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Durability assessment of a confinement cut-off wall for a phosphogypsum landfill

Durabilité des parois d'isolation d'une décharge de phosphogypse

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ABSTRACT
An experimental investigation on the interaction between a contaminant and cement-bentonite mixtures is described and discussed. The best mixture had to be selected to construct a self-hardening cut-off wall to isolate a hazardous waste landfill. The contaminant (i.e. the landfill leachate) is a very acidic inorganic solution which could adversely affect the hydraulic performance of cement mixtures. Immersion and permeation tests were performed to investigate the mixture performance under very aggressive environmental conditions in order to accelerate the effects of chemical reactions. The results of the tests are discussed, showing that the mixture-leachate interaction does not result in a worsen hydraulic performance. An assessment of the durability of the mixture is given.

RÉSUMÉ
Cet article analyse le compatibilité et la durabilité d'une parois moulée en mélange ciment-bentonite (CB) pour l'isolation d'une décharge de phosphogypse. Le lixiviat est très acide et il peut influencer négativement la perméabilité des mélanges CB. Des essaies d'immersion et de perméation ont été réalisées afin d'investiguer les performances hydrauliques en présence de conditions agressives et de accélérer l'effet des interactions chimiques. On montre que l'interaction CB-lixiviat ne comporte toujours une augmentation de perméabilité. On cherche d'estimer aussi la durabilité du matériel.

1 INTRODUCTION

Phosphogypsum (calcium sulphate) is a solid by-product of phosphoric acid production. Each ton of phosphoric acid is accompanied by the production of 4.5-5.5 tons of phosphogypsum. It is estimated that more than 22 million tonnes of phosphoric acid (as P_2O_5) are produced annually worldwide, generating an excess of 110 million tonnes of phosphogypsum by-product; more than 95% of it still has to be disposed.

Phosphogypsum is a damp, powdery material; the permeability for unstabilized phosphogypsum ranges from 10^{-4} to 10^{-5} cm/s (Lopez and Seals, 1992; US EPA web site). As its major constituent is calcium sulphate, it exhibits acidic properties (the free water in the gypsum cake is highly acidic, having a pH as low as 1.0); therefore, the leachate of a phosphogypsum landfill can be very aggressive for the bottom and sidewall liners.

In particular, when a remediation system has to be designed to prevent the leachate migration, circumferential diaphragms made of cementitious mixtures are the typical solution considering the most proved technologies currently available (self-hardening slurry walls, jet-grouting, deep-mixing). In such cases the main problem is to assess the wall durability since it is well known that acids can cause disintegration of an alkaline material like a cementitious mixture; even if disintegration does not occur, acids can adversely affect the hydraulic behaviour of the cement-bentonite mixtures; indeed, acids can alter the cement hydration processes which are responsible for typical reduction in the hydraulic conductivity with time of the cement-bentonite mixtures.

In this paper an experimental investigation is described and discussed concerning the interaction of typical cement-bentonite mixtures to be used for a cut-off wall to isolate a phosphogypsum landfill and the very acidic leachate of the landfill.

2 SITE DESCRIPTION AND REMEDIATION DESIGN

The phosphogypsum landfill, located in Sicily (Italy), is about 55 hectares wide with a maximum thickness of 10 m; a total volume of about 4.000.000 m^3 of phosphogypsum has been disposed, starting from early 80's until 1992.

The waste body is surrounded by compacted clay embankments and lies on a clayey-silty low permeable soil ($k \leq 10^{-10}$ m/s) with sandy lenses and thin sandy strata which were found to be responsible for potential leachate migration. The groundwater table is strongly dependent on the meteoric events; its level can change of several meters below the ground surface.

The landfill has been always without any cover so that the rain leaching through the waste body must be pumped and treated continuously.

The proposed remediation design (by Aquater S.p.A. Snam-Progetti, Eni Group) mainly consists of: a cover system including a HDPE geomembrane coupled with a geosynthetic clay liner; a circumferential composite cut-off wall (Figure 1), made by a self-hardening cement-bentonite (CB) mixture and a HDPE geomembrane (the cut-off wall is embedded into the clayey substratum); an internal permeable diaphragm all around the waste body and pumping wells, to remove the leachate.

Performance trials of the proposed CB mixture were required to verify its general properties in the short and long term, to control its resistance to the aggressive leachate and to assess its durability. To this purpose a special experimental program was properly designed to investigate the mixture performance under the aggressive environmental conditions of the site. In particular, test conditions simulating those expected in the field are fundamental and special tests have to be carried on for sufficient time to show the full effects of physical or chemical reaction (Jefferis, 1992; Pasqualini and Fratalocchi, 2000).

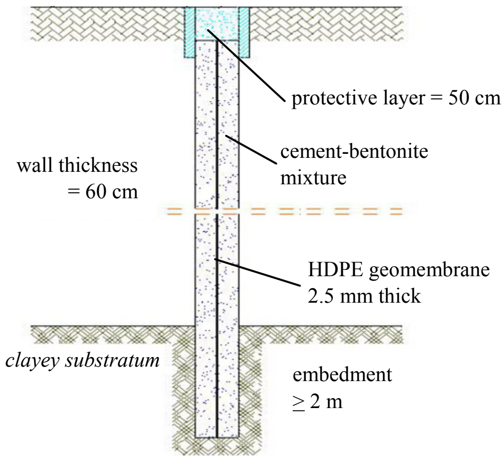

Figure 1. Cross section of the cut-off wall.

3 MATERIALS

After preliminary tests on different CB mixtures, two of them were investigated in detail to select the best one in terms of workability, permeability and chemical compatibility; then its long term performance was analysed, a test panel was constructed close to the phosphogypsum landfill with this mixture and samples of fresh and aged mixture were taken for lab tests.

The two mixtures consist of tap water, sodium-activated bentonite, blast furnace slag cement (type III/A or III/B) and an additive to improve their workability at fluid state, in order to facilitate installation of geomembrane panels. Table 1 shows the composition and dosages of the mixtures as well as their main characteristics at the fluid state.

The mixtures were prepared in the laboratory according to the procedures recommended by ETC8 (1993): bentonite powder and water were thoroughly mixed using a high shear mixer and the bentonite mud was allowed about 24 hours to hydrate prior to its use in the slurry mix. Cement was then added to the bentonite suspension while mixed. The additive was added to the bentonite mud before cement addition, according to the sequence of mixing used in the field. Samples were prepared by pouring the fresh mixture into cylindrical plastic moulds of 7 cm in diameter and 10 cm high. Soon after casting, the samples were stored under water or in leachate.

Immersion and permeation tests were purposely performed to simulate very severe chemical conditions for the diaphragm, in order to accelerate the effects of chemical reactions. The chemical composition of the leachate is shown in Table 2.

4 TESTING AND RESULTS

4.1 *Immersion test*

Immersion tests are considered severe tests mainly because of the lack of effective confining pressure and of the large surface exposed to aggression in relation to the volume of the sample. Therefore they give rapid indications of the barrier material durability.

Samples of the mixture were immersed soon after casting in the leachate; for 3-4 days only their upper surface was exposed to leachate; then the samples were extracted from the moulds and the whole surface was put in contact with the leachate. The leachate was changed almost each day in order to avoid variation in the chemical composition and in the pH value, due to reactions with the cement-bentonite.

Pocket penetrometer tests were performed during the first two weeks of curing on the surface always in contact with leachate in order to control possible variation in the mechanical performance. Figure 2 shows that the samples cured under water and under leachate have the same resistance; this means that the contact with leachate does not appreciably alter the cement hydration processes of the selected mixtures at brief curing.

No visible damage and cracking were observed on the samples immerged in the leachate up to about 5 months of curing. Only a light variation in the colour occurred together with the growth of crystals as reaction products on the samples surface. Some very small fissures appeared after about 3 months of immersion in the leachate, however this did not produce any worsening of the performance, as shown by the results of compression and permeation tests discussed in the following. The crystals were recognised (by SEM micro-analyses) to be mainly $MgHPO_4$ and $CaHPO_4$ (Fratalocchi et al., 2004). Their growth and probably the sulphate ions bound in the mixture structure (Kledyński, 2002) produced an increase in weight of the samples up to about 4 months of curing, as shown in Figure 3. After this period, the weight did not change significantly.

Unconfined compression tests were performed on the samples stored under water and under leachate and the results were compared, curing time being equal. No significant differences in the compression strength and stiffness occurred during the observed curing time (Tables 3 and 4).

On the basis of all the results, both the mixtures were considered to have a good performance in the aggressive chemical conditions of the landfill. Anyhow, the long term performance and durability of the mixtures must necessarily be based on the results of permeation tests, which put continuously the aggressive liquid in contact with all the sample volume.

4.2 *Permeability tests*

Permeation tests give a direct indication of variations in the hydraulic performance due to interaction of the barrier material with an aggressive liquid. The main disadvantage consists in the long time (months to years) often necessary to measure the permeability post-reaction. For this reason the permeation tests with leachate were performed with a high hydraulic gradient (i = 200) in order to verify any reaction of the mixtures under a great number of pore volumes of flow.

Table 1. Main data of the mixtures.

Mixture	R	B
Water (dose by weight)	1000	1000
Bentonite (dose by weight)	50	60
Cement (type, dose by weight)	III/A, 230	III/B, 240
Additive (g/l of water)	5	5
Density (g/cm^3)	1.17	1.18
Marsh viscosity (s)	35	39
Bleeding (%)	2	1

Table 2. Main data of the leachate.

pH	2.0
Electrical Conductivity (mS/cm)	22.2
SO_4^{--} (ppm)	3965
HPO_4^{--} (ppm)	23867
F- (ppm)	619
Cl- (ppm)	4088
Fe (ppm)	22
Zn (ppm)	18
Al (ppm)	27

Table 3. Mixture B: results from unconfined compression tests performed on samples cured under water and in leachate (compression strength, σ_f and axial deformation at failure, $\varepsilon_{a,f}$).

Curing time	Water		Leachate	
(days)	σ_f (kPa)	$\varepsilon_{a,f}$ (%)	σ_f (kPa)	$\varepsilon_{a,f}$ (%)
14	681	1.3	672	1.6
30	1036	1.4	1001	1.3
60	1247	1.5	1185	1.6
135	1269	1.4	1257	1.3

Table 4. Mixture R: results from unconfined compression tests performed on samples cured under water and in leachate (compression strength, σ_f and axial deformation at failure, $\varepsilon_{a,f}$).

Curing time	Water		Leachate	
(days)	σ_f (kPa)	$\varepsilon_{a,f}$ (%)	σ_f (kPa)	$\varepsilon_{a,f}$ (%)
14	609	2.1	836	1.1
30	1042	1.4	893	1.6
70	1293	2.9	1354	1.9
120	1444	2.3	1472	1.6

Samples of both the mixtures were continuously permeated with the leachate over a period of about six months; then only the R mixture was kept under permeation because it was selected for the cut-off wall (the test is still going on, after more than 18 months of continuous permeation). Samples of the same mixtures were permeated with tap water, as reference tests.

The permeability values measured over time are shown in Figures 4 and 5. The permeability with water showed the typical reduction with time (Fratalocchi and Pasqualini, 1998). During the first two months of curing, permeation with the leachate resulted in a more rapid reduction in the permeability than the permeation with water, probably due to clogging. Thereafter a light increase in the permeability was observed on both mixtures, followed by a reduction with time similar to that observed in water.

According to Jefferis (2003), the reduction in permeability followed by an increase is typical of cement-bentonite mixtures when subjected to aggressive reaction. The rapid reduction in the permeability observed during the first two months of permeation can be due to the growth of crystals and to precipitation of iron and zinc as hydroxides, often of fine particle size.

The overall increase in the permeability with leachate measured during the third month of curing could be due to predominant reactions of sulphate attack to calcium hydroxide leading to the formation of gypsum and then thaumasite and ettringite; however, this increase in the k values cannot be considered as the definitive effect of the leachate-mixture interaction because the sulphate and phosphate concentration at the effluent was found to be lesser than the inlet one also after the increase of k; moreover, the pH was almost constant in the range from 7 to 7.5 during the whole test. Therefore, a combination of both effects of clogging and advance in the reaction front could explain the increase and the subsequent decrease in the overall permeability. Further investigations and analyses are currently being performed.

From a practical point of view the results from permeability tests show a good performance, considering the severe conditions in terms of chemical concentrations (constant at the inlet) and pore volumes of flow induced through the sample.

Supposing the reaction effects to be only dependent on the numbers of pore volumes of flow of leachate through the mixture, the cut-off durability can be roughly assessed; in particular, considering that a good hydraulic performance was observed on the mixture after 550 days of continuous permeation with the leachate (Figure 5) under a hydraulic gradient equal to 200, in case of a hydraulic gradient across the in situ barrier equal, on average, to 2, the barrier should be efficient one hundred times longer than the length of the test, that is, about 150 years.

It is necessary to point out that the chemical conditions adopted in the tests are very severe, as they are not expected to occur continuously in the field; indeed, the leachate used in all the tests was the worst one measured in the landfill, in terms of concentration levels and pH; moreover, a pumping system is provided into the landfill to reduce the leachate level inside the encapsulation, so that there should be an advective flow of groundwater and not of leachate across the barrier.

Samples taken from the test panel close to the waste body were stored under leachate occasionally refreshed. One of these samples was permeated with leachate after about 7 months of curing and its k values resulted of the order of 10^{-9} cm/s (Figure 6), very close to the values measured on the sample of the same mixture prepared in the laboratory and permeated with leachate, thus confirming a good performance of the in situ R mixture in the long term.

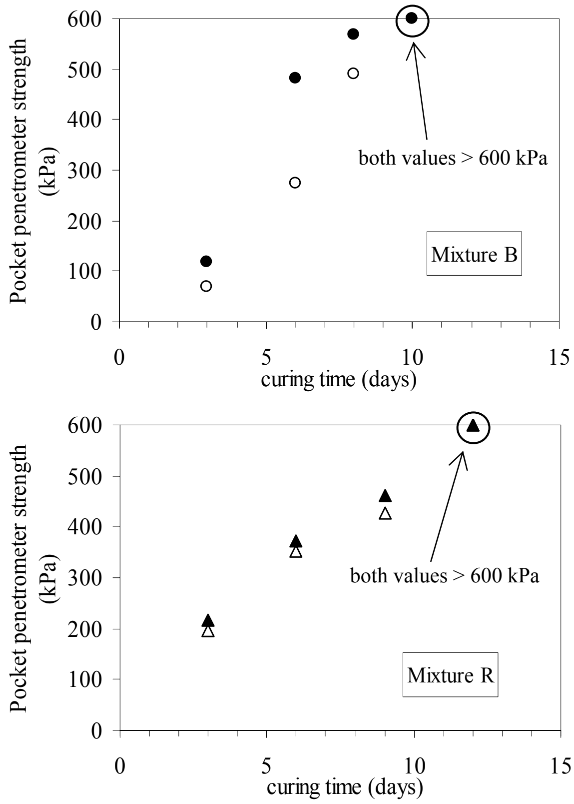

Figure 2. Results from pocket penetrometer tests on samples cured under water (open symbols) and under leachate (solid symbols).

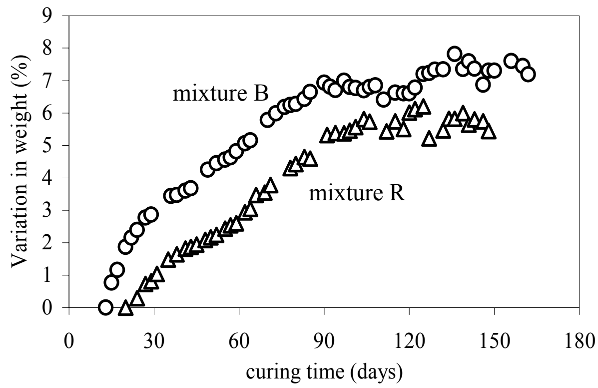

Figure 3. Weight increment of the samples cured under leachate.

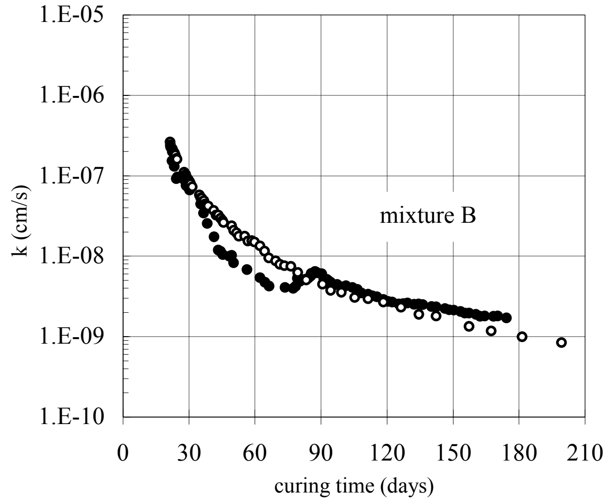

Figure 4. Permeability values of the mixture B permeated with water (open symbols) and with the leachate (solid symbols).

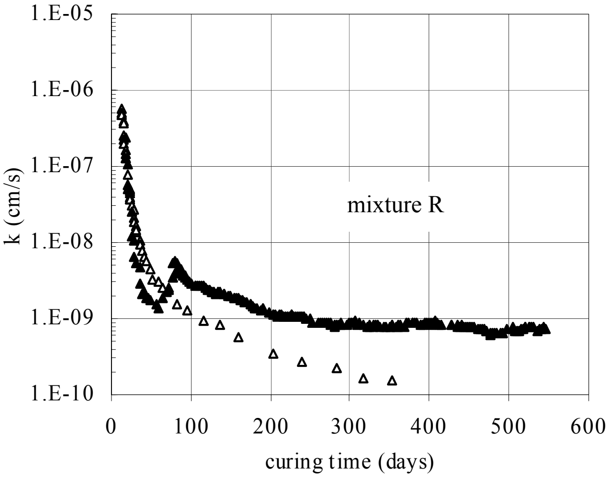

Figure 5. Permeability values of the mixture R permeated with water (open symbols) and with the leachate (solid symbols).

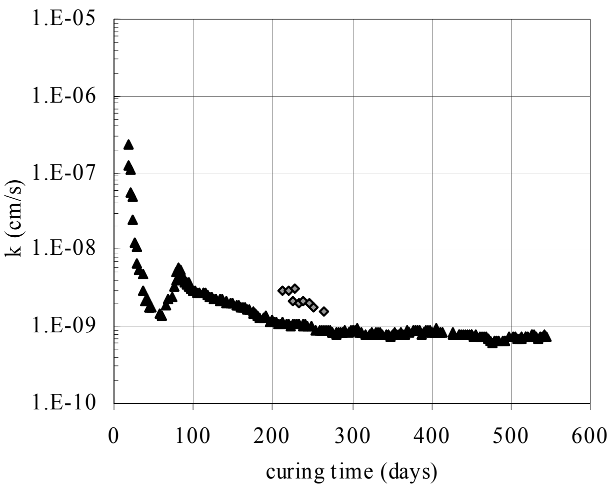

Figure 6. Permeability of the mixture R prepared in the laboratory and always permeated with the leachate and permeability of a sample taken from the in situ panel (grey symbols).

5 SUMMARY AND CONCLUSIONS

A composite cement-bentonite cut-off wall was proposed as confinement system for a phosphogypsum landfill with a very acidic leachate. Performance trials were required to select a proper mixture and to verify its general properties in the short and long term, to control its resistance to the aggressive leachate and to assess its durability. Immersion tests and permeation tests were performed to simulate very severe chemical conditions for the diaphragm, in order to accelerate the effects of chemical reactions. The results from all the tests and in particular from permeability tests showed a good performance, considering the severe conditions in terms of chemical concentrations (constant at the inlet) and pore volumes of flow induced through the samples.

Supposing the reaction effects to be dependent only on the numbers of pore volumes of flow of leachate through the mixture, the cut-off durability was roughly assessed to be of about 150 years. Even if the assumption of reaction effects dependent only on pore volumes of flow can be questionable, it is important to consider that the chemical conditions adopted in the tests are not expected to occur continuously in the field because the leachate used in all the tests was the worst one measured in the landfill, in terms of concentration levels and pH; moreover, a pumping system is provided into the landfill to reduce the leachate level inside the encapsulation, so that there should be an advective flow of groundwater and not of leachate across the barrier.

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