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## SUITABILITY OF A CRETACEOUS NATURAL Na-BENTONITE AS CONSTRUCTION MATERIAL FOR LANDFILL LINERS

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### Abstract

A natural Cretaceous sodium (Na) bentonite from North Patagonia, Argentine, is considered a potential construction material for landfill liners. It is studied alone and in mixtures with sand. Physical and chemical properties of the Na-bentonite were determined to verify the suitability of this material for the construction of low permeability mineral liners. Hydraulic conductivity measurements were conducted on pure bentonite and on mixtures of poorly graded sand with 3, 6, 9 and 12% of Na-bentonite. Compaction and hydraulic conductivity tests were performed using deionized water and a 1000 mol/m<sup>3</sup> CaCl<sub>2</sub> solution to determine the effects of a high concentrated saline solution on the hydraulic properties of compacted sand-bentonite mixtures. The experimental results confirm that the pure powdered bentonite meets current specifications to be used as part of a geosynthetic clay liner (GCL), and that ageing effects may affect the hydraulic behavior of this material. Compacted sand-bentonite mixtures required a minimum bentonite content of 6% in order to achieve hydraulic conductivities lower than 1 x 10<sup>-9</sup> m/s which is a regular requirement for hydraulic containment liners. However, in contact with a strong saline solution (1000 mol/m<sup>3</sup> CaCl<sub>2</sub>), the permeability of the mixtures increased between three and five orders of magnitude. This indicates that a higher percent of bentonite is required to reach an acceptable hydraulic conductivity value when the mixtures are permeated with a CaCl<sub>2</sub> solution. The results show that a new applicability was found for bentonites of regional provenance as well as high reserves which have a naturally very high amount of sodium.

*Key words:* clay liner, GCL, landfill liner, Na-bentonite, sand-bentonite mixture

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### 1. Introduction

Bentonite has been used for many years as a component of containment systems to construct low permeability layers in landfills. The usefulness of bentonites in such application can be attributed to their small particle size, high surface area, and high sorbing and swelling capacity (Churchman et al., 2002). Most of these characteristics are enhanced when the main exchangeable cation is sodium, making sodium bentonites especially suitable for base liner construction because of their low hydraulic

conductivity and high contaminant retention potential (Czurda, 2006). In addition, the presence of expandable clay minerals is preferred due to their self sealing capacity as consequence of the clay mineral hydration (Luo et al., 2015).

In this work, bentonite is generally utilized in combination with other natural or synthetic materials (e.g. compacted clay liners CCL or geosynthetic clay liners GCL) (Daniel, 1993). Bentonite requirements for this use are listed in Table 1.

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**Table 1.** Sodium bentonite requirements for a GCL or a CCL (data compiled from Daniel, 1993 and Maubeuge, 2002)

Requirement	Value
Montmorillonite content	75 - 90%
Plasticity index	140 - 380 %
Hydraulic conductivity	$< 5 \times 10^{-11}$ m/s
Swell index	$> 24$ mL
CEC	$> 94$ meq/100g

Sand-bentonite mixtures are increasingly been used for construction of CCL when clayey soils are not available (Chalermyanont and Arrykul, 2005; Stewart et al., 2003). These mixtures can meet hydraulic conductivity requirements without suffering shrinkage cracking (Mollins et al., 1996; Studds et al., 1998; Tay et al., 2001). The addition of bentonite to local soils is frequently related to the minimum hydraulic conductivity of  $1 \times 10^{-9}$  m/s for the construction of mineral barriers in landfills as required by the US Environmental Protection Agency (EPA) and current European Commission regulations. Bouazza and Van Impe (1998) compiled minimum requirements for containment barriers in different countries. The determination of the amount of bentonite required to achieve the desired hydraulic conductivity becomes very important when local soils are used as construction material for mineral liners. It is also important that the clay content is kept close to the minimum percentage required from hydraulic reasons in order to minimize costs and to avoid significant shrinkage cracking (Daniel and Benson, 1990; Gebrenegus et al., 2011).

The hydraulic conductivity of a liner built with a Na-bentonite can be altered due to the exchange of  $\text{Na}^+$  ions by polyvalent cations (Kolstad et al., 2004), due to precipitation/dissolution processes (VanGulck and Rowe, 2004) and due to clogging mechanisms (Francisca and Glatstein, 2010). In this way, properties of a GCL or a CCL can be modified when bentonite comes in contact with a leachate containing high concentrations of inorganic species (e.g. leachate from municipal solid waste containment facilities) (Benson et al., 2010; Guyonnet et al., 2009; Jo et al., 2001, 2004; Katsumi et al., 2008; Kolstad et al., 2004; Lee et al., 2005; Razakamanantsoa et al., 2012; Rowe, 2005; Shackelford et al., 2010; Shariatmadari et al., 2011; Studds et al., 1998).

Ageing effects of highly compacted swelling clays have been studied in the past to evaluate the long term performance of the barriers during the life of a waste repository system (Delage et al., 2006; Subba Rao and Tripathy, 2003). However, there are few studies which evaluate changes in the clay mineral properties during storage, and before their use as a construction material for GCL or CCL (Gaidzinski et al., 2011). The aims of this research included the evaluation of a Cretaceous natural Na-bentonite from North Patagonia Argentina as a suitable material for the production of geosynthetic clay liners or for the construction of compacted sand-

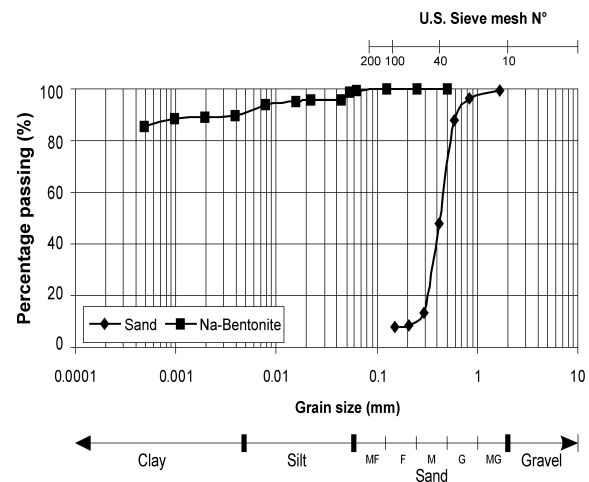
bentonite liners. The influence of the ageing of bentonite and permeating liquid properties on the hydraulic conductivity and intrinsic permeability of the tested sand-bentonite mixtures was also evaluated.

## 2. Materials and methods

### 2.1. Materials

The clay used in this study was a powdered natural Na-bentonite manufactured by Castiglione Pes & Cía., named CATAE (Table 3). This bentonite belongs to the facies associations B and C (Barrio, 1990) from the Upper Cretaceous Allen Formation, located in Neuquén basin, Argentina, and is often used as an additive in drilling muds. The bentonite deposit from which CATAE is extracted, is one of the biggest in South America with a calculated reserve of more than 3.000.000 tons and an extension of 69  $\text{Km}^2$  (Vallés and Impiccini, 1999).

The poorly graded sand ("SP" according to the Unified Soil Classification System) used in the experiments was a commercial fluvial sand. Uniformity and curvature coefficients of the tested sand were  $C_u = 1.58$  and  $C_c = 0.99$ , respectively. The grain size distribution curves of the bentonite and the sand are shown in Fig. 1.

**Fig. 1.** Grain size distribution curves

### 2.2. Methodology

Physical and chemical properties of the bentonite were determined by following standard tests. Grain size analyses were done by the pipette method. Atterberg limits and free swell tests were performed following ASTM D4318-10 (2010) and ASTM D5890-11 (2011), respectively. Specific gravity was measured by the pycnometer method on clay dried at 105 °C. X-ray diffraction (XRD), differential thermal analysis (DTA) and thermal gravimetric analysis (TGA) were conducted in order to determine the mineralogical composition of the bentonite. Physicochemical and chemical analyses included bulk lithochemical analysis by total digestion using the lithium metaborate/tetraborate

fusion/nitric acid method followed by ICP emission spectrometry (ICP-AES), specific surface determination by the EGME method and cation exchange capacity and exchangeable cations following the silver thiourea method (Chhabra et al., 1975).

Atterberg limits and swell index were measured using deionized water and a 1000 mol/m<sup>3</sup> CaCl<sub>2</sub> solution. The CaCl<sub>2</sub> was chosen as permeating liquid because previous studies had shown that calcium-dominated solutions produced significant alterations in the hydraulic conductivity of sodium bentonites (Stern and Shackelford, 1998).

### 2.2.1. Compaction test

Compaction tests were carried out to assess the optimum water contents and maximum dry unit weights of sand-bentonite mixtures. The amount of Na-bentonite used to prepare the samples were 0%, 3%, 6%, 9%, and 12% (weight by weight). For each sand-bentonite mixture, the optimum water content and the maximum dry unit weight were determined using the standard Proctor compaction method, ASTM D698-07e1 (2007). Five samples of the soil-bentonite mixture were prepared with different fluid contents ranging from 3 to 17%. Either water or CaCl<sub>2</sub> solution was added to the sand-bentonite mixtures to obtain the desired fluid contents. The sand-bentonite mixtures were allowed to hydrate for at least 24 hours prior compaction.

### 2.2.2. Hydraulic conductivity measurements

Two different cells were used to determine hydraulic properties. Test performed on pure bentonite were carried out in an oedometer cell while a rigid wall compaction-cell permeameter was used for the compacted sand-bentonite mixtures. In both cases, the hydraulic conductivity was determined by following the falling head test procedure.

Consolidation tests were carried out on Na-bentonite following ASTM D2435/D2435M-11 (2011), with the following effective vertical pressures: 53.64, 107.28, 268.21 536.52 and 1073 kPa. The range of vertical pressure represents overburden pressures exerted on clay liners used to seal hazardous waste landfills, sanitary landfills, waste piles and mining waste surface impoundments (Peirce et al., 1986). The experiments therefore simulate vertical loads applied to the bottom liners in landfills. Cells had a diameter of 6.3 cm and a height of 2.5 cm, respectively. Different specimens were prepared by adding deionized water to the powdered clay. The initial humidity was adapted equally to the bentonite liquid limit ensuring the maximum water content possible. Filter paper was placed at the bottom and top of the specimen. A top cap with a porous stone was placed above the sample. The entire assembly was placed in the consolidation cell and positioned in the loading frame.

The consolidation ring was immersed in deionized water. Primary consolidation of samples

was reached after seven days in all cases, and the degree of saturation at the end of the tests ranged from 90 to 98%.

Hydraulic conductivity of the pure bentonite was determined at two different times to evaluate if the properties of the clay had changed during storing. Identical samples were prepared and tested immediately after opening the CATAE bentonite bag and after 365 days of storing the open bag in the laboratory, under controlled environment conditions (temperature: 21 °C; relative humidity: 52% ).

Compacted sand-bentonite mixtures were prepared by adding a molding water content 2% wet of optimum which according to Mitchell et al. (1965) ensures optimum hydraulic properties and expected dispersed clay microstructure. The specimens were compacted in the testing cell in three layers of equal height by giving 25 blows per layer with a 10.95 mm in diameter blunt tip bar.

Deionized water and CaCl<sub>2</sub> solution were permeated through the specimens of 9 cm in width and 5 cm in height, from top to bottom. A burette was connected to the permeameters (either the oedometer or the compaction-mold cell) via a Teflon tube. The hydraulic gradient was set by filling the burette with the testing liquid. After placement in the permeameters, the specimens were hydrated during 48 hours and thereafter the permeating liquid was introduced from the top of the cell. Hydraulic gradients varied from 25 to 16 according to the expected hydraulic conductivity values (ASTM D5856-95, 2007). The hydraulic conductivity was measured at different times and the tests were terminated when the following criteria was achieved (ASTM D5856-95, 2007; Rosin-Paumier and Touze-Foltz, 2012): 1) a steady hydraulic conductivity; 2) the ratio of outflow to inflow rate was between 0.75 and 1.25; and 3) a minimum of two pore volumes of flow (PVF) passed through the specimen.. The time of testing varied between 30 and 120 days for the highest and lowest permeable sample, respectively. The evolution of hydraulic conductivity with the time of permeation was monitored in order to achieve steady state values. At that stage no changes in the saturation degree are expected. The specimens were disassembled after testing, and samples that were permeated with the CaCl<sub>2</sub> solution were tested by XRD to verify the presence of exchangeable cations in the clay mineral. Table 2 summarizes the different tests, soils, liquids and standards used in this research. All experiments were conducted at least in duplicate and the results represent the mean value.

The hydraulic conductivity of a soil,  $k$  (m/s), depends on various physical factors, including properties of the soil and fluid. Conversely, the intrinsic permeability,  $K$  (m<sup>2</sup>/s), is considered as a property of the porous media, independent of fluid properties. Therefore, the intrinsic permeability of the sand-bentonite mixtures was also calculated to assess the influence of the permeant.

**Table 2.** Summary of testing program for the geotechnical properties

Property	Tested material	Permeant liquid	Method
Atterberg limits	Powdered bentonite	DW and CaCl <sub>2</sub>	ASTM D4318-10
Swell Index	Powdered bentonite immediately after open the storing bag	DW and CaCl <sub>2</sub>	ASTM D5890-11
Swell Index	Powdered bentonite after 365 days of storing	DW and CaCl <sub>2</sub>	ASTM D5890-11
Compaction tests	Sand-Bentonite mixtures	DW and CaCl <sub>2</sub>	ASTM D698-07e1
Hydraulic conductivity	Sand-Bentonite mixtures	DW and CaCl <sub>2</sub>	ASTM D5856-95
Hydraulic conductivity	Powdered bentonite immediately after open the storing bag	DW	ASTM D2434-11
Hydraulic conductivity	Powdered bentonite after 365 days of storing	DW	ASTM D2434-11

**Table 3.** Selected physical and chemical properties of the Na-bentonite CATAE

Properties	Na-Bentonite CATAE
Clay (<2 μm) content (%)	90
Cation exchange capacity (meq/100g)	104.6
Specific surface area (m <sup>2</sup> /g)	754.5
Montmorillonite content (%)	90
Activity	3.14
Specific gravity	2.70
Exchangeable cations (%)	
Na <sup>+</sup>	76.9
Ca <sup>2+</sup>	12.4
Mg <sup>2+</sup>	10.9
K <sup>+</sup>	0.3
<b>Major elements (%)</b>	
SiO <sub>2</sub>	56.6
Al <sub>2</sub> O <sub>3</sub>	18.7
Fe <sub>2</sub> O <sub>3</sub>	4.9
MgO	2.6
CaO	1.1
Na <sub>2</sub> O	3.1
K <sub>2</sub> O	0.3
TiO <sub>2</sub>	0.4
P <sub>2</sub> O <sub>5</sub>	0.09
MnO	0.06
Ignition loss	11.3

The Kozeny-Carman equation presents a theoretical relationship between k, K and relevant soil and fluid properties as follow (Eq. 1) (Mitchell and Soga, 2005):

$$K = k \left( \frac{\mu}{\rho g} \right) = \frac{1}{k_0 T^2 S_0^2} \left( \frac{e^3}{1+e} \right) S^3 \quad (1)$$

where *g* = gravity, *ρ* = fluid density, *μ* = fluid viscosity, *T* = tortuosity, *k<sub>0</sub>* = pore shape factor, *S<sub>0</sub>* = wetted surface area per unit volume of particles, *e* = void ratio and *S* = degree of saturation.

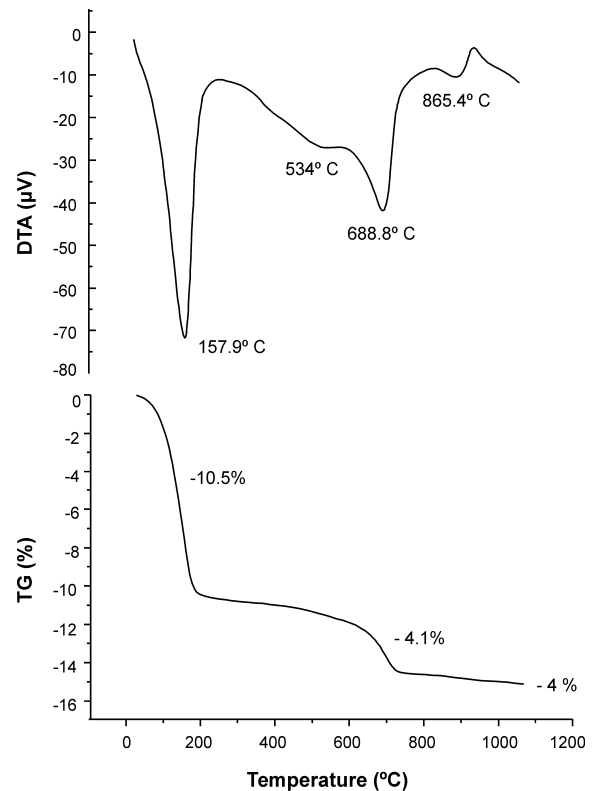
### 3. Results and discussion

#### 3.1. Na-Bentonite properties

According to the grain size analysis, the bentonite contained 90% of particles with a grain size lower than 2 μm (Fig. 1). The XRD test showed that the Na-bentonite was mainly composed by montmorillonite (90% in weight) which is consistent with the obtained plasticity index (PI), high cation exchange capacity and specific surface area (Tables 1 and 2). In addition, chemical analysis showed that the

bentonite was mainly composed by silicon oxide and aluminum oxide, and that the main exchangeable cation was Na<sup>+</sup>.

The obtained DTA and TGA curves evidenced all the typical thermal effects of a montmorillonite (Fig. 2). The weight loss in the TGA curve and the prominent endothermic peak near 150 °C in the DTA curve were ascribed to interlayer water associated with the exchangeable Na<sup>+</sup> ions. The weight loss and the endothermic peaks at 688 °C were associated with the loss of structural water through dehydroxylation of the montmorillonite layers.



**Fig. 2.** Differential Thermal Analysis (DTA) and Thermal Gravimetric Analysis (TGA) curves of the Na-bentonite CATAE

The Atterberg limits confirmed that the bentonite has a very high plasticity and swelling potential. However, in contact with the highly concentrated CaCl<sub>2</sub> solution, the liquid limit and swell index decreased drastically (Table 4). Decreased swelling and water retention (PI) occurred as a consequence of an increased electrolyte concentration and cation valence in the pore fluid. This is expected in accordance with the diffuse

double layer theory (DDL) for clays with such a high amount of expandable clay minerals (Arasan and Yetimoglu, 2008; Gleason et al., 1997; Jo et al., 2001; Katsumi et al., 2008; Mitchell and Soga, 2005; Montoro and Francisca, 2010; Shackelford et al., 2000).

**Table 4.** Atterberg limits and swell index of the Na-bentonite CATAE determined with deionized water (DW) and a 1000 mol/m<sup>3</sup> CaCl<sub>2</sub> solution

Properties	DW	CaCl <sub>2</sub>
Liquid Limit (%)	340	96
Plasticity index (%)	291	23
Swell index (mL)	36	7
Swell index in aged clay (mL)	28	7

Swell index of the powdered bentonite, after one year of storing, was lower than the obtained for the freshly mined bentonite. This behavior can be attributed to weathering or ageing processes that take place during the storing of the clay mineral (Table 4). In this process, water molecules placed in metastable positions may evaporate, whereas water molecules of the air become fixed in more stable positions (Gaidzinski et al., 2011). The bentonite CATAE revealed, after one year of ageing, a loss of moisture from 11% to 10% which may have caused the aggregation of clay particles affecting the microstructure of the clay and therefore the water retention capacity (Delage et al., 2006). This behavior is in good agreement with the decrease observed in the swell index.

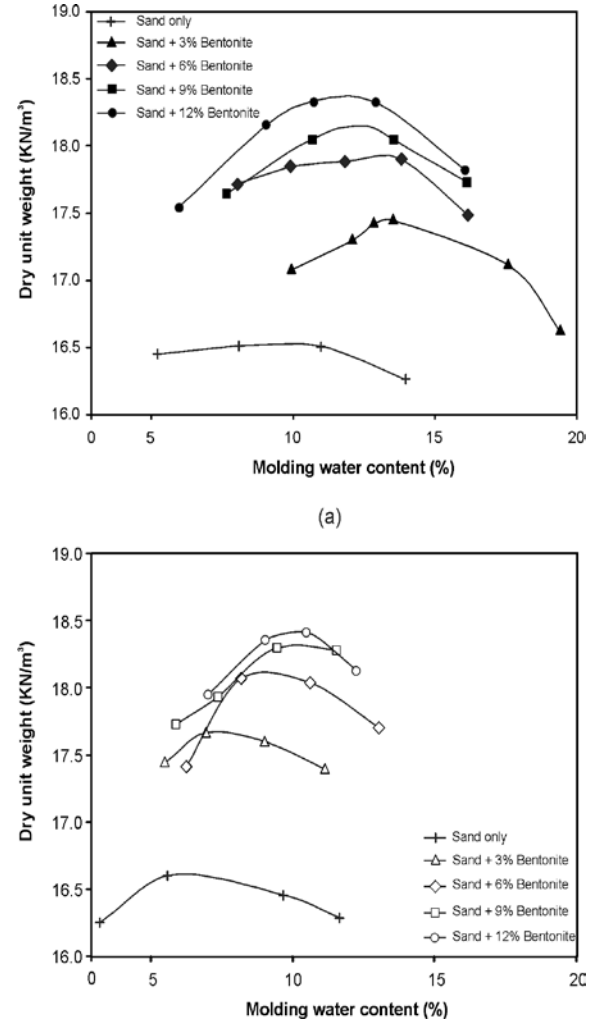
3.2. Compaction test results

The maximum dry unit weights and corresponding optimum water contents were obtained from the peak of compaction curves for the sand-bentonite mixtures which were compacted with either deionized water or CaCl<sub>2</sub> solution (Fig. 3). The presence of high ionic concentration in the compaction fluid affected the shape of the compaction curves.

All specimens compacted with deionized water (Fig. 3a) showed very similar optimum water contents (sand only 10%; 3% bentonite 13%; 6% bentonite 14.5%; 9% bentonite 12.5%; 12% bentonite 12%), while maximum dry unit weights increased with the amount of bentonite (sand only 16.5 kN/m<sup>3</sup>; 3% bentonite 17.4 kN/m<sup>3</sup>; 6% bentonite 17.8 kN/m<sup>3</sup>; 9% bentonite 18.0 kN/m<sup>3</sup>; 12% bentonite 18.3 kN/m<sup>3</sup>).

Compaction tests of sand-bentonite mixtures using the CaCl<sub>2</sub> solution as compaction fluid showed an increase in optimum water content (sand only 5.5%; 3% bentonite 7%; 6% bentonite 9.5%; 9% bentonite 10.5%; 12% bentonite 11%) and dry unit weight (3% bentonite 17.6 kN/m<sup>3</sup>; 6% bentonite 18.1 kN/m<sup>3</sup>; 9% bentonite 18.3 kN/m<sup>3</sup>; 12% bentonite 18.4 kN/m<sup>3</sup>) with the bentonite content (Fig. 3b). In addition, optimum water contents determined for the specimens compacted with the CaCl<sub>2</sub> solution were

approximately 2% lower than those obtained using deionized water, showing that the use of a saline strong solution facilitated to achieve a more flocculated state obtained at lower water contents and higher dry densities. This trend evidences the effect of the exchangeable cations on water retention capacity (Kaufhold et al., 2010).



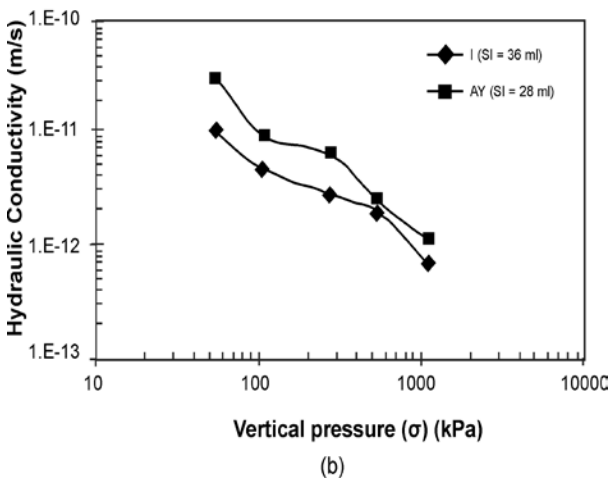
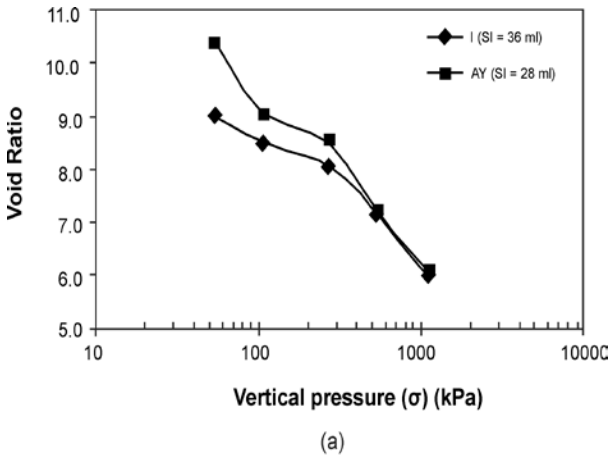
**Fig. 3.** Compaction curves of sand-bentonite mixtures: (a) Specimens compacted with distilled water; (b) Specimens compacted with a 1000 mol/m<sup>3</sup> CaCl<sub>2</sub> solution

3.3. Hydraulic conductivity of pure bentonite

Hydraulic conductivity and void ratio of the Na-bentonite as function of effective vertical pressure are shown in Fig. 4. Consolidation and hydraulic conductivity of the pure bentonite was measured on twin samples: one sample was prepared and tested immediately after opening the bentonite bag and the second one after one year of open storage.

Both void ratio and hydraulic conductivity decreased with the applied pressure demonstrating how close both parameters are related. The hydraulic conductivity of the bentonite increased approximately half order of magnitude with the time of air exposure of bentonite before the experiment

(Fig. 4b). This change in hydraulic conductivity can be also associated with ageing effects giving that particle aggregation, which would be responsible for the decrease in the swell index, would also affect the hydraulic properties of the clay. However, the causes of ageing of the clays are not yet thoroughly understood at this moment, and the mechanisms responsible for the changes in hydraulic conductivity and swell index through time should be evaluated in future research.



**Fig. 4.** Influence of the effective vertical pressure on compressibility (a) and hydraulic conductivity (b) of pure Na-bentonite hydrated with deionized water tested immediately (I) and after 1 year (AY) of open storage. SI= swell index

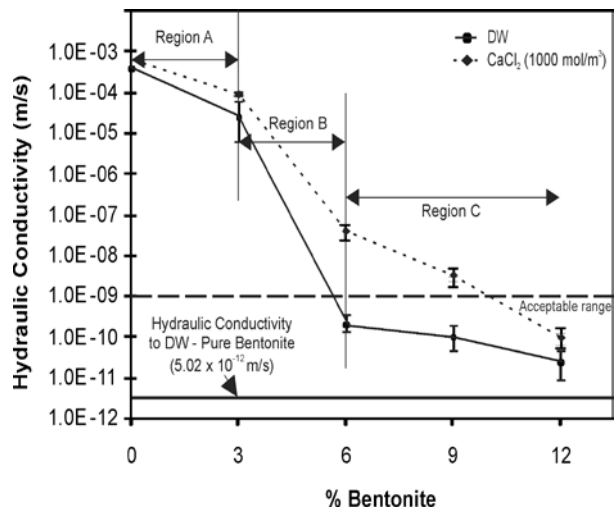
There are no previous studies reporting an increase in the hydraulic conductivity of bentonite associated to ageing processes of the material under storing, before its use. This effect should be taken into account if the bentonite will be used for the construction of clay liners.

The pure bentonite showed an extremely high compressibility, and can only be used in clay liners mixed with other -mechanically more stable-materials (Musso et al., 2010). However, according to the properties listed in Table 2 and results shown in Fig. 4b, the Na-bentonite meets the requirements in terms of montmorillonite percent, swell index,

plasticity index, cation exchange capacity and hydraulic conductivity stipulated in Table 1, and may be suitable to be used as a component of geosynthetic clay liners (GCL).

### 3.4. Hydraulic conductivity of sand-bentonite mixtures

The hydraulic conductivity of the sand-bentonite mixtures measured with water, decreased with increasing bentonite content (Fig. 5). This phenomenon was previously observed by many investigators (Chalermyanont and Arrykul, 2005; Chapuis, 1990; Kenney et al., 1992). The decreasing hydraulic conductivity was characterized by three regions in agreement with the results obtained by Abichou et al. (2002). In region A, the hydraulic conductivity showed a slight decrease as the bentonite content increased from 0 to 3%. In region B, the hydraulic conductivity dropped dramatically by more than four orders of magnitude when the bentonite content increased from 3 to 6%. Further increase of the bentonite content above 6% did not generate a significant reduction of the hydraulic conductivity (region C). At the highest bentonite content, the hydraulic conductivity of the sand-bentonite mixtures approached that of pure bentonite indicating that the flow rate was mainly controlled by the clay particles while the sand grains behaved as impervious inclusions in the clay matrix (Kenney et al., 1992).

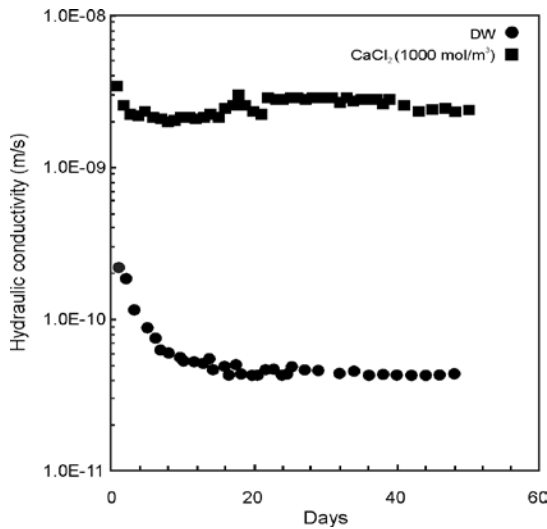


**Fig. 5.** Hydraulic conductivity vs. bentonite content of sand-bentonite mixtures permeated with deionized water (DW) and a CaCl<sub>2</sub> (1000 mol/m<sup>3</sup>) solution. Error bars indicate a 95% confidence interval

The hydraulic conductivity of the mixtures permeated with a CaCl<sub>2</sub> (1000 mol/m<sup>3</sup>) solution was higher than that measured with water, but the influence of bentonite content resulted quite similar in both cases. However, region C was more sensitive to the bentonite content and the hydraulic conductivity decreased three orders of magnitude from 6% to 12%. In this case, the acceptable range of

hydraulic conductivity was reached only with 12% of bentonite. This result is in good agreement with the more flocculated state of the specimen in contact with the CaCl<sub>2</sub> solution.

Hydraulic conductivity of sand-bentonite mixtures permeated with deionized water showed a gradual decrease with time reflecting progressive hydration of the swelling clay minerals (Fig. 6). In the case of the samples permeated with a CaCl<sub>2</sub> (1000 mol/m<sup>3</sup>) solution, hydraulic conductivity firstly decreased and after 20 days of testing increased slightly until reaching an equilibrium condition (i.e., hydraulic conductivity steady over time). This trend is related to the partial exchange of Na<sup>+</sup> with Ca<sup>2+</sup> during the test.

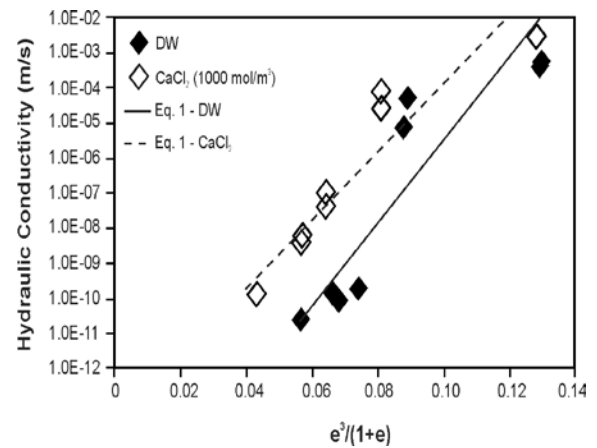


**Fig. 6.** Hydraulic conductivity as a function of time. Example for a sand-bentonite mixture with 9% of bentonite content

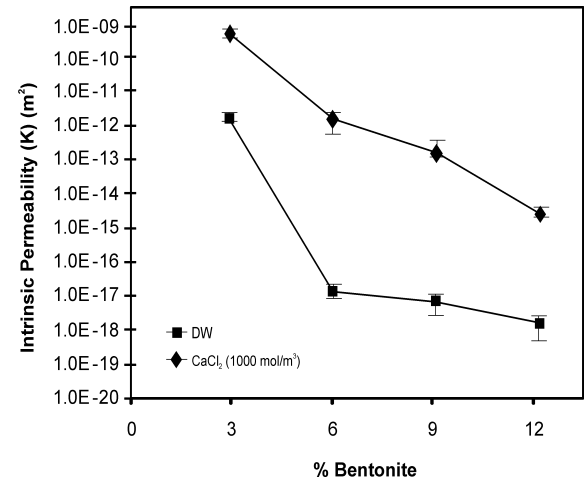
Liquid displacement in porous media depends on pore space properties such as void ratio, pore shape, tortuosity and spatial variability of pore sizes (Fetter, 1993). As the bentonite content increases, the void ratio of the sand-bentonite mixtures decreases because macro-pores are occupied by the swelling bentonite creating a more tortuous flow path.

Hydraulic conductivity of the compacted sand-bentonite mixtures permeated with either distilled water or the CaCl<sub>2</sub> solution showed a fairly well linear dependence with the theoretical void ratio function revealed in the Kozeny-Carman's equation (Fig. 7). The obtained coefficients of determinations were R<sup>2</sup> = 0.76 and R<sup>2</sup> = 0.88 for the specimens permeated with deionized water and the CaCl<sub>2</sub> solution, respectively. This trend confirms that the experimental results could be well represented by the theoretical Kozeny-Carman equation (Bear, 1972). Comparisons between hydraulic conductivities determined with water and CaCl<sub>2</sub> solution were made in terms of their intrinsic permeability K (m<sup>2</sup>) to eliminate the influence of the liquid properties (viscosity and density). The data shows that very high bentonite content is required to reach a

satisfactory hydraulic conductivity when the mixture is exposed to a 1000 mol/m<sup>3</sup> CaCl<sub>2</sub> solution (Fig. 8).



**Fig. 7.** Relationship between Kozeny-Carman void ratio function and hydraulic conductivity of sand-bentonite mixtures permeated with deionized water (DW) and a 1000 mol/m<sup>3</sup> CaCl<sub>2</sub> solution



**Fig. 8.** Influence of the bentonite content on the intrinsic permeability of sand-bentonite mixtures permeated with deionized water (DW) and a 1000 mol/m<sup>3</sup> CaCl<sub>2</sub> solution

Intrinsic permeability increased from 3 to 5 orders of magnitude when the sand-bentonite mixtures were permeated with this solution, confirming that there was a change in pore structure (Fig. 8). This change is determined by the related decrease of the DDL thickness at higher ion concentrations. As the bentonite contains expandable minerals (weak interlayer bonding) and most of water is forming the DDL around particles, any change of intrinsic permeability can be associated to expansion/shrinkage mechanisms in response to changes in the DDL thickness (Montoro and Francisca, 2010).

This evolution of soil fabric emerges at macroscale as the observed increase of intrinsic permeability shown in Fig. 8. The results are in line with previous studies which reported increments from 2 to 5 orders of magnitude in permeability

values of sand-bentonite mixtures (Jo et al., 2001; Lee and Shackelford, 2005; Petrov and Rowe 1997).

The observed increase in hydraulic conductivity of the sand-bentonite mixtures upon permeation with a highly concentrated  $\text{CaCl}_2$  solution confirmed that different microstructures develop in the clay fraction due to the integration of  $\text{Ca}^{2+}$  ions into the diffuse double layer. Flocculation of the montmorillonite particles probably occurred due to the presence of a higher calcium concentration (Gleason et al., 1997; Shackelford, 1994) and generated an increase in effective pore size, as well as a decreasing micropores and increasing macropores volume (Rao and Mathew, 1995).

Mineralogical analysis of the Na-bentonite was performed before and after permeation with the  $\text{CaCl}_2$  solution. A variation in basal spacing  $d_{001}$  from 12.22 Å to 14.92 Å (air dried condition) was observed after permeation with  $\text{CaCl}_2$  due to the exchange of  $\text{Na}^+$  by  $\text{Ca}^{2+}$ . Therefore, tests conducted with deionized water as permeating fluid described the behavior of a high-swelling bentonite; whereas tests conducted with  $\text{CaCl}_2$  (1000 mol/m<sup>3</sup>) illustrated the expected behavior for low-swelling bentonites (Kenney et al., 1992).

#### 4. Conclusions

This paper shows that the studied Cretaceous natural Na-bentonite from North Patagonia Argentina can be effectively used as a component of a GCL or a CCL. Ageing effects during storing and the contact with a highly concentrated saline solution ( $\text{CaCl}_2$  1000 mol/m<sup>3</sup>) may affect the hydraulic behavior of this material as a consequence of changes in the microstructure of the bentonite.

A new applicability for bentonites of regional provenance which have a naturally very high amount of sodium was found. This becomes of relevance given that most of the sodium bentonites used in geoenvironmental engineering nowadays are originally calcium bentonites that have been sodium activated.

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