A parametric study on the hydromechanical properties of claystone/bentonite mixtures

Danai Tyri[1,2,3*], *Irini* Djeran-Maigre[2], *Jean-Claude* Robinet[3] and *Jad* Zghondi[4]

[1]EDAFOS Engineering Consultants S.A., 10558 Athens, Greece, formerly: Univ Lyon-INSA Lyon, Euro-Géomat Consulting
[2]Univ Lyon, INSA Lyon, 69621 Villeurbanne, France
[3]Euro-Géomat Consulting, 45100 Orléans, France
[4]Andra, Meuse/Haute-Marne Underground Research Laboratory, 55290 Bure, France

Abstract. Mixtures of crushed excavated COx claystone and bentonite are considered as a potential backfill solution for the French disposal concept of radioactive waste. In the present work, the hydro-mechanical behaviour of claystone/sodium bentonite mixtures was experimentally investigated for various bentonite contents (≤40% in total mass) and densities. Demineralized, site water and cementitious alkaline solution were used. Test on mixtures physicochemical properties showed that Plasticity Index (PI) and Free Swelling Index (FSI) values increase with bentonite content. Introducing bentonite in the mixture induces to lower density for the same compaction energy. No clear impact of the cementitious solution was observed compared to the site water. The expected swelling pressure increase upon bentonite increment was obtained. The effect was more important at higher densities. Mixtures primary wetting, resulted in lower swelling pressure.

1 Introduction

In France, the national radioactive waste management agency (Andra) oversees the Industrial Centre for Geological Disposal (Cigéo), designed to host the French High-Level Waste (HLW) and Intermediate-Level Long-lived Waste (ILW/ILW-LL). The centre will be constructed at the mid-level of Callovo-Oxfordian (COx) claystone formation, at approximately 500m depth. Cigéo safety system relies principally on COx claystone as a natural barrier and includes also structures as passive post-closure safety systems. Upon the closure of geological disposal facilities, all tunnels will be backfilled to limit the propagation of the excavation damaged zone around the drifts and contribute to the sealing maintenance [1]. Different mixtures are considered as a potential backfill solution. In particular, COx/bentonite mixtures are examined due to their well-known swelling properties, or COx/sand based mixture are investigated due to their mechanical behaviour and their high gas permeability which is also another benefit behaviour of the backfill. On that framework, Andra studies different potential backfill mixtures and one of the considered solutions is to reuse crushed excavated COx mixed with bentonite additive, placed at an adequately initial dense state.

After the backfill installation, the material absorbs groundwater, swells and fills inevitable technological voids. In parallel, concrete degradation of the engineering barrier (liners along drifts and ILW cells) produces alteration of groundwater's composition. Hence, the alkaline perturbation on the installed materials constitutes an aspect for examination [2].

The claystone/bentonite backfill will be installed in situ, using available technological means. Andra studies different backfill solutions alone or combined: pellets (i.e., highly compacted granules), prefabricated blocks and/or in-situ compaction.

In the present study, the hydro-mechanical behaviour of claystone/bentonite mixtures is experimentally investigated for various bentonite contents and densities. This analysis served as an initial investigation since it was later incorporated into a pelletized claystone/bentonite mixture. The presented parametric investigation focuses on the effect of the sodium bentonite content, initial dry density and infiltrated solution on basic physicochemical properties and swelling capacity of the studied materials (bentonite content ≤ 40% in total mass).

2 Materials and Methods

2.1 Materials and infiltrated fluids

Mixtures of COx claystone and sodium bentonite in various fractions were studied. COx claystone was excavated from Andra's Underground Research Laboratory (URL) and subsequently crushed. COx presents a rather homogeneous composition (20% tectosilicates, 20-25% carbonates and 50-55% clay minerals) [1]. The studied bentonite is a commercial sodium bentonite from Wyoming (commercial name

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[*] Corresponding author: dtyri@edafos.gr

WH2 Gelclay), containing 80% smectites and other non-clay minerals [3]. Both materials present maximum grain size of 2 mm at the as-received condition. The materials grain size distribution obtained by the dry sieving method is presented on Fig.1. Table 1 summarises the physical and chemical properties of the studied materials. In the present experimental work, the incorporated bentonite content was no higher than 40% (in mass).

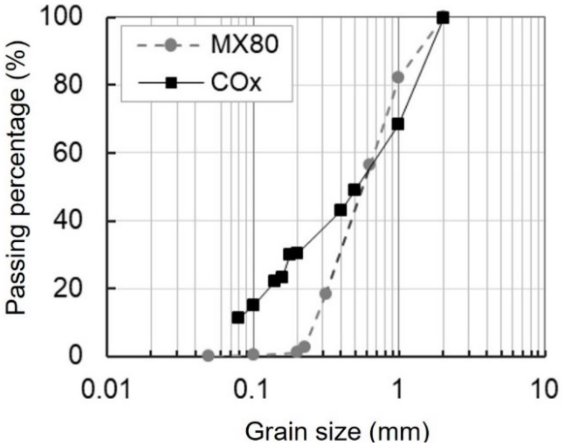

Fig. 1. Grain size distribution of the studied materials.

Table 1. Physical and chemical characteristics of studied materials.

Material	***w*** **(%)**	***LL*** **(%)**	***PL*** **(%)**
Claystone	6.2	47	21
Bentonite	12.5	465	57

w: water content
LL: Liquid Limit obtained from Atterberg limits
PL: Plastic Limit obtained from Atterberg limits

Three infiltrated fluids were used for the materials hydration to follow design conditions. Two synthetic fluids were prepared: site water (SW) having the groundwater's composition and cementitious water (CW), representing the groundwater that has undergone alkaline perturbation. Tests were also conducted with demineralised water (DW) as reference. Table 2 summarises their characteristics. CW presents alkaline conditions. Nonetheless, the ionic strength of the synthetic solutions does not significantly alter.

Table 2. Properties of infiltrated fluids used in the experiments.

Fluid	**pH**	**Ionic strength, *I* (mmol/L)**
Demineralised water DW	6.1	-
Site water SW	7.9	83
Cementitious water CW	12.8	87

2.2 Experimental methods

The physicochemical properties of claystone/bentonite mixtures were studied by following conventional soil testing methods. Atterberg limits, free swelling index (FSI) and dynamic compaction characteristics were determined according to French Standards [4–7]. FSI was determined following the (AFNOR 2002), where two grams of dried powdered clay (d<160μm) are progressively poured into a graduated cylinder, containing 100ml of aqueous solution. The volume of swollen clay in the cylinder is measured after 24h. Atterberg limits and FSI tests were performed by using demineralized, site and cementitious water. The dynamic compaction characteristics were determined by performing Standard Proctor tests, where mixtures were hydrated by spaying the proper amount of water (corresponding to the target water content) on Cox claystone and then adding the necessary bentonite on the hydrated material. The decision focused on avoiding the immediate swelling of bentonite, which would prevent a homogeneous mixing process. The mixtures were stored in hermetic plastic bags at least 12h. Site water and cementitious water were used.

Swelling pressure developed under constant volume was measured using isochoric oedometric cells (D=40 mm). The mixtures were compacted by means of a hydraulic press at the desired initial dry density. The cell was placed on a device consisting of a metallic frame and a force sensor was positioned to measure the developed pressure upon wetting. A large screw was fixed to prevent the axial displacement upon wetting. The screw was reinforced by using a "pre-stressed" spring to minimise as possible the system's deformability. During the experiment, the displacement of the piston was monitored by a displacement transducer. The axial strains remained < 0.4%. Fig. 2 illustrates the experimental system used on swelling pressure tests. Mixtures of claystone/bentonite at the original conditions of reception were studied having bentonite proportions of 10%, 20%, 30% and 40%. The samples were compacted at 1.45Mg/m^3 and 1.60Mg/m^3, corresponding at the investigated density range [8]. In addition, the effect of water content on the swelling pressure was studied by carrying out a supplementary series of tests on specimens prepared at water contents corresponding to the optimum compaction conditions. Details regarding the experimental program presented on Table 3.

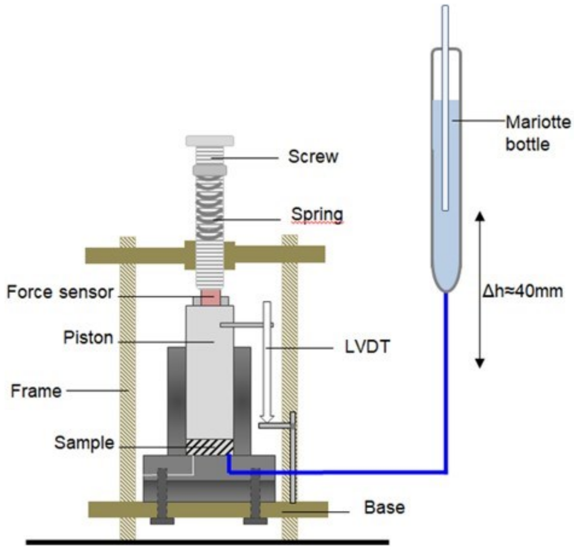

Fig. 2. Experimental system used on swelling pressure tests.

Table 3. Experimental program of the conducted swelling pressure tests.

No.	Bentonite content	Initial dry density, ρ_d (Mg/m3)	Initial water content, w (%)	Initial Saturation degree, S_r (%)
1	10	1.46	6.7	21.4
2	20	1.45	7.5	23.4
3	30	1.45	8.5	26.4
4	40	1.46	7.9	24.9
5	10	1.61	6.7	27.1
6	20	1.60	7.3	28.9
7	30	1.60	8.1	31.2
8	40	1.61	8.6	33.6
9	10	1.59	17.2	67.1
10	20	1.60	21.1	83.1
11	30	1.62	24.0	96.4
12	40	1.59	24.8	96.4

3 Experimental Results

3.1 Physicochemical properties

The evolution of mixtures plasticity wetted with demineralised, site and cementitious water is presented on Fig.3 for the tested claystone/bentonite samples. A clear increase of the Plasticity Index (PI) is observed with bentonite fraction increase regardless the hydration water. Decrease on plasticity can be noticed when synthetic solutions are used. The impact is better defined at high bentonite fractions. Even though a slightly higher PI values are observed when CW is used compared to SW, no clear behavior can be obtained.

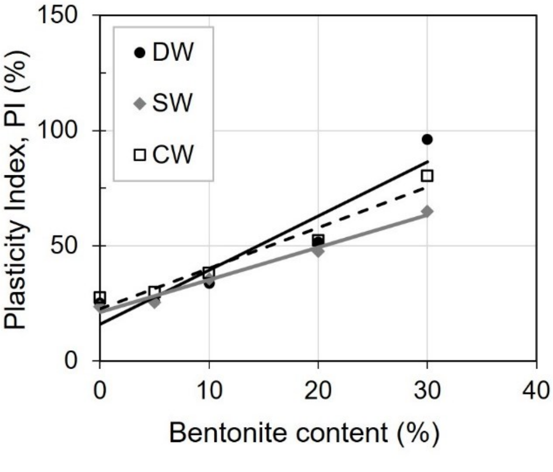

Fig. 3. Plasticity index of claystone/bentonite mixtures in relation to bentonite content.

The FSI measurements illustrated on Fig.4 show that FSI increases with bentonite content for all types of fluids. Nevertheless, FSI remains relatively low (FSI<6cm^3/2g). Similarly, to the Atterberg limits, mixtures swelling appeared to be relatively lower with synthetic solutions. The impact relates to synthetic solutions salinity, compared to the clear demineralized water. A comparison between site and cementitious water shows that the alkaline solution resulted in decrease of swelling capacity that site water, especially for bentonite content < 20%).

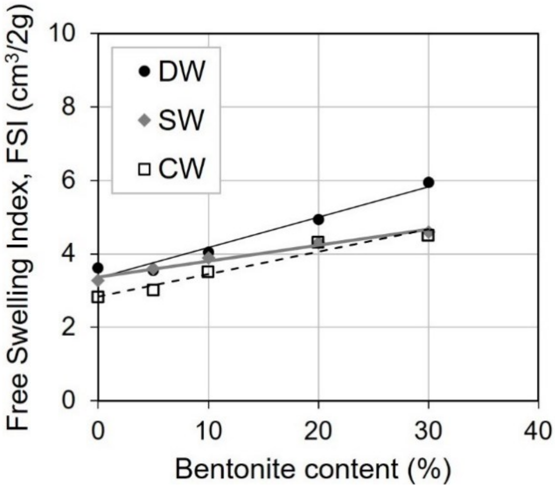

Fig. 4. Results of Free Swelling Index of claystone/bentonite mixtures in relation to bentonite content.

Results of dynamic compaction tests are presented on Fig.5 for different claystone/bentonite mixtures prepared with site and cementitious water. A clear behavior in material's compaction is observed for both fluids: the higher the presence of bentonite in the mixture, the lower the maximum dry density is achieved (Fig.5a), while the optimum water content appears to be shifted on higher values (Fig.5b). Observations are in accordance with past researchers [9–11]. This tendency is caused due to bentonite's swelling upon hydration. Swelling of bentonite particles means that more volume is occupied by bentonite in the sample and lower compaction can be obtained for the same compaction energy. Therefore, dynamic compaction properties serve as an additional "indirect" method to evaluate the mixtures swelling capacity. Samples prepared with site water could be compacted at higher dry densities than cementitious water, implying that the mixtures wetted with the alkaline solution had swollen more than those wetted with site water.

3.2 Swelling pressure

Swelling pressure developed by the tested claystone/bentonite mixtures is illustrated on Fig.6 in relation to bentonite content and dry density. A non-linear increase of final swelling pressure with bentonite percentage is observed, with more notable effect at higher dry densities. As expected, swelling pressure is improved upon density increase, which is well-reported on pure bentonites and on swelling-clay based materials [12–15].

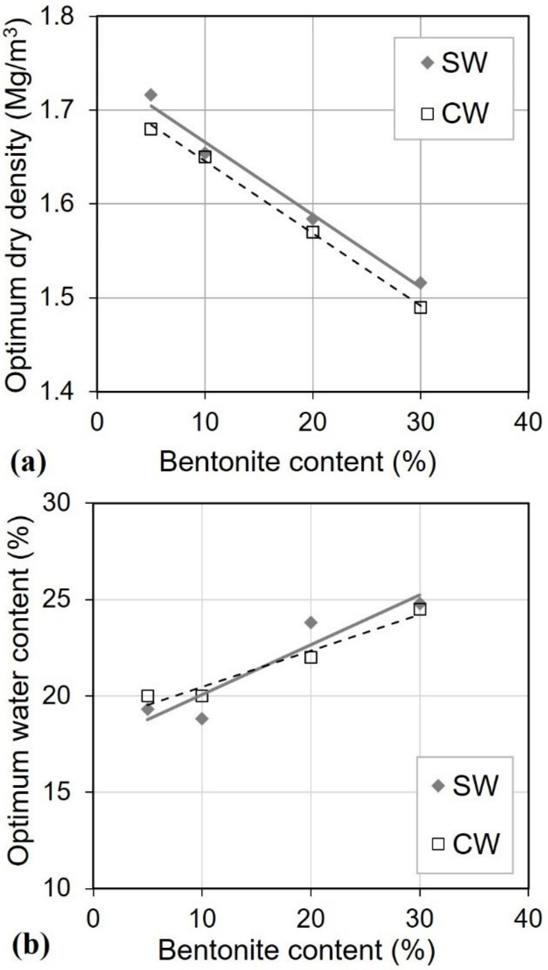

Fig. 5. Results of Proctor test of claystone/bentonite mixtures in relation to bentonite content: (a) optimum dry density and (b) optimum water content.

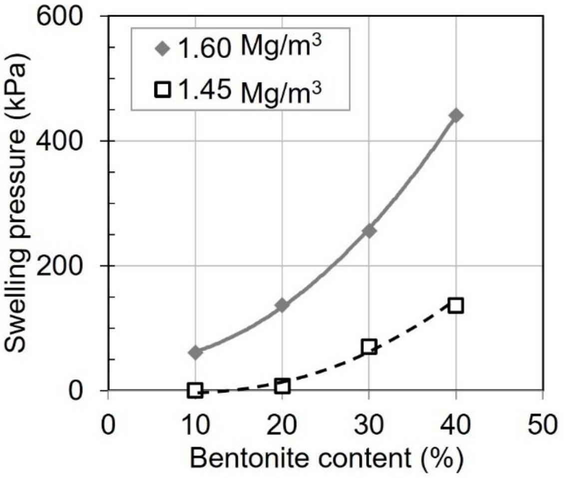

Fig. 6. Swelling pressure of claystone/bentonite mixtures compacted at different initial dry densities.

The supplementary series of swelling pressure tests on mixtures prepared at the optimum water content shows the influence of initial water content conditions on the final swelling pressure. The obtained results are presented on Fig.7 for the mixtures at dry density of 1.60Mg/m^3. Lower swelling is observed upon bentonite addition, while on low bentonite contents (<20%), the impact becomes insignificant. The results show the sensibility of bentonite's hydro-mechanical behavior on interaction with water. When the material is pre-wetted, swelling capacity decreases.

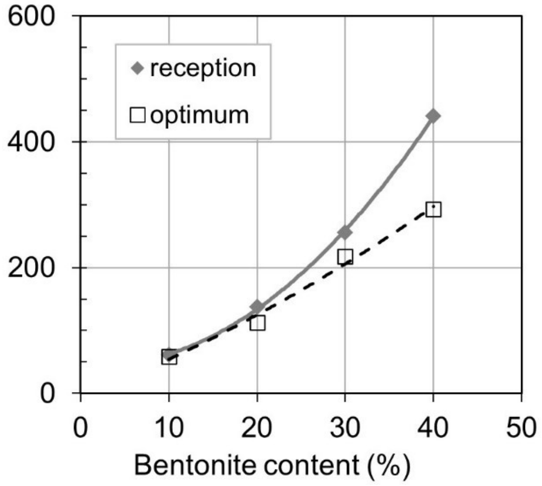

Fig. 7. Swelling pressure of claystone/bentonite mixtures compacted at initial dry density $\boldsymbol{\rho_d}$ = 1.60Mg/m^3, at water content of reception and optimum water content conditions.

4 Conclusions

The present experimental study focused on the hydro-mechanical properties of claystone/bentonite mixtures, (Bentonite content ≤40% in total mass). The experimental analysis constituted an initial investigation, for the subsequent examination of a pelletized claystone/bentonite mixture.

Crushed excavated COx clayestone mixed with commercial sodium bentonite was studied, evaluating the mixtures plasticity, free swelling capacity, and compaction characteristics. The swelling pressure of compacted samples with 1.45Mg/m^3 and 1.60Mg/m^3 of dry density were also tested. Demineralized and synthetic solutions (site water pH ≈7.8, cementitious water pH ≈12.8) were used.

Increase of PI and FSI upon bentonite addition was obtained for all solutions used. Site and cementitious solutions presented relatively lower values, compared to demineralized water. The chemical impact was more evident at higher bentonite contents, due to higher smectite presence which exhibits sensibility to water chemistry. Proctor tests showed decrease of optimum dry density with bentonite addition when the mixtures are compacted at the optimum water content. The effect was caused by bentonite's swelling upon hydration which hindered compaction. No significant impact of the cementitious solution is observed compared to the groundwater (site water). In general, the two synthetic solutions present similar ionic strength and therefore the effect is considered insignificant.

Swelling pressure follows a non-linear increase with the imposed bentonite content, while the effect is more notable for higher dry densities. For bentonite contents higher than 20%, mixtures with higher initial water content exhibited lower final swelling pressure.

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