in the case a), it is compared the Wyoming bentonite while in case b), the type of bentonite is Kunigel. There are more investigations studied, but the result is the same in each case.

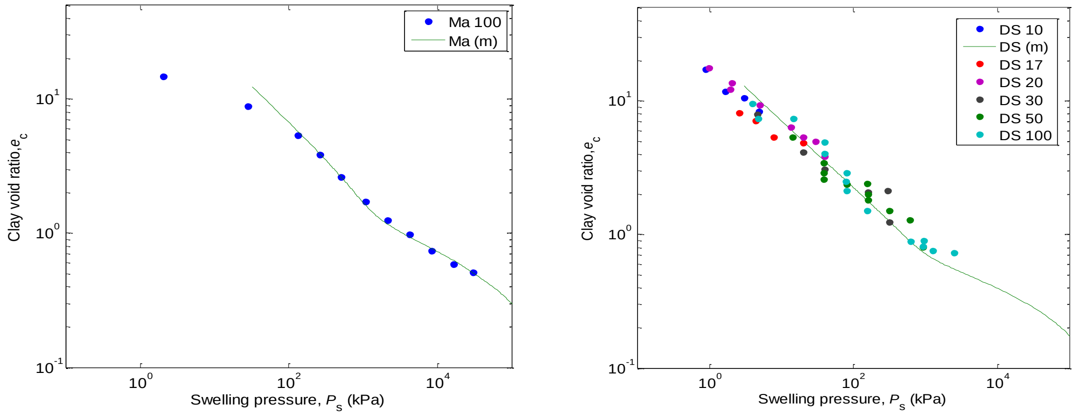

Figure 15 Comparison of measured swelling pressure between model prediction and laboratory test results. a) Marcial [22], b) Sun [33]

6.3. Sensitivity analysis

The last aim raised in this thesis is the study of different variables through the model developed to predict the swelling pressure of bentonite and sand mixture. The sensitivity analyses of the swelling pressure will be carried out by varying:

- the number of stacked unit layers, n_s
- montmorillonite content, C_m
- the amount of the exchangeable cations, $ExCa$
- the ratio, B
- the pre-exponential factor, k
- the decay length, l

Every sensitivity analysis has been carried out independently, meaning that all the changes on response have been studied with the variation of a single parameter at once.

The following table shows the values of the parameters used in the model of swelling pressure

Table 3 Physical constants used in the predictions from Liu [19]

Parameters	Unit	Value
Faraday constant (F)	C/mol	9.64853x10^{4}
Gas constant (R)	$J \cdot K/mol$	8.31446
Temperature (T)	*K*	298
Permittivity of vacuum (e_0)	F/m	8.8542x10^{-12}
Relative dielectric constant of water (e_r)	-	78.54
Density of water (ρ_w)	kg/m^3	1000
Density of montmorillonite (ρ_m)	kg/m^3	2770
Thickness of the unit layers (σ_s)	*nm*	0.96
Fixed separation between the Na-m unit layers (d_i)	*nm*	1.2
Hamaker constant (A_H)	*J*	2.2x10^{-20}
Reference pressure (p_r)	*Pa*	1000

6.3.1. Varying the number of stacked unit layers

The first sensitivity analyses studies the swelling pressure respect to the void ratio of bentonite with the number of stacked unit layers from one to ten.

The meaning of the stacked unit layers can be described in Fig. 16. It is composed of a determinate number of unit layers of montmorillonite and they are separated by a fixed distance between the unit layers, d_m

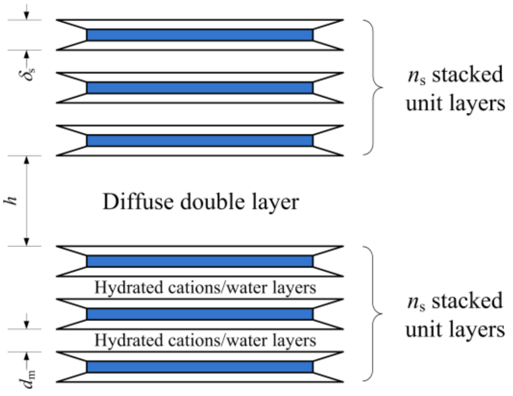

Figure 16 Representation of the montmorillonite particles [19]

In addition, the distance between the montmorillonite particles depends on the shacked unit layer as we can observed in Eq. 13

$$h = n_s \cdot \delta_s \cdot e_m - (n_s - 1) \cdot d_m \tag{13}$$

where δ_s is the thickness of the unit layers of montmorillonite.

In figure 17, the variation of the model prediction is shown because of the change of the number of stacked unit layers. In this case, the variation is shown between swelling pressure and clay void ratio.

When the number of stacked unit layers increases, it observes that as the process happens, the swelling pressure decreases. This happens because the distance between the layers is getting larger.

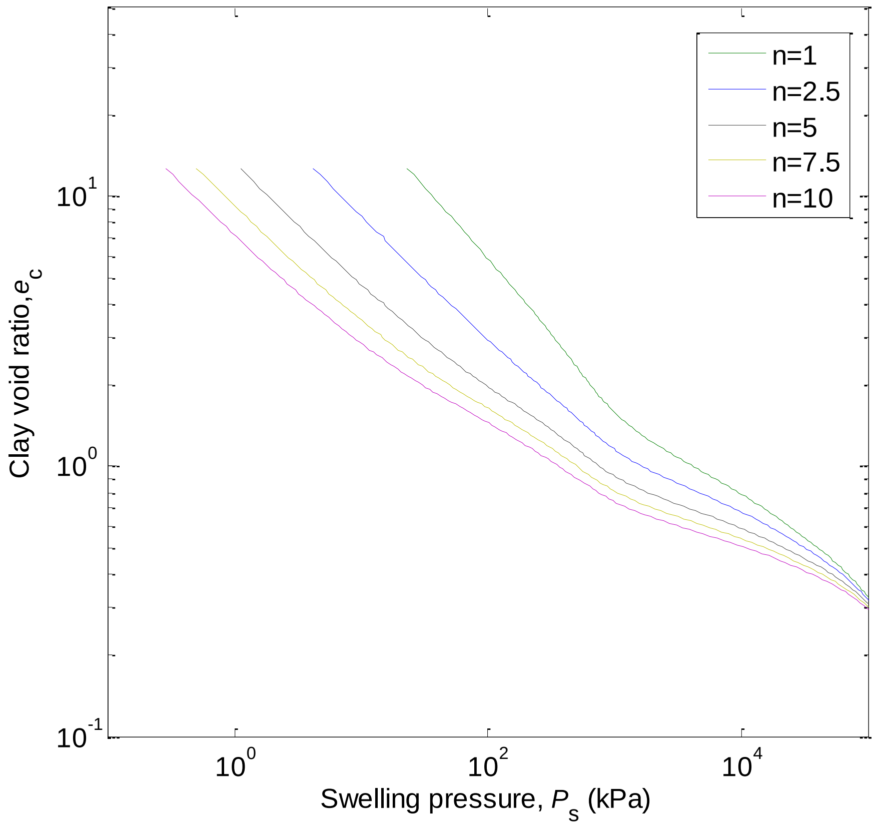

Figure 17 Comparison of the predicted model of the swelling pressure as a function of the clay void ratio varying the number of stacked unit layers

6.3.2. Varying the montmorillonite content

The second variable compared is the montmorillonite content.

The montmorillonite is the only component of bentonite which absorbs water and the only component that can swell. Hence, the relationship between the swelling pressure and the clay void ratio depending on the montmorillonite content is shown in Fig. 18. It is observed that at a same clay void ratio, the swelling pressure is increasing proportionally as the montmorillonite content is increasing.

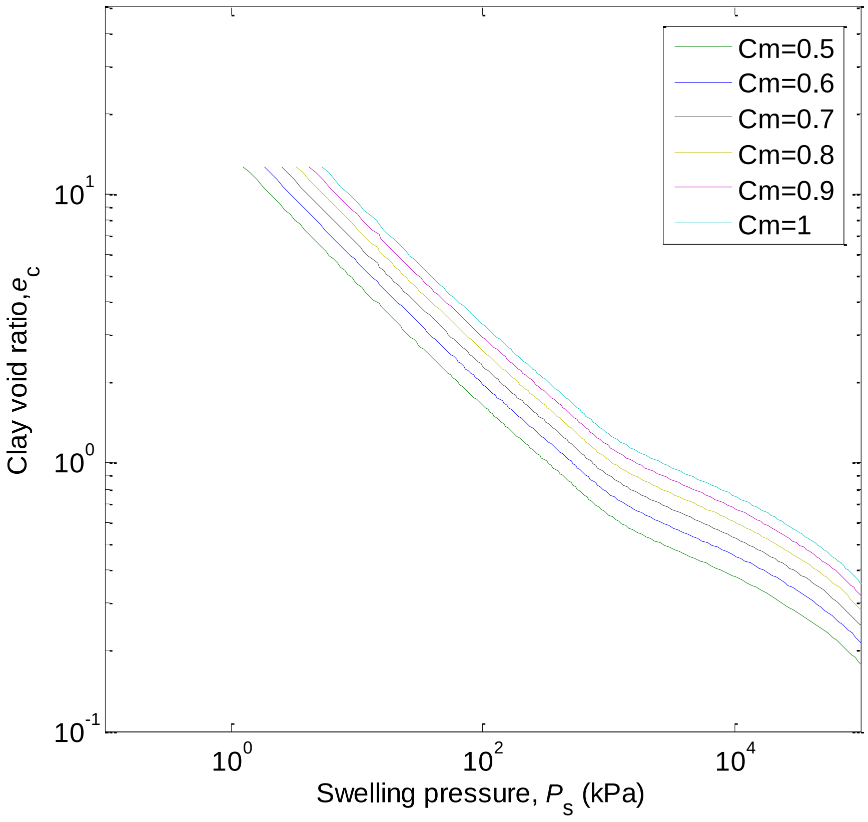

Figure 18 Comparison of the predicted model of the swelling pressure as a function of the clay void ratio varying the content of montmorillonite

In addition, the relation between the swelling pressure and the montmorillonite void ratio depending on the montmorillonite content does not change instead of the clay void ratio in Fig. 18. This is because the only component that can absorb water and swell is montmorillonite.

6.3.3. Varying the amount of the exchangeable cations

In this case, it has been varied proportionally the amount of the exchangeable cations increasing the amount of the sodium cation and decreasing proportionally the amount of the other cations. The cations are sodium, potassium, magnesium and calcium. The table of values is shown below:

Table 4 Different amounts of the exchangeable cations from the experimental data of Studds [32]

	ExCa			
Test	Na	K	Mg	Ca
Data	0.534	0.015	0.125	0.276
1	0.500	0.016	0.135	0.299
2	0.600	0.013	0.105	0.232
3	0.700	0.009	0.075	0.166
4	0.800	0.005	0.045	0.100
5	0.900	0.002	0.015	0.033

It shows in the Fig. 19 that when the Na- bentonite dominant, the variation of the swelling pressure is almost insignificant. The reason is that the swelling pressure depends mainly on the distance between particles.

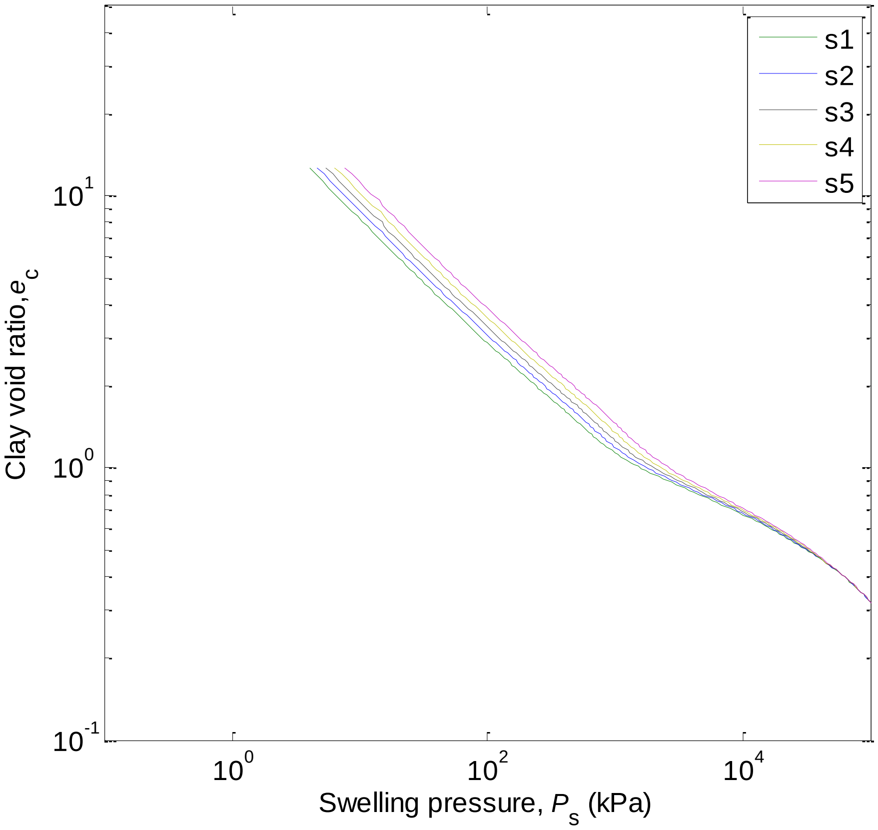

Figure 19 Comparison of the predicted model of the swelling pressure as a function of the clay void ratio varying the content of montmorillonite

6.3.4. Varying the ratio of crystalline swelling region

Figure 20 shows that the variation of the ratio of crystalline swelling region from one to six does not depend on the swelling pressure. A slight change can be detected in the crystalline swelling region. This parameter describes the extent of "just initiated" crystalline swelling and generally, this value is fixed as unity to disregard the process of "just initiated" crystalline swelling, unless otherwise stated [19].

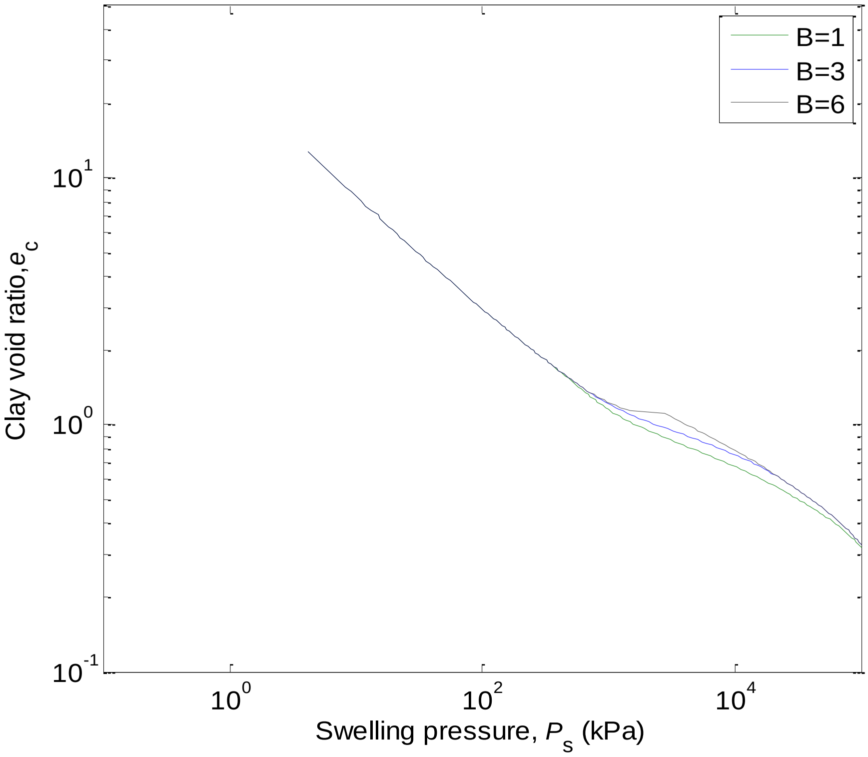

Figure 20 Comparison of the predicted model of the swelling pressure as a function of the clay void ratio varying the ratio of crystalline swelling region

6.3.5. Varying the pre-exponential factor and the decay length

The pre-exponential factor k and the decay length l are two parameters that are associated with the crystalline swelling. The crystalline swelling is a mechanism very complex to define because there are many forces involved. However, it can be describe with an empirical equation. As it is shown in chapter 5, the Eq. 10 describes the swelling pressure referred to the crystalline swelling.

$$\boldsymbol{p_{CCS} = k \cdot e^{-h/l}} \tag{14}$$

On the one hand, if the distance between particles h is equal to zero, the crystalline swelling pressure is equal to k. Therefore, the crystalline swelling pressure reaches its maximum value. On the other hand, if h is equal to the decay length l, the crystalline swelling pressure decreases.

In Fig. 21, the parameter studied is the pre-exponential factor. It observes that the swelling pressure respect to the clay void ratio only changes in the crystalline swelling region being greater when the pre-exponential factor increases for a same value of the clay void ratio.

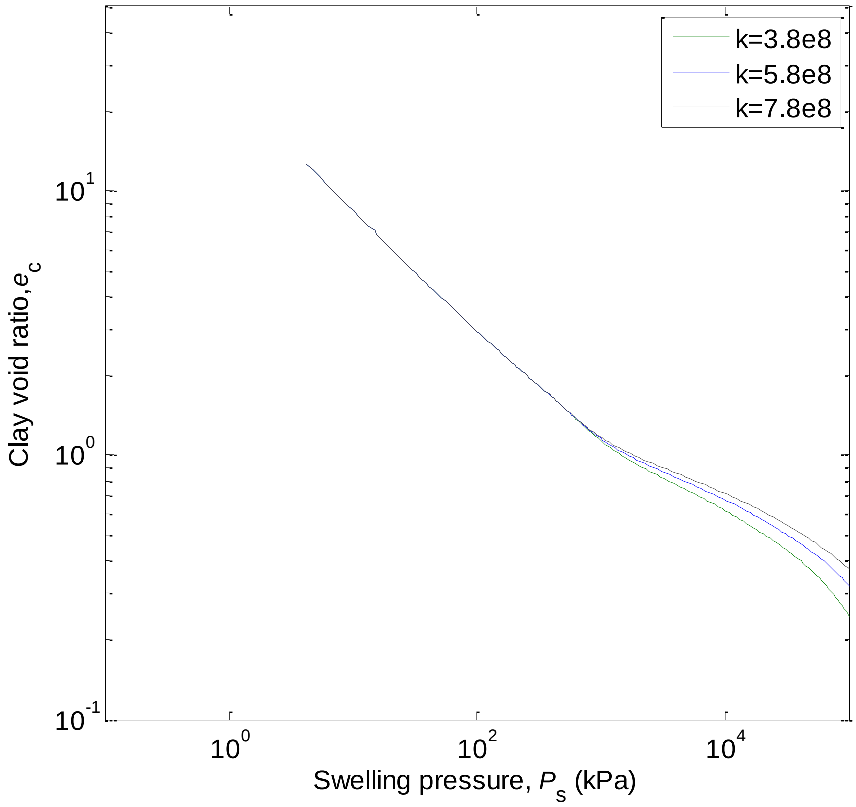

Figure 21 Comparison of the predicted model of the swelling pressure as a function of the clay void ratio varying the pre-exponential factor

Furthermore, if the parameter that varies is the decay length, it observes the same process as varying the pre-exponential factor. The relation between the swelling pressure and the clay void ratio only changes in the crystalline swelling region and when the decay length increases, the swelling pressure also increases for a same value of the clay void ratio. However, the difference between the Fig. 21 and the Fig. 22 is that in Fig. 22 the variation of the swelling pressure is more accentuated.

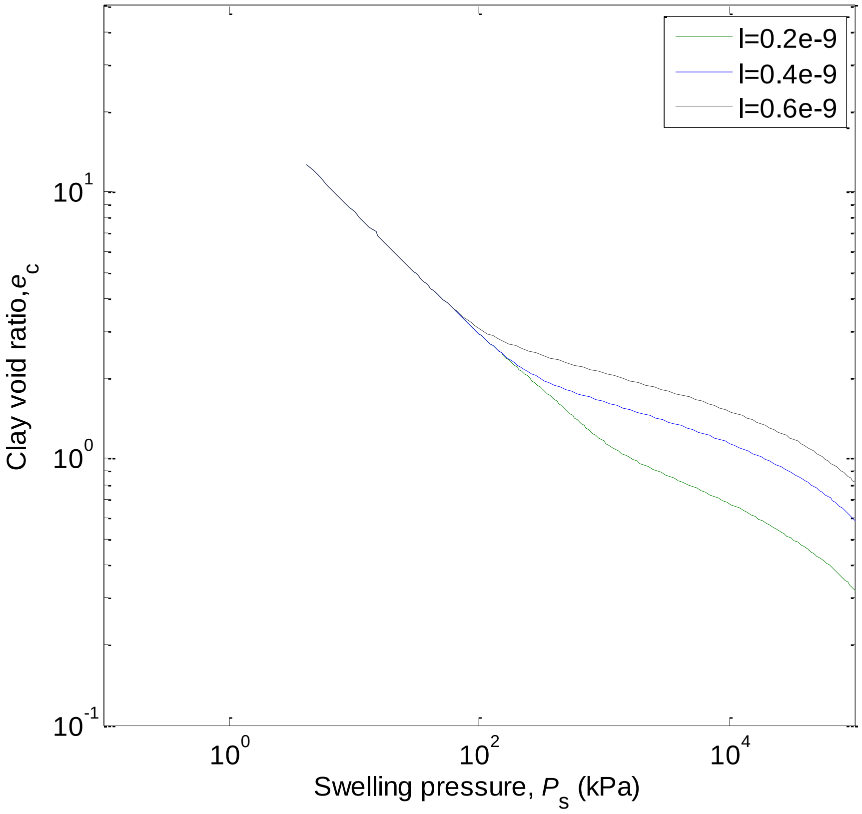

Figure 22 Comparison of the predicted model of the swelling pressure as a function of the clay void ratio varying the decay length

7. Conclusions

By summing up the results of this study on the behavior of the swelling pressure, this chapter summarizes the conclusions obtained from those results.

The conclusions have been divided into two different sections. Firstly, it will describe the conclusions from the results of the experimental test and then, the conclusions from the results taken from the model of the swelling pressures.

From the results of the different investigations aforementioned, the results of the swelling pressure of different kind of bentonites respect to the clay void ratio, the void ratio of montmorillonite and to the final dry density are compared. The sample used in each test is considered to be completely saturated and the Na- montmorillonite is predominant. Also the test solution is distilled water.

The first conclusion is that in the Na-montmorillonite there is only a unique relation among different bentonites. It is also observed that the compacted bentonite and sand mixture swollen depend on their final dry density. These two conclusions are very useful in engineering applications. The unique relation and the linear relation are important for calculations in engineering applications to predict the swelling pressure of Na- montmorillonite.

The other conclusion from the results of experimental tests is that the relation between the swelling pressure and the clay void ratio show the mechanism on the swelling pressure, crystalline swelling and osmotic swelling.

On the other hand, a mechanistic model has been developed and validated. The model is used to predict the swelling pressure of bentonite and sand mixture in distilled water, considering the Na-dominant bentonite and sand mixture.

Comparing to the predictions of the model and the results of experimental tests it can be concluded that the model in each kind of bentonite works well because both processes describe a same relation.

The model is also tested with a sensitivity analysis to obtain the different behaviours of the swelling pressure by varying several parameters. The crystalline swelling depends on the pre-exponential factor and on the decay length and the ratio of the crystalline swelling region describes the extent of "just initiated" crystalline swelling. It has been noted that the swelling pressure depends mainly on the distance between particles.

Also, it has been observed that the swelling pressure increases when the number of stacked unit layers decreases. Moreover, the only component of bentonite that can absorb water and swell is montmorillonite.

The sensitivity analyses indicate that the number of stacked unit layers and the decay length are parameters which should be fitted more carefully for future studies to validate this model with different bentonites.

To conclude this thesis, an idea of the future research should be focused on trying to find an empirical expression on the relationship between swelling pressure and final dry density. Also, this model can be used for engineering applications.

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