Hydraulic conductivity and small-strain stiffness of a cement-bentonite sample exposed to sulphates

Conductivité hydraulique et module de cisaillement initial d'un échantillon de ciment-bentonite exposé aux sulfates

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ABSTRACT: Cement-bentonite slurries have often been used for geoenvironmental applications to isolate contaminated areas. A potential issue with all cement-treated materials is their durability, especially when applied in chemically aggressive environments. In this paper, the small-strain shear modulus (G_0) and the hydraulic conductivity (k) of a cement-bentonite sample in contact with water and an aggressive sodium sulphate solution were investigated. Bender elements were installed in a flexible-wall hydraulic conductivity cell, to simultaneously monitor both G_0 and k. As expected, permeation with clean water had no significant effect on the cement hydration, e.g. G_0 continued to increase and k decreased gradually with time. However, after prolonged permeation with sulphates, a decrease of G_0 and a gradual increase of k were recorded. These observations suggest that contact with sulphates produces degradation of the cemented structure that results in loss of strength and development of a network of interconnected fissures within the sample that increases the hydraulic conductivity.

RÉSUMÉ : Des barrières de ciment-bentonite ont été souvent utilisées pour des applications géo-environnementales afin d'isoler les zones contaminées. Un problème potentiel avec tous les matériaux cimentés est leur durabilité, en particulier lorsqu'ils sont mis en place dans des environnements chimiquement agressifs. Dans cet article, le module de cisaillement initial (G0) et la conductivité hydraulique (k) d'un échantillon de ciment-bentonite en contact avec de l'eau et une solution agressive de sulfate de sodium ont été étudiés. Des bender elements ont été installés dans une cellule de conductivité hydraulique avec une paroi flexible, afin de surveiller simultanément G0 et k. Comme prévu, l'infiltration à l'eau claire n'a eu aucun effet significatif sur l'hydratation du ciment, par exemple, G0 a continué d'augmenter et k a diminué progressivement avec le temps. Cependant, après l'infiltration avec les sulfates, soit une diminution de G0 et une augmentation progressive de k ont été enregistrées. Ces observations suggèrent que le contact avec les sulfates produit la dégradation de la structure cimentée qui entraîne une perte de la résistance au cisaillement et la formation d'un réseau de fissures interconnectées dans l'échantillon qui augmente la conductivité hydraulique.

KEYWORDS: clay, cement, sulphate attack, hydraulic conductivity, small-strain shear modulus.

1 INTRODUCTION

Low permeability vertical barriers (cut-off walls) are often used to control groundwater flow and to isolate polluted soil. They are constructed by excavating a vertical trench. During excavation, the trench is filled with a slurry to prevent collapse. When the slurry is a mix of cement, bentonite clay and water, the barrier is denominated cement-bentonite (CB) cut-off wall (Jefferis 1981). The design of a CB cut-off wall is based on the characterization of the hydraulic conductivity, the strength of the cement-clay mix and eventually chemical compatibility with local groundwater.

Traditionally, the mechanical properties and hydraulic properties of the cement-clay mix are studied separately on different specimens (e.g. Opdyke and Evans 2005). Mechanical properties are usually evaluated by unconfined compression testing; however, the amount of data obtained is often limited to a few curing times and is usually subjected to scatter. On the other hand, hydraulic properties are evaluated by hydraulic conductivity tests; however, it is difficult to relate the hydraulic conductivity data alone to variations of strength, stiffness or microstructure of the cement-clay mix.

In this paper, an advanced testing method was used to simultaneously monitor both mechanical and hydraulic properties of a single sample. To that aim, a flexible-wall hydraulic conductivity cell was combined with a non-destructive technique to monitor the hardening of cement-clay samples. This technique uses bender elements (Dyvik and Madshus 1985) to measure the small-strain shear modulus, G_0.

Such stiffness modulus is typically associated with small shear-strain levels (lower than 10^{-3} %). In general, G_0 is governed by a number of factors such as stress history, stress level, void ratio, soil fabric, and the stiffness of the porous medium skeleton (Santamarina et al. 2001). Then, an increase of G_0 can be expected with increasing interparticle cementation due to cement ageing. Conversely, a decrease of G_0 can be expected when interparticle cementation is disrupted due to either mechanical or chemical degradation.

Experimental work was carried out on bentonite clay mixed with blast furnace slag cement. Monitoring of the small-strain shear modulus of cement-treated clay proved to provide valuable additional information to study the degradation of these materials.

2 MATERIALS AND SAMPLE PREPARATION

The samples studied in this research consist of a mixture of clay, cement and water. A sodium-activated bentonite clay, blast furnace slag cement of the type CEM III/B (ENV 197-1) with a nominal strength of 42.5 MPa and purified water with an electrical conductivity EC ≤ 2 μS/cm and a pH of about 7.6 were used. During hydraulic conductivity testing, the CB sample was initially permeated with purified water for 1 month to allow for further hydration of the cement products. After that period, the sample was permeated with a 25 g/L solution of Na_2SO_4. Such high Na_2SO_4 concentration was chosen here to accelerate the degradation process; however, it may be too high to represent common sulphate exposure levels in the field.

Some properties of the clay and the chemical composition of the clay and cement used here are summarized in Table 1 and Table 2, respectively.

Table 1. Physical properties of the bentonite clay

Parameter	*Value*
Liquid limit	541.9
Plastic limit	67.0
Plasticity index	474.9
Swell index (ml/2g)	34
Cation exchange capacity (meq/100g)	73

Table 2. Chemical composition of the blast furnace slag cement & clay

Main component	*Cement*	*Bentonite*
SiO_2 (%)	29.3	53.7
Al_2O_3 (%)	8.8	23.4
Fe_2O_3 (%)	1.2	5.9
MgO (%)	6.7	2.4
CaO (%)	47.1	1.9
Na_2O (%)	0.2	2.2

The prepared samples consist of 80% water, 16% cement and 4% bentonite (by weight). This composition is in agreement with other studies in the literature (e.g. Ryan and Day 1986, Jefferis 1992, Opdyke and Evans 1995). First, a slurry of bentonite and water was mixed with a high-speed shear mixer for 5 minutes. The slurry was poured in a closed container and allowed to hydrate for about 24 hours. Subsequently, a slurry cement and water (accounted for in the final composition) was prepared to obtain a water cement ratio of about 0.5. Finally, the cement slurry and the 24-hour hydrated bentonite slurry were mixed in a dough mixer for about 10 minutes.

Then the fresh CB slurry was poured in stainless-steel moulds to prepare cylindrical specimens. The moulds were lightly vibrated to ensure that any trapped air bubbles were removed. The bottom and top ends of the mould were sealed with plastic foil to prevent moisture loss. Then, the samples were allowed to cure for 7 days in a conditioned room at 18°C. After that period, when the samples showed enough strength to be handled, their ends were flattened with a spatula and they were carefully extruded out of the moulds. Samples with a diameter of 100 mm and a height of 60 mm were used for hydraulic conductivity tests.

3 METHODS

The hydraulic conductivity and small-strain shear modulus of a CB sample were studied in a flexible-wall hydraulic conductivity cell provided with bender elements. In parallel, the hardening of the CB mix in contact with water was monitored through bender element testing in a bench top setup to provide a reference of G_0 increase under normal curing conditions.

3.1 *Bench top bender element test*

The small-strain shear modulus of the CB mix was evaluated (starting from a fresh state) by bender element testing (Shirley and Hampton 1978, Dyvik and Madshus 1985) in a bench top setup (Fig. 1). The bender elements used here are of the type T220-A4–203Y (Piezo Systems, Inc.). The effective bender element length penetrating in the sample was about 4.5 mm.

The bench top bender element setup consists of two translucent polymethyl methacrylate (PMMA) plates that hold a U-shaped rubber mould with an open space for housing a cemented sample. The bender element transmitter and receiver are fixed to the PMMA plates, one in front of the other and vertically aligned. All parts are held together by four sets of screws and nuts resting on rubber disks to avoid wave propagation through the apparatus itself. More details on this setup are given by Verastegui-Flores et al. (2010).

Testing was started immediately after a fresh CB mix was prepared. The mix was poured into the rubber mould and it was allowed to cure in a conditioned room at constant temperature (18°C). In order to avoid desiccation, the sample was kept all the time under a thin layer of purified water. Bender element measurements were performed on a regular basis from the first day of curing up to 2 months approximately.

In bender element testing, G_0 is determined out of the propagation velocity (V_s) of shear waves generated and detected by the transmitter and receiver bender elements installed on opposite sides of a sample. G_0 is estimated as:

$$G_0 = \rho V_s^2 \quad (1)$$

where ρ is the density of the sample. V_s is evaluated as follows:

$$V_s = L / t_s \quad (2)$$

where L is the tip-to-tip distance between the transmitter and receiver bender elements, and t_s is the travel time of the shear waves from the transmitter to the receiver. t_s is evaluated out of the signal recordings. In this research, t_s was evaluated by means of two methods. The first one consists of visually identifying the first direct arrival from the output signal (e.g. Dyvik and Madshus 1985, Jovičić et al. 1996; Viggiani and Atkinson 1995). Clearly, the success of this method depends on the quality of the recorded signal. The second method used here was the cross-correlation method, first introduced by Viggiani and Atkinson (1995). The cross-correlation analysis measures the level of correspondence or interrelationship between two signals of similar nature and it produces the time shift between them, which is equivalent to the travel time of the shear wave. Although some authors argue that the cross-correlation method may not be suitable for bender element interpretation (e.g. Arulnathan et al. 1998), it produced very consistent results. Both methods produced a similar outcome.

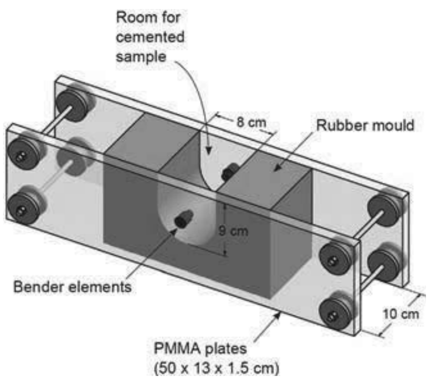

Figure 1. Bench top bender element setup

3.2 Flexible-wall permeability cell with bender elements

A flexible-wall permeability cell was provided with bender elements (one in the base pedestal and the other on the top cap) to enable the simultaneous monitoring of G_0 and k of a cement bentonite sample. Moreover, height changes during permeation could also be monitored through a cathetometer placed next to the cell (Fig. 2).

The parameter k was evaluated out of a falling-head test performed in a conditioned room at constant temperature (18°C) and at an isotropic effective stress of 30 kPa. The sample was first permeated with deionized water for about 1 month (1.7 pore volumes of flow). Next, the deionized water was replaced with the 25 g/L solution of Na_2SO_4 and the test was continued for a period of about 250 days (about 10 pore volumes of flow).

Figure 2. Flexible-wall hydraulic conductivity cell provided with bender elements and a cathetometer

4 RESULTS

4.1 Small-strain shear modulus

Figure 3 summarizes all G_0 measurements carried out in the benchtop bender element setup and in the modified flexible-wall hydraulic conductivity cell. As expected, the results of the benchtop bender element setup, where the CB sample was cured in pure water, showed a gradual increase of G_0 in time due to normal cement hydration. Verastegui-Flores et al. (2010) showed that the G_0 increasing trend of clay treated with blast furnace cement could be fairly-well characterized through a logarithmic function. Clearly, all measurements up to a sample age of 90 days were in excellent agreement with such rule.

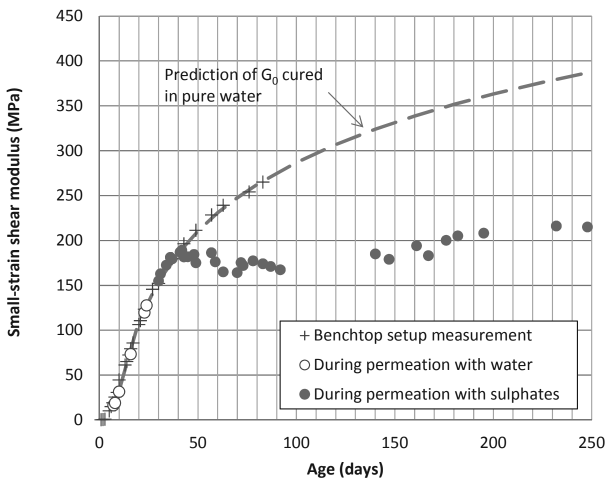

Figure 3. Impact of sulphate attack on the small-strain shear modulus of a cement-bentonite sample

Similary, G_0 measurements in the flexible-wall permeability cell are in agreement with measurements out of the benchtop bender element setup during the first phase of the tests, when the sample was permeated with pure water for about one month. However, when the permeation with the 25 g/L Na_2SO_4 solution started, the normal cement hydration process was clearly disrupted (Fig. 3). G_0 measurements up to 250 days of permeation, when compared to the expected G_0 trend in contact with pure water, show that the stiffness of the CB sample was significantly reduced due to contact with sulphates. Such reduction is the result of interparticle cementation degradation and it could also indicate severe fissuring affecting the original structure of the CB sample. Clearly, a decrease in G_0 suggests a decrease of strength as well as both parameters are strongly linked to interparticle cementation.

Deterioration of the cement hydration products by sulphates is a well-known durability problem in cement mortars exposed to high concentrations of sulphate ions. The most common manifestations of sulphate attack in concrete are expansion, caused by formation of ettringite and gypsum within the matrix of a specimen, and loss of strength. A similar phenomenon was observed in CB samples.

4.2 Hydraulic conductivity

Figure 4 summarizes all k measurements as well as sample height changes during permeation with water (for one month) followed by permeation with a 25 g/L Na_2SO_4 solution for a total period of about 250 days.

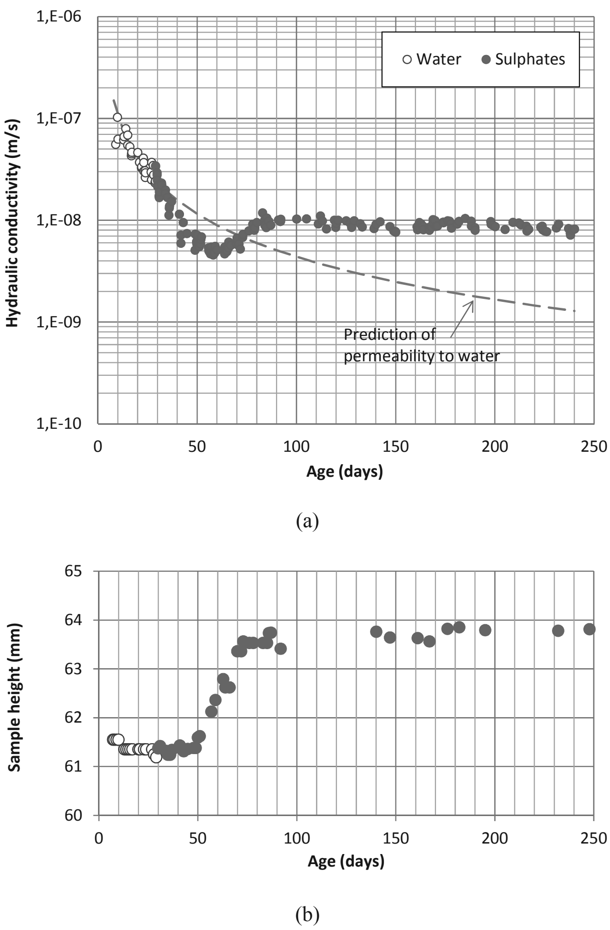

Figure 4. Impact of sulphate attack on the (a) hydraulic conductivity and (b) the height of a cement-bentonite sample during permeation with water and a 25 g/L Na_2SO_4 solution in a flexible wall cell.

As expected during the first phase of permeation with pure water, a gradual decrease of k with increasing time was observed. This feature of cement-bentonite samples has been reported before in the literature (e.g. Fratalocchi et al. 1998, ICE 1999) and an equation describing such trend has also been proposed:

$$k(t) = k_{28d} \, (t / 28)^{-n} \quad (3)$$

where k_{28d} is the hydraulic conductivity at an age of 28 days, t is the age of the sample in days and n is a constant. Based on existing data and Eq. 3, it was possible to predict the permeability to water of the CB sample vs. time (Fig. 4a).

In the second phase of the test, the CB sample was permeated with a Na_2SO_4 solution. As a result of deterioration of the sample an immediate increase of *k* was expected. However, in the early phase of sulphate attack (age between 30 to 70 days) the permeability to sulphates seems to be lower than the expected permeability to water. This feature may have been caused by formation of gypsum in the pores within the sample (Santhanam et al. 2003) which may result in a gradual clogging of the pores. Gypsum primarily deposits in the fissures and in voids, because these provide the best sites for nucleation. After a while, when the formation of ettringite becomes significant, the affected areas tend to expand, then, fissures start to appear which will gradually lead to an increased hydraulic conductivity. In fact, figure 4 shows that expansion of the sample and increase of hydraulic conductivity start at approximately the same time.

The hydraulic conductivity of a cement-clay mix is not a simple function of the porosity, but depends also on the size, distribution, shape, tortuosity and continuity of the pores that change during the cement hydration and sulphate attack.

5 CONCLUSIONS

Traditionally, the mechanical and hydraulic behaviour of cement bentonite samples are studied separately on different specimens. In this research, a flexible-wall hydraulic conductivity cell was provided with bender elements to measure the hydraulic conductivity as well as the small-strain shear modulus.

Monitoring of G_0 was shown to provide valuable quantitative information to study the deterioration effects of sulphate attack on a cement-clay mix. As expected, during permeation with deionized water an increase of G_0 and a decrease of permeability were observed due to normal cement hydration. On the other hand, after sustained contact with sulphates a significant decrease of G_0 and increase of k were measured. These observations suggest that the sulphate attack produces severe degradation of interparticle cementation and perhaps also severe fissuring affecting the macrostructure of the CB sample which in turn may lead to a loss of strength.

6 ACKNOWLEDGEMENTS

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