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Abstract

Excavation operations during construction produce several tons of soil, which frequently contain high concentrations of sulfates (>0.5 wt%). In accordance with the requirements of the French decree for waste classification for disposal, a soil containing sulfates is classified as "inert and non-hazardous waste" if the leachable sulfate concentration is lower than a mass fraction of 0.1%. To prevent sulfate leaching from excavated soils, solutions for immobilizing sulfates are needed and the associated stabilization mechanisms must be understood. On the other hand, the reuse of soils containing sulfates for civil engineering purposes can lead to significant risks after their treatment with ordinary cementitious binders because of chemical reactions involving sulfates. These reactions can promote the formation of massive ettringite crystals resulting in expansion, cracking and eventually catastrophic damage of materials or structures. For these reasons, the stabilization of soils containing sulfates by adding alternative hydraulic binders is studied in this paper. Several binders were used to treat a sulfate-spiked soil. It was observed that treatment with cementitious binders having high C_3A content led to volume expansions greater than 5%, while treatments with binders containing a high fraction of ground granulated blast furnace slag (GGBS) showed volume expansions of less than 5% and about 89% of sulfates were immobilized in the solid matrices. These preliminary results suggest that GGBS binders are effective for the treatment of soils containing sulfates. Moreover, numerical calculations using PHREEQC were compared with experimental results to improve the understanding of sulfate immobilization mechanisms.

Keywords: Sulfates; Immobilization; Soils; Volume expansion; PHREEQC

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1. Introduction

In France, excavated soils are considered as waste and they are classified in different categories depending on their concentration of leachable characteristic constituents. In accordance with the French decree on waste classification for disposal (Legifrance, 2014), a soil containing sulfates is classified as "inert and non-hazardous waste" if the leachable sulfate concentration is lower than a mass fraction of 0.1%. To decrease sulfate leaching from excavated soils, solutions for immobilizing sulfates are needed and the associated stabilization mechanisms must be understood. On the other hand, the reuse of soils containing sulfates for civil engineering purposes can be considered.

In civil engineering, the mechanical properties of soil are improved by adding lime or cement. However, it has been reported that lime treatment of a soil containing sulfates represents a potential risk to the stability and durability of the material application because of expansive reactions induced in the treated soil. Several studies have highlighted that sulfate-rich soils treated with either ordinary Portland cement or lime lead to expansion of the material (Cabane, 2004; Celik and Nalbantoglu, 2013; Colas, 2012; Harris et al., 2006; Hunter, 1988; Kolani et al., 2012; Puppala et al., 2003; Talluri et al., 2013; Wang et al., 2003).

Moreover, it has been shown that soils containing clays with a mass fraction of at least 10% and 1% of sulfates produced significant swelling after treatment with lime (Dermatas, 1995; Hunter, 1988). In a high pH system (pH>12), an expansive mineral known as hydrated calcium sulfoaluminate or ettringite ($3CaO·Al_2O_3·3CaSO_4·32H_2O$) can precipitate when sulfate, alumina and calcium ions, and water are available to react. In treated soils, alumina ions could come from either the cement used in the treatment or from the dissolution of clays in the soil because of the increase of pH after the lime-treatment (Celik and Nalbantoglu, 2013; Dermatas, 1995; Hunter, 1988; Puppala et al., 2004).

It has also been reported that the swelling potential of sulfate-rich soils is decreased when they are treated with binders other than lime (Celik and Nalbantoglu, 2013; Harris et al., 2006; Kota et al., 1996; Mahedi et al., 2018; Puppala et al., 2004, 2003; Talluri et al., 2013; Wang et al., 2003). Wang et al. (2003) investigated the stabilization of sulfate-rich soils by partially replacing ordinary Portland cement (OPC) by alternative materials such as ground granulated blast furnace slag (GGBS), class C fly ash (CFA) and amorphous silica (AS). The authors showed that the replacement of OPC by GGBS decreased the amount of expansion in the soil and no expansion was detected when CFA and AS were used. Similarly, Puppala et al. (2003) studied the effectiveness of the treatment of sulfate-rich soils (2000 mg/kg to 5000 mg/kg of dry mass) with sulfate-resisting cement of types I/II and V. By adding 5% of such cements, they observed a decrease in the free swelling from 30% to less than 5%.

In the literature, preliminary work in this field focused primarily on the evaluation of the swelling phenomena in sulfate-contaminated soils. However, not much is known about the decrease in leachable sulfate concentration after treatment and the sulfate stabilization mechanisms. This work

aims to (i) compare the capacity of several alternative binders to immobilize sulfates in a sulfate-spiked soil in natural pH conditions (pH 7) and (ii) understand the sulfate immobilization mechanisms by using a single step batch method. The stabilization technique was used to decrease the sulfate leaching from treated soils and the swelling phenomenon was studied for all the treatments. Stabilization groups together the chemical processes permitting the decrease in the potential risk of a hazardous waste due to the decrease in the toxicity and the water solubility of pollutants (Chen et al., 2009; Peysson, 2005).

In this study, four different binders were chosen to treat a sulfate-spiked soil: one Ordinary Portland Cement (OPC), one alternative clinker mainly composed of ye'elimite $C_4A_3\bar{S}$ and belite C_2S, one GGBS CEM III/C cement and one experimental binder composed of 90% GGBS and 10% OPC. Leachable sulfate concentrations were determined by using ion chromatography by analyzing eluates extracted from leaching tests carried out in accordance with the European standard NF EN 12457-2, which is required by French law (Legifrance, 2014). Leachable heavy metals concentrations were also determined by using Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES). Swelling of untreated and binder-treated soils was evaluated by performing volume expansion tests on compacted specimens. Additionally, mechanical properties were evaluated by determining the indirect tensile strength. Finally, sulfate stabilization mechanisms were studied using Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM/EDS) and X-ray diffraction (XRD) by analyzing mineral phases that could contain sulfates. In order to better understand the underlying sulfate stabilization mechanisms, experimental results were compared with numerical calculations obtained from a geochemical model using the PHREEQC code (Parkhurst and Appelo, 1999).

2. Materials and methods

2.1. Materials

2.1.1. Sulfate-spiked soil

Since a natural sulfate-rich soil was not available, it was chosen to artificially contaminate a soil from the Paris region. This soil was classified as a "silt" using the Unified Soil Classification System (USCS). Its chemical composition is presented in Table 1. It had a total organic carbon (TOC) content of about 2450 mg/kg of dry mass ± 6% and no heavy metal contamination was detected. In order to study the sulfate stabilization, the soil was spiked with 1.8% of gypsum ($CaSO_4 \cdot 2H_2O$) powder in order to obtain a mass fraction of 1% of sulfates (SO_4^{2-}) (10000 mg/kg of dry mass of soil). The sulfate dosage of 1% is reported as the level where sulfate-rich soils usually present swelling phenomena after cement treatment (Centre de recherches routières, 2004). In addition, gypsum was selected as the sulfate source because sulfates are generally found in this form in the soils of France (Société Française de Chimie, 2016; Wild et al., 1999). Moreover, gypsum is reported to be the major source of sulfate that produces swelling in soils treated with lime (Burkart et al., 1999). This mineral has the highest water

1 solubility among the common sulfate minerals found in soils (2.4 g/l at 25 °C) (Alexander et al., 2013;

2 Escadeillas and Hornain, 2008; Eswaran and Gong, 1991; Klimchouk, 1996).

3 *2.1.2. Binders*

4 Four cementitious binders, with the ability to chemically consume sulfates during their hydration

5 processes, were tested to stabilize sulfates contained in the soil. The chemical compositions of each

6 of the anhydrous binders are summarized in Table 1. The binders tested were:

7 -   One Ordinary Portland cement (OPC), CEM I 52.5 N CE CP2 NF, with a C_3A content of 8.6%. In

8   this study, it is referred to as "CEM I". It was expected that this binder would consume a large

9   amount of sulfates due to the reaction between the aluminate phases (C_3A) and the gypsum

10   contained in the soil to form ettringite. In this system, ettringite is not a suitable phase because it

11   can lead to volume changes. It should be noted that the main hydration products of this binder are

12   portlandite (CH) and calcium silicate hydrate (C-S-H) gel.

13 -   One sulfoaluminate clinker mainly composed of ye'elemite ($Ca_2Al_6O_{12}SO_4$ or $C_4A_3\bar{S}$ in cement

14   notation) and belite (Ca_2SiO_4 or C_2S in cement notation). It is referred to as "Clinker Y". It should

15   be noted that the SO_3 concentration in this binder is about 8 wt%, which is about half of the SO_3

16   content found in classic calcium sulfoaluminate binders (Glasser and Zhang, 2001). It was expected

17   that this binder would also consume a large amount of sulfates because its main hydration product

18   is ettringite, which is formed until sulfates are consumed and, then, monosulfate is precipitated

19   (Midgley and Bhaskara Rao, 1978; Winnefeld and Lothenbach, 2010).

20 -   One ground granulated blast furnace slag (GGBS) CEM III/C cement (according to EN 197-1),

21   composed of 82% GGBS, 15% Portland clinker and 3% of secondary constituents. The designation

22   of this binder is CEM III/C 32.5 N–LH/SR PM and it is referred to as "CEM III/C". This binder was

23   chosen to verify the effect of the proportion of slag and Portland clinker on the immobilization of

24   sulfates. The main hydration products in this type of binders (slag blended with Portland clinker)

25   include C-S-H phases, C-A-S-H (when alumina replaces some silica), hydrotalcite (M_5AH_{13}),

26   ettringite and AFm phases such as C_4AH_{13} (Chen and Brouwers, 2007; Kolani et al., 2012).

27 -   One experimental binder composed of 90% GGBS and 10% OPC, with the OPC designation

28   CEM I 52.5 N R CE CP2 NF. In this study, it is referred to as "90%GGBS+10%OPC". The addition

29   of Portland cement to GGBS is required to increase the pH and enable the hydration reaction of

30   GGBS and, thus, the precipitation of cementitious phases (Hewlett, 2004). In this binder, the main

31   hydration products are C-S-H, C-A-S-H, hydrotalcite, ettringite and AFm phases.

32 **Table 1** - Chemical compositions of the soil and each of the anhydrous binders (% by weight). The GGBS and

33 OPC binders presented in this table were used to fabricate the experimental binder "90%GGBS+10%OPC".

Oxide/phase composition	Soil	CEM I	Clinker Y	CEM III/C	Experimental binder	
					GGBS	OPC
CaO	13.23	64.48	45.10	46.09	43.80	64.04
SiO_2	57.70	20.22	10.56	31.29	37.41	21.87
Al_2O_3	7.20	5.23	23.47	10.46	10.20	3.96
Fe_2O_3	2.86	3.32	9.71	0.91	0.60	3.08
MgO	1.06	0.80	1.00	5.84	6.40	1.25
Mn_2O_3	0.11	0.00	0.01	0.00	0.20	0.03
Na_2O	0.74	0.28	0.17	0.53	0.21	0.15
K_2O	1.59	0.76	0.27	0.65	0.28	0.74
TiO_2	0.64	0.00	1.29	0.00	0.70	0.17
P_2O_5	0.21	0.00	0.11	0.00	0.10	0.10
SO_3	0.05*	3.02	8.07	2.52	0.10	3.35
Cl-	-	0.07	0.01	0.30	-	-
C_3A	-	8.60	-	8.40	-	8.00
C_3S	-	61.00	-	63.00	-	-
C_2S	-	-	20.30	-	-	-
C_4AF	-	11.10	-	10.50	-	-
$C_4A_3\bar{S}$	-	-	54.30	-	-	-
C_3MS_2	-	-	4.50	-	-	-
C_3FT	-	-	9.30	-	-	-
$C\bar{S}$	-	-	0.40	-	-	-
LOI (loss on ignition)	14.60	1.80	0.23	1.40	0	1.26

1 *SO_3 concentration in the natural soil. This soil was additionally spiked with 1.8% gypsum to obtain 1% of sulfates

2 (SO_4^{2-}).

3 *2.1.3. Sample preparation and curing*

4 All of the samples were fabricated in accordance with the French standard NF P94-100. The aim

5 of this standard is to evaluate the mechanical performance of a soil treated with either lime or hydraulic

6 binders. Before sample fabrication, the soil was dried at 40 °C and reduced to a particle size below

7 5 mm. Then, it was mixed dry with a mass fraction of 1.8% of gypsum in the form of powder in order to

8 obtain a mass fraction of 1% of sulfates (1% SO_4^{2-}).

9 The sulfate-spiked soil without binder was used as the reference or control formulation (F1). For

10 the other mixtures (F2, F3, F4 and F5), the sulfate-spiked soil was mixed dry with the binders using a

11 dosage of 10% of dry mass of soil. This binder dosage level was selected to ensure sufficient

12 mechanical properties for all of the treatments. Before compaction of the samples, the water content of

13 all the formulations was determined in accordance with the European standard NF EN 13286-2. This

14 standard specifies a method for determining the relationship between the water content and the dry

15 density of untreated or treated soils with hydraulic binders using Proctor compaction. Compaction was

16 carried out by using cylindrical test molds (50 mm in height and 50 mm in diameter) at 96% of the

17 maximum dry density. All of the samples were protected in hermetically sealed bags in order to

18 preserve the water content and were stored in a room at 20 °C until tests were conducted. The five

19 formulations tested are described in Table 2.

Table 2 – Designations and compositions of all the formulations studied (% of dry mass of soil).

Designation/Formulation	Composition	Water* to solid ratio (E/S)
F1 (untreated soil)	Silt + 1% SO_4^{2-}	0.17
F2	Silt + 1% SO_4^{2-} + 10% CEM I	0.15
F3	Silt + 1% SO_4^{2-} + 10% Clinker Y	0.15
F4	Silt + 1% SO_4^{2-} + 10% CEM III/C	0.17
F5	Silt + 1% SO_4^{2-} + 10% (90% GGBS+10% OPC)	0.17

* Water content determined in accordance with the European standard NF EN 13286-2

2.2. Test methods

2.2.1. Leaching tests

In this study, leaching tests were carried out in accordance with the European standard NF EN 12457-2 as required by French law (Legifrance, 2014). This standard allows the characterization and evaluation of leachable constituents of waste materials in a single step batch procedure at natural pH condition (pH 7). This method is useful to compare the capacity of several binder treatments to immobilize sulfates in a soil using distilled water as the leachant solution. Thus, the influence of leachant pH on the leachability of constituents from the sulfate-spiked soil is not covered in this study.

In order to perform the NF EN 12457-2 test, samples were cured for 28 days as described in section 2.1.3. Samples were crushed to a particle size below 4 mm and placed in 1 L polypropylene bottles containing distilled water with a liquid to solid ratio (L/S) of 10 L/kg. All bottles were shaken for 24 h in an end-over-end tumbler at 7 rpm. The solid residue was separated by filtration using a vacuum device and 0.45 µm membrane filters. After filtration, the conductivity and pH were measured in each of the eluates and then stabilized using a 2 v% nitric acid (HNO_3) solution. All of the resulting solutions were stored at 4 ºC until the chemical analyses were performed. A minimum of two specimens were used for each formulation. Arithmetic average results are presented with a confidence interval of 95%.

Sulfate immobilization was evaluated by measuring the leachable sulfate concentration using ion chromatography (Dionex Ion Chromatography System (ICS-3000)). Before analyses, all solutions were diluted by a factor of 100 using ultrapure water (18 MΩ.cm). Additionally, the conductivity, pH and heavy metal concentrations were measured in the leachate solutions. Heavy metal concentrations were determined using Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) (Optima 7000 DV machine). In this case, all solutions were diluted by factors of 10, 25 or 100 using ultrapure water (18 MΩ.cm) with 2 v% HNO_3.

2.2.2. Evaluation of swelling and mechanical properties

Swelling phenomena and mechanical properties of all mixtures were evaluated in accordance with the French standard NF P 94-100. This standard proposes the immersion of all treated soil samples in water at 40 ºC for 7 days. The aim of this immersion is to accelerate the hydration process and to verify whether the treatment leads to expansion of the material (Wang et al., 2003). Before the immersion period, this standard recommends a specific curing time for treated soils. When soil is treated with lime, the curing period is 3 days ± 4 h. For soil treatment with other binders, the minimum curing time recommended is 4 h ± 15 min and the maximum curing time is 7 days. Therefore, in this study, the minimum and maximum curing times were tested on all binders. Preliminary testing showed that a 4-h curing appeared to be insufficient to meet the requirements for splitting tests (results not presented here). For this reason, a curing of 7 days was chosen for both volume expansion and splitting tests.

For volume expansion tests, initial geometrical measurements of all samples were taken after 7 days of curing using a digital sliding caliper and final volumes were measured at the end of the immersion by applying Archimedes' principle. Specimens used to evaluate mechanical properties followed a preparation procedure similar to that for volume expansion tests. At the end of the immersion period, the indirect tensile strength was assessed by carrying out splitting tests in accordance with European standard NF EN 13286-42, which proposes diametral compression tests on compacted soil specimens. For both volume expansion and splitting tests, three specimens were used per formulation. Arithmetic average results are presented with a confidence interval of 95%.

This standard takes two criteria into account to assess the performance of a soil treated with hydraulic binders: (1) volume expansion, and (2) indirect tensile strength. These criteria and their guideline values are presented in Table 3.

Table 3 – Criteria and guideline values to evaluate the performance of a soil treated with hydraulic binders according to the French standard NF P 94-100.

Performance of the treated-soil	Parameter (after immersion for 7 days)	
	Volume expansion (Gv) in %	Indirect tensile strength (Rit) in MPa
Suitable	≤ 5	≥ 0.2
Doubtful	5 < Gv ≤ 10	0.1 ≤ Rit < 0.2
Unsuitable	> 10	< 0.1

2.2.3. Mineralogical and microstructural characterizations

In order to understand the sulfate-stabilization mechanisms, mineralogical and microstructural analyses were carried out on treated soil samples using Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM/EDS). SEM observations were made on carbon-coated fracture surfaces and on polished sections, previously mounted in resin. Two different scanning electron microscopes were used, a JEOL JSM-6380LV and a JEOL JSM 7800, equipped with Rontec XFLASH 3001 and SDD X-Max 80mm2 EDS detectors, respectively. Images were obtained with both secondary (SE) and back-scattered (BSE) electrons with magnifications between 100x and

1   10000x. SEM observations coupled with EDS mapping and EDS punctual analyses were also carried

2   out in order to identify correlations between various chemical elements and sulfate.

3   Additionally, X-ray diffraction (XRD) analyses were performed in order to identify mineral phases

4   that could contain sulfates. A Bruker D8 diffractometer with the Bragg-Brentano configuration and

5   copper radiation source (Cu Kα, λ=1.54 Å) was used. The anode voltage was 40 kV and the electric

6   current intensity was 40 mA. XRD analyses were carried out on powder previously crushed to a particle

7   size below 40 µm.

8   *2.3. Modeling*

9   Experimental results were compared to numerical calculations in order to better understand the

10   sulfate stabilization mechanisms. Simulations were carried out using the PHREEQC code version 3.0

11   (Parkhurst and Appelo, 1999). In this code, mineral-solution equilibria are calculated considering the

12   extended Debye-Hückel activity coefficient model and using the Debye-Hückel B-dot relation (C.

13   Helgeson, 1969). The thermodynamic properties used in this work came from the Thermoddem

14   database (Blanc et al., 2012). Hydrogarnet phase ($C_3(A,F)S_{0.84}H_{4.32}$), which is a typical cementitious

15   phase in the studied system, was added to this database using the Gibbs free energy of formation

16   presented by Lothenbach et al. (2019).

17   In the geochemical model, the initial solution was defined using an *L/S*=10 L/kg, a temperature of

18   *T*=25 °C and a *pH* of 7.0. According to chemical analyses (Table 1) and XRD analyses, the soil was

19   defined as a combination of four main constituents represented in the form of quartz (SiO_2), calcite

20   ($CaCO_3$), illite ($K_{0.85}Al_{2.85}Si_{3.15}O_{10}(OH)_2$) and magnetite ($Fe_2O_3$) in molar proportions (mol%) of 72.8%,

21   21.2%, 4.4%%% and 1.6%, respectively. Sulfate was introduced into the model using 10.4 mmol of

22   gypsum (a constant sulfate concentration of 1% was used for all of the simulations). Finally, the binder

23   was defined by its elemental chemical composition expressed in moles.

3. Results

3.1. Leaching tests

26   Leachable sulfate and heavy metal concentrations, conductivity, and pH of eluates extracted from

27   leaching tests for each of the formulations are presented in Table 4. Concentrations exceeding the

28   thresholds are in bold. Leachate concentrations are expressed in mg/kg of dry mass of solid (soil and

29   binder) and are compared to the "inert and non-hazardous waste" thresholds established by French

30   law (Legifrance, 2014) in order to verify that no significant heavy metal concentrations were released

31   after binder treatment. Leachate concentrations of all formulations are schematically represented in

32   Figure 1. In this figure, the radial axes represent the concentrations measured for each element

1 normalized by their thresholds, which are established by French law and represented by the red dotted

2 line. Outside this line, thresholds are not respected.

3 **Table 4** - pH, conductivity values and leachable sulfate, chloride, fluoride and heavy metal concentrations of all

4 formulations. Leachate concentrations are expressed in mg/kg of dry mass. Bold values indicate the

5 concentrations exceeding the thresholds established by French law (Legifrance, 2014).

Chemical species	Units	Inert and non-hazardous waste Threshold	F1	F2	F3	F4	F5
pH	-	-	7.5	12.4	11.7	11.3	11.4
Conductivity	µS/cm	-	1662	3425	747	653	1050
Sulfates (SO_4^{2-})	mg/kg	1000	**9174**	138	759	986	785
Arsenic (As)	mg/kg	0.5	<0.2	<0.2	<0.2	<0.2	<0.2
Barium (Ba)	mg/kg	20	0.5	0.6	0.2	0.2	0.3
Cadmium (Cd)	mg/kg	0.04	<0.002	<0.002	<0.002	<0.002	<0.002
Chromium (Cr)	mg/kg	0.5	<0.1	**0.8**	**2.3**	<0.1	<0.1
Copper (Cu)	mg/kg	2	<0.2	0.4	<0.2	<0.2	<0.2
Mercury (Hg)	mg/kg	0.01	<0.001	<0.001	<0.001	<0.001	<0.001
Molybdenum (Mo)	mg/kg	0.5	0.1	0.1	0.3	<0.01	0.2
Nickel (Ni)	mg/kg	0.4	<0.1	0.3	<0.1	<0.1	0.1
Lead (Pb)	mg/kg	0.5	<0.1	<0.1	<0.1	<0.1	<0.1
Antimony (Sb)	mg/kg	0.06	<0.002	<0.002	<0.002	<0.002	<0.002
Selenium (Se)	mg/kg	0.1	<0.01	<0.01	<0.01	<0.01	<0.01
Zinc (Zn)	mg/kg	4	<0.2	<0.2	<0.2	<0.2	<0.2
Chlorides (Cl^-)	mg/kg	800	18.6	3.7	50.4	156.7	27.1
Fluorides (F^-)	mg/kg	10	3.3	6.9	2.0	2.2	2.1

F1: Untreated soil
F2: CEM I
F3: Clinker Y
F4: CEM III/C
F5: 90%GGBS+10%OPC

– – Inert and non-hazardous waste threshold

6

As mentioned previously, the initial sulfate concentration in the soil was 10000 mg/kg of dry mass. When leaching tests were carried out in the untreated sulfate-spiked soil (formulation F1), it was found that the leachable sulfate concentration was approximately 10 times higher than the guideline value for "inert and non-hazardous waste" (1000 mg/kg of dry mass) established by French law (Legifrance, 2014). As shown in Table 4 and Figure 1, for the untreated soil, the leachable sulfate concentration was about 9174 mg/kg of dry mass ± 16%. In contrast, treatment of the sulfate-spiked soil with CEM I binder (formulation F2) showed a noteworthy decrease in leachable sulfate concentration, which was below the guideline value. In this case, the sulfate concentration released in solution was about 138 mg/kg of dry mass ± 9%. Similarly, treatment of sulfate-spiked soil with the sulfoaluminate binder "Clinker Y" (formulation F3) significantly decreased the leachable sulfate concentration. In this treatment, a sulfate concentration of 759 mg/kg of dry mass ± 12% was detected in solution. However, in both formulations F2 and F3, chromium (Cr) was also detected in solution with a higher concentration than the limit for "inert and non-hazardous waste" (0.5 mg/kg of dry mass). Therefore, the use of these two binders in the treatment of the sulfate-spiked soil represents a disadvantage due to the high concentration of Cr released in solution.

On the other hand, sulfate-spiked soil treated with GGBS-binders (formulations F4 and F5), showed the decrease in leachable sulfate concentration and leachable heavy metal concentrations were well within the established limits. For the formulation F4 (CEM III/C), the sulfate concentration released in solution was 986 mg/kg of dry mass ± 21% and for formulation F5 (90%GGBS+10%OPC), the leachable sulfate concentration was 785 mg/kg of dry mass ± 8%.

It should be noted that, for all formulations, leachable chlorides (Cl⁻) and fluorides (F⁻) concentrations were below the "inert and non-hazardous waste" thresholds established by French law, with less than 800 mg/kg of dry mass for Cl⁻ and less than 10 mg/kg of dry mass for F⁻.

3.2. Evaluation of swelling phenomena and mechanical properties

Figure 2 shows the indirect tensile strength and the volume expansion results for each formulation. For formulation F1 (untreated soil), splitting tests were not performed because of insufficient mechanical strength. Therefore, an indirect tensile strength of 0 MPa is indicated in Figure 2(a).
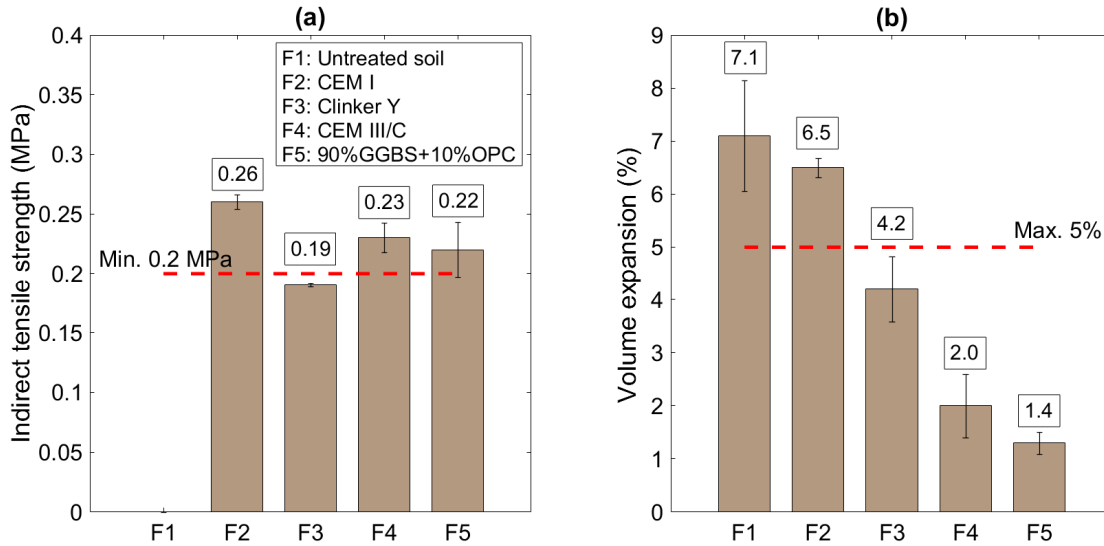
Figure 2 – Evaluation of mechanical properties and swelling for all formulations. The red dotted lines represent the guideline established by French standard NF P 94-100. **(a)** Indirect tensile strength is expressed in MPa. **(b)** Volume expansion is expressed in percentage.

It can be observed that formulation F1 (untreated soil) had a volume expansion of 7.1% ± 1.1%, which is greater than the maximum guideline value of 5%. The soil expansion is probably due to the presence of swelling clays. On the other hand, soil treatment with CEM I (formulation F2) presented an indirect tensile strength of 0.26 MPa ± 0.01 MPa, which is higher than the minimum guideline value of 0.20 MPa. However, the volume expansion value was higher than 5%, classifying this treatment as "doubtful" (cf. Table 3). Treatment of the sulfate-spiked soil with the "Clinker Y" binder (formulation F3) was also classified as "doubtful" because the indirect tensile strength reached approximately 0.19 MPa ± 0.002 MPa, which was lower than the accepted guideline value (0.2 MPa). In addition, the volume expansion was close to the limit value of 5% (4.2% ± 0.6%).

Treatment of the sulfate-spiked soil with binders containing high slag levels, such as CEM III/C and "90%GGBS+10%OPC" (formulations F4 and F5, respectively) resulted in a significant decrease in the swelling and maintained an indirect tensile strength above 0.20 MPa. For formulations F4 and F5, the volume expansion reached 2.0% ± 0.6% and 1.4% ± 0.2%, respectively; and the indirect tensile strengths were about 0.23 MPa ± 0.03 MPa and 0.22 MPa ± 0.06 MPa, respectively. As such, both treatments F4 and F5 were classified as "suitable".

3.3. Microstructural characterization: general observations

SEM observations were carried out on fracture sections of all the formulations and the images obtained are presented in Figure 3. SEM images of formulation F1 (untreated soil) are presented in Figure 3(a) and Figure 3(b). As shown, gypsum was the sulfate specie observed and its chemical composition was verified by punctual EDS analyses, having a CaO to SO_3 molar ratio of 1. SEM images

of fracture surfaces of formulations F2 and F3 are shown in Figure 3(c) and Figure 3(d), respectively. In both formulations, ettringite crystals were easily identified. For soil treated with CEM I binder (F2), ettringite was observed at higher magnifications than for soil treated with "Clinker Y" binder (F3). Conversely, for formulations containing high GGBS levels (F4 and F5), no expansive phases were observed on the fracture surfaces. As presented in Figure 3(e) and Figure 3(f), the SEM images of F4 and F5 formulations showed a microstructure consisting of an agglomeration of phases, without the typical morphology of sulfate-bearing minerals such as ettringite or gypsum.
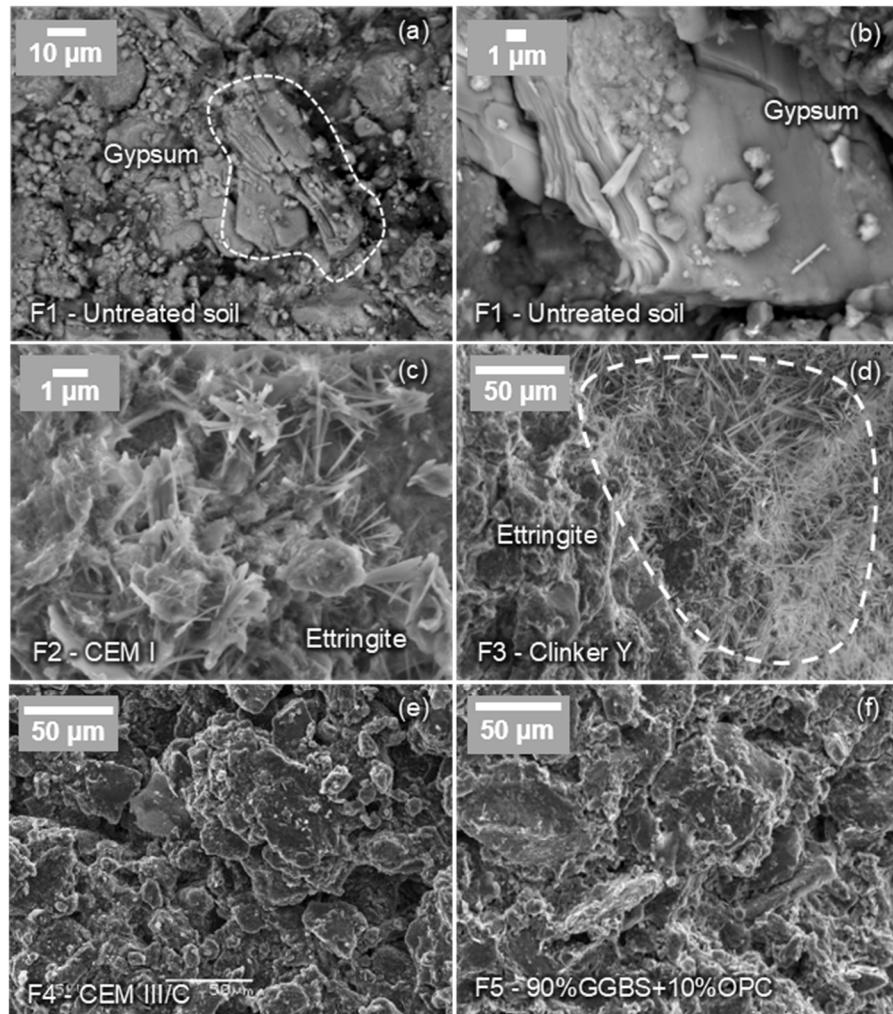
Figure 3 – SEM images obtained from untreated and treated soils. **(a)**, **(b)** Formulation F1 (untreated soil) showing sulfate in the form of gypsum. **(c)** Formulation F2 (CEM I) showing ettringite crystals. **(d)** Formulation F3 (Clinker Y) showing ettringite precipitates. **(e)** Formulation F4 (CEM III/C) showing agglomeration of phases **(f)** Formulation F5 (90GGBS+10OPC) showing agglomeration of phases without typical morphology of sulfate-bearing crystal.

3.4. Focus on formulation F5: experimental and numerical results

Particular attention was paid to the experimental binder composed of 90% GGBS and 10% OPC (formulation F5) due to its capacity to immobilize sulfates (89% of sulfates immobilized in the solid)

without releasing heavy metals into solution. Additionally, this treatment did not show significant volume expansions, as was seen in Figure 2. In order to better understand the sulfate stabilization mechanisms in this formulation, it was decided to extend the microstructural analyses and to compare experimental data with numerical calculations.

3.4.1. Microstructural characterization: formulation F5

As explained in section 3.3, the identification of mineral phases containing sulfates was difficult and no typical morphology of sulfate-bearing crystals such us ettringite and gypsum was observed on the fracture surfaces of formulation F5 (cf. Figure 3(f)). Therefore, SEM observations coupled with EDS mapping and EDS punctual analyses were carried out on polished sections of this formulation. One of the EDS mapping analyses is shown in Figure 4(a). EDS mapping analyses provided information about the distribution of sulfates in this sample and revealed that sulfates were always associated with calcium. In order to verify the composition of the species containing sulfates, EDS punctual analyses were performed in these zones. Ettringite was the dominant phase identified in the zones presenting high concentrations of sulfates (SO_3 to CaO molar ratio of 0.50 and an Al_2O_3 to CaO molar ratio of 0.17).

SEM/EDS analyses were completed by carrying out XRD analyses in the soil before and after treatment. XRD patterns obtained from formulation F5 were then compared to XRD patterns obtained from untreated sulfate-spiked soil (F1). XRD analyses are presented in Figure 4(b). The pattern for untreated soil (F1) revealed a large peak between 2θ 11° and 12°, which is characteristic of the presence of gypsum. The XRD pattern of formulation F5 showed the decrease in intensity of gypsum peaks and revealed the appearance of new peaks between 2θ 9° and 10° and 15° and 16°, which are characteristic of the ettringite phase. However, residual gypsum in formulation F5 revealed that gypsum was not completely dissolved by the (probably insufficient amount of) water provided to the system during the fabrication of the samples.
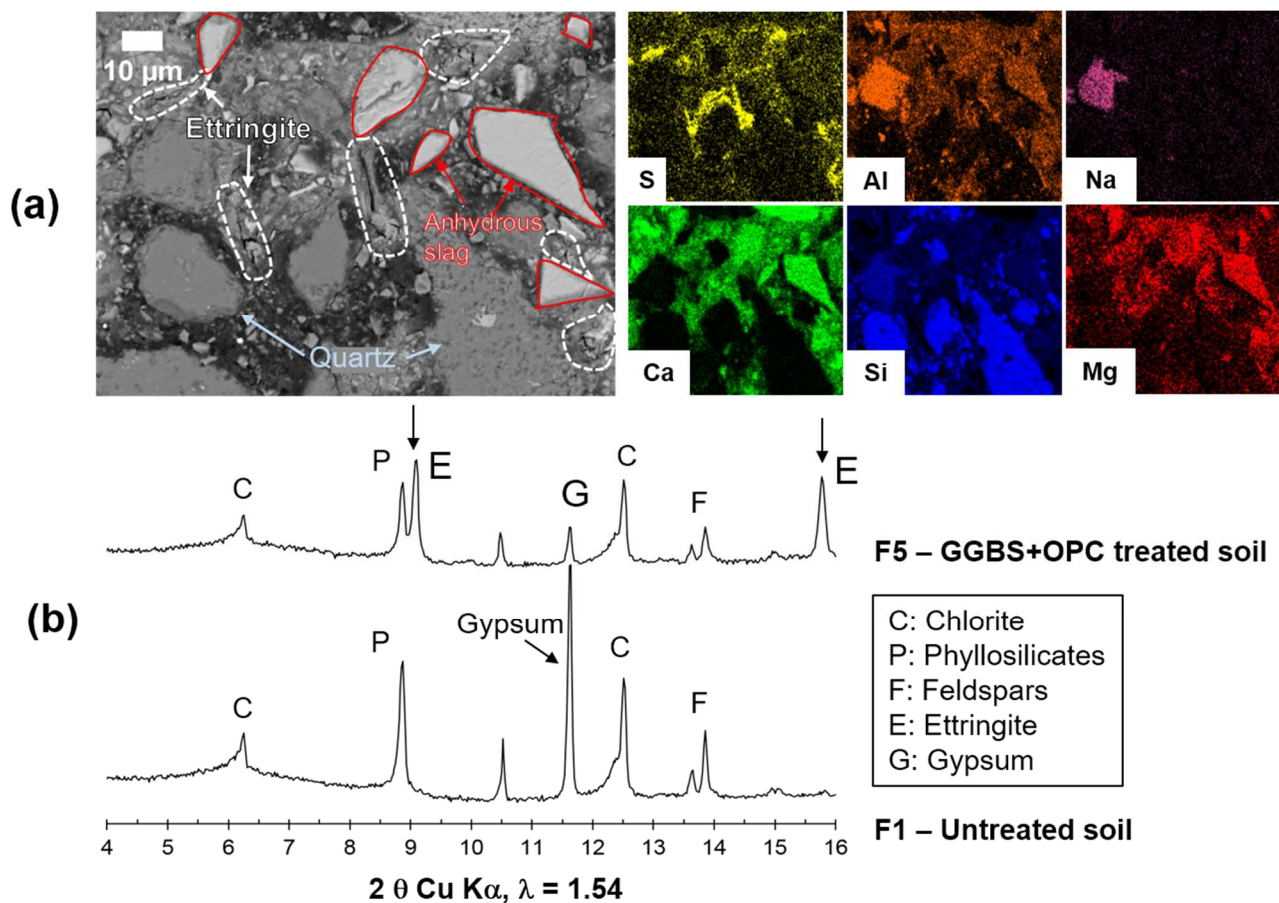
Figure 4 – Identification of ettringite in formulation F5 at 28 days of curing. **(a)** At left, BSE image of polished section indicating the zones where ettringite phase was found. At right, EDS elemental mapping showing the zonation of sulfur, calcium, aluminum, sodium, silicon and magnesium elements. **(b)** Selected range of the XRD pattern obtained from formulation F1 (untreated soil) and formulation F5 showing the reduction of intensity of the gypsum peak (G) and the appearance of ettringite peaks (E) in formulation F5.

3.4.2. Experimental vs. modeling results: formulation F5

In order to better understand sulfate stabilization mechanisms, numerical modeling was performed for formulation F5 and experimental and numerical results were compared. Thus, the influence of the proportion of GGBS+OPC binder on sulfate immobilization in the sulfate-spiked soil was evaluated by varying the dosage of the binder from 0% to 20% (in percentage by mass) as presented in Table 5. Assuming congruent dissolution in the model, this experimental binder was defined by its elemental chemical composition expressed in moles (cf. Table 1).

The results of batch-reaction calculations were compared to leachable sulfate and leachable major element concentrations, which were experimentally determined after leaching tests at 28 days of curing. Consequently, the kinetics of chemical reactions were not taken into account in this model, which represents only the state of equilibrium reached by the system. For some minerals defining the soil constituents, such as quartz and iron(III) oxide (magnetite), the dissolution was not permitted due to

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their very slow kinetics of dissolution at 20 °C in alkaline conditions (Brady and Walther, 1990; Dove and Crerar, 1990; Schwertmann, 1991).

Since few thermodynamic data exist for GGBS binder, several calculations were made by assuming its hydration at four different degrees: 10%, 20% 30% and 40%. As reported in the literature, the hydration of GGBS and OPC mixes is more complex than that of Portland cement. GGBS reacts more slowly with water and not all the GGBS binders react in the same way (Hewlett, 2004). It is largely recognized that the rate of hardening of GGBS binders is slower than that of Portland cement during the first 28 days of curing (Hewlett, 2004). Lothenbach et al. (2012) showed that, over one month, approximately 70% of the slag had reacted in a system containing about 60% slag and a water to binder (W/B) ratio of 1.1. Taylor et al. (2010) reported that, over two years, approximately 68% of the slag had reacted in a Portland-slag system containing about 50% slag and an W/B ratio of 0.4. It seems that higher hydration degrees are estimated in systems containing large amounts of available water. Taylor et al. (2010) showed that the degree of slag hydration in a slag-blended system also depended on the proportion of slag provided. This means that, over the same time period, a system containing high proportions of slag presents lower rates of slag reaction compared with systems containing lower slag amounts. Taylor et al. (2010) estimated that, over two years of reaction, only 36% of slag had reacted in a formulation containing 90% GGBS and 10% OPC.

In the current investigation, it is, therefore, important to consider the hydration degree of the GGBS as a significant factor influencing the immobilization of sulfates in formulation F5.

Table 5 – Formulations used to verify the sulfate stabilization mechanisms in formulation F5 (90% GGBS+10%OPC) (% of dry mass of soil).

Designation	Composition
F5-20	Silt + 1% SO_4^{-2} + 20% 90GGBS+10OPC
F5-15	Silt + 1% SO_4^{-2} + 15% 90GGBS+10OPC
F5-12	Silt + 1% SO_4^{-2} + 12% 90GGBS+10OPC
F5-10	Silt + 1% SO_4^{-2} + 10% 90GGBS+10OPC
F5-8	Silt + 1% SO_4^{-2} + 8% 90GGBS+10OPC
F5-7	Silt + 1% SO_4^{-2} + 7% 90GGBS+10OPC
F5-5	Silt + 1% SO_4^{-2} + 5% 90GGBS+10OPC
F5-2	Silt + 1% SO_4^{-2} + 2% 90GGBS+10OPC

Experimental and numerical data of pH values, sulfate, calcium, aluminum, silicon and magnesium concentrations in solution are plotted in Figure 5, where element concentrations are given in mM (mmol/L). All the data were plotted as a function of the binder dosage (ranging from 0% to 20%). Experimental data are represented by black circles while model calculations are given by lines. Solution concentrations are also completed by the molar quantities of solid phases involved in the modeled reactions (Figure 6). These quantities are divided into two groups here for ease of reading: silicon

dominant phases (M-S-H, C-S-H and illite) are shown in Figure 6(a) and aluminous dominant phases (ettringite, hydrogarnet and gibbsite) in Figure 6(b).

As shown in Figure 5(a), experimental and numerical data were in good agreement for the sulfate concentrations in solution. Experimental data and simulations showed that sulfate concentration in solution decreased when the content of "GGBS+OPC" binder increased. Experimentally, the sulfate concentration in solution remained relatively stable for formulations containing more than 7% of binder. This behavior can be explained by the ettringite stability as shown in Figure 6(b). In addition, simulations showed that the GGBS hydration level seemed to have an impact on the slope of the curve representing the sulfate concentration in solution. Depending on the GGBS hydration level considered, a modification on the sulfate concentration in solution was observed. However, the GGBS hydration level no longer affected the sulfate concentrations calculated beyond 15% of binder dosage. The best agreement between experimental and numerical data was obtained when a GGBS hydration level of 30% was assumed.

The results obtained for other leached elements are shown from Figure 5(b) to Figure 5(e). As for sulfates, the most important changes are observed for formulations containing less than 10% of binder. Calcium, silicon and magnesium concentrations calculated in solution are mostly controlled by C-S-H and M-S-H as shown in Figure 5(b), Figure 5(d), Figure 5(e) and Figure 6(a). On the other hand, aluminum concentrations in solution presented a more complex behavior due to the competition between aluminous phases (cf. Figure 5(c)). For formulations ranging from 0% to 10% of binder, aluminum presented a first evolution mainly dominated by ettringite formation as shown in Figure 6(b).

Experimental data for formulations with high binder dosage (>10%) showed that a maximum aluminum concentration was reached between 10% and 15% of binder dosage and then remained relatively stable. This behavior was not well modeled in the calculation results. The authors think that the difference found between numerical and experimental results in aluminum behavior could be explained by the aluminum mineral phases used in the model. In fact, it was verified that a decrease of 1% in the aluminum content present in the illite (used in the model to represent the soil), decreased the aluminum concentrations in solution by about 50%, without modifying the trend of the curve shown in Figure 5(c).

The pH values measured and calculated in solutions are shown in Figure 5(f). The first value measured in the system without binder (untreated soil) was close to 7.4. After the addition of the binder to the soil, pH increased significantly in the solution (pH 11-12). It seems that the proportion of binder in the treatment had a little effect on the variation of pH in the solution since it stayed relatively constant between pH values 11 and 12. This evolution was well reproduced by the model (but was slightly lower than experimental values) and was mainly controlled by the C-S-H composition.
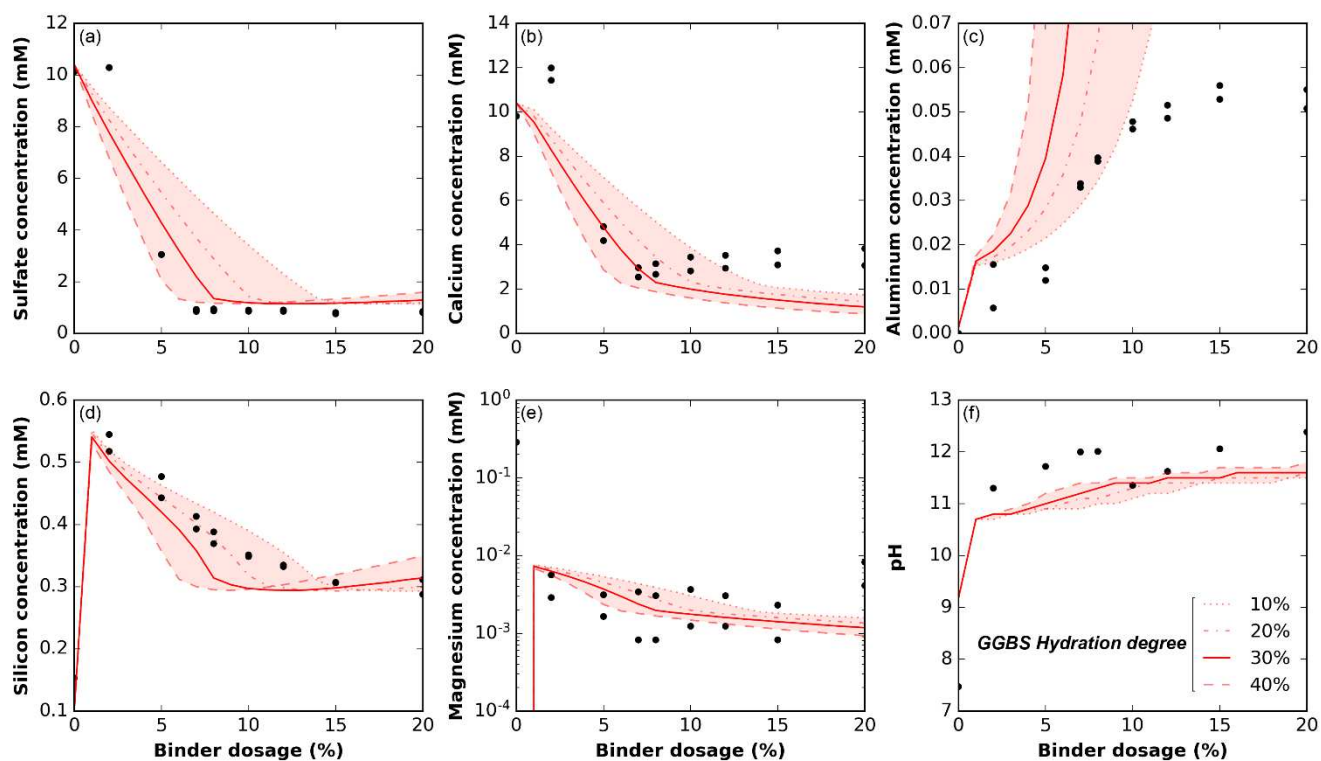
Figure 5 – Comparison between numerical and experimental data obtained from formulation F5 (90%GGBS+10%OPC). All the data are plotted as a function of the binder dosage. Black circles: experimental data. Line with larger dots: 40% GGBS hydration level. Solid line: 30% GGBS hydration level. Line with smaller dots: 20% and 10% GBBS hydration. **(a)** Leachable sulfate concentration, **(b)** leachable calcium concentration, **(c)** leachable aluminum concentration, **(d)** leachable silicon concentration, **(e)** leachable magnesium concentration (using a logarithmic scale), **(f)** pH values.
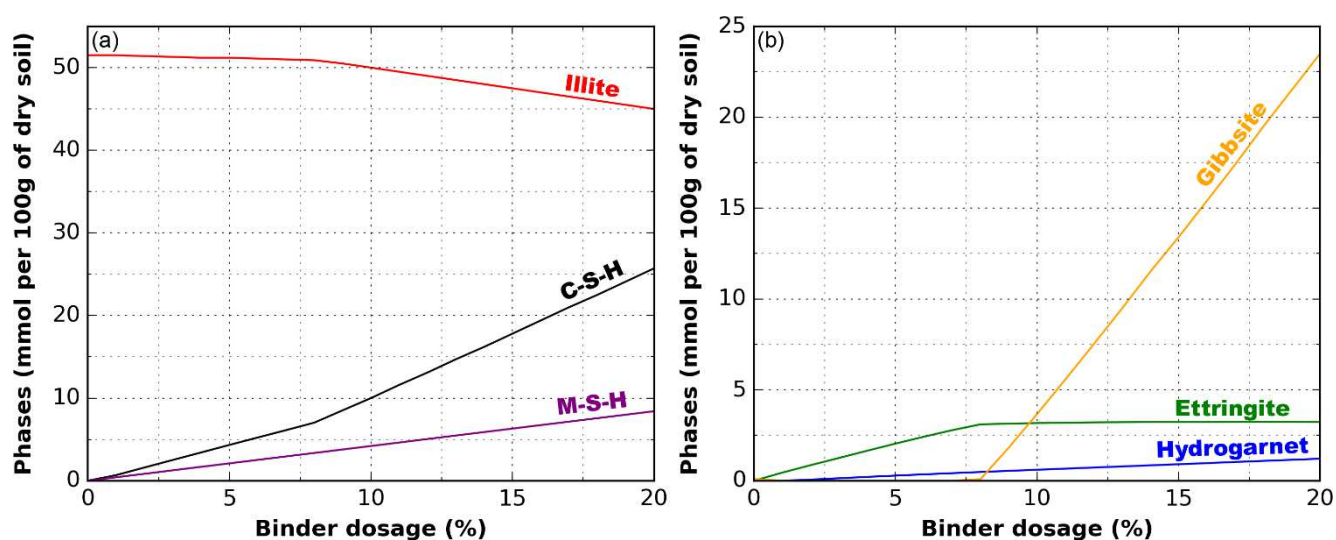

Figure 6 – Molar quantities of major phases calculated from the geochemical simulation of formulation F5 (90%GGBS+10%OPC) assuming a GGBS hydration level of 30%. All the data are plotted as a function of the binder dosage. **(a)** Illite, C-S-H and M-S-H phases, **(b)** Ettringite, gibbsite and hydrogrdarnet.

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4. Discussion

4.1. Sulfate stabilization with CEM I and Clinker Y binders

Results presented in this paper confirm that treatment of sulfate-contaminated soils with cementitious binders is useful to decrease sulfate leaching. As presented previously, untreated sulfate-spiked soil released of about 90% of sulfates in solution. It was verified that the treatment with ordinary Portland cement (formulation F2) and Clinker Y (formulation F3) showed a large decrease in sulfate concentration in solution after leaching tests. For formulation F2, it was calculated that a mass fraction of about 99% of sulfates were fixed in the solid. For formulation F3, the percentage of sulfates fixed in the solid was 92%. As explained in the literature, these binders provided aluminum to the system and also increased the pH (Hunter, 1988), which led to the instability of gypsum contained in the soil and to the precipitation of ettringite as shown in Figure 3 and Table 4. Ettringite is reported to be the most stable phase under these conditions permitting the decrease in leachable sulfate concentration (Brown and Bothe, 1993; Cody et al., 2004; Feng et al., 2016; Warren and Reardon, 1994).

Nevertheless, the stabilization of sulfates in soils by these two binders (CEM I and Clinker Y) is limited because of: (i) the release of chromium and (ii) the significant swelling measured. In fact, chemical analyses of eluates after leaching tests showed that both formulations F2 and F3 released chromium (Cr) at concentrations that were higher than the established "inert and non-hazardous waste" threshold (0.5 mg/kg of dry mass of soil) (cf. Table 4 and Figure 1). Previous studies have investigated the presence of chromium in cements (Eštoková et al., 2012; Sinyoung et al., 2011; Sorrentino, 2011). Sinyoung et al. (2011) conducted leaching tests in cement clinkers and found that chromium exceeded the allowable leachable concentration established by the United States Environmental Protection Agency (US EPA). Eštoková et al. (2012) indicated that the concentration of soluble chromium in binders was proportional to the OPC clinker content. That explains why some types of cements, such as CEM III or CEM V, have a low chromium content; in these cases, chromium content decreases with OPC content. In addition, it has been shown that slag can reduce Cr(VI) to Cr(III) due to the reducing properties of this material. As reported in the literature, Cr(III) is less soluble than Cr(VI), therefore, slag binders decrease the leachable Cr concentration (Allan and Kukacka, 1995; Kindness et al., 1994).

In terms of swelling, both formulations F2 and F3 had larger volume expansions than the expansions measured in formulations F4 and F5. Swelling phenomena in F2 and F3 formulations can be attributed to the presence of ettringite crystals in the matrices, as shown in in Figure 3 (ettringite was observed at higher magnifications in F2 than for F3). The marked expansion in formulation F2 (CEM I-treatment) was expected due to the large aluminate content in the form of C_3A (8.6%), which reacted with gypsum to form ettringite. These results support previous findings in the literature where noteworthy swelling was reported on sulfate-contaminated soils treated with OPC or lime (Dermatas, 1995; Puppala et al., 2004, 2003; Wang et al., 2003). Moreover, volume expansion on formulation F2

was greater than that measured for formulation F3 (6.5% and 4.2%, respectively) (cf. Figure 2). The authors think that this can be explained by the difference in the type of ettringite formed. Mehta, (1983) reported that differences in the crystal habit and size of ettringite can affect the expansion behavior of the material. Mehta, (1983) proposed two types of ettringite crystals. Ettringite crystals of type 1 has large lath-like crystals ranging from 10 μm to 100 μm and several micrometers thick and it is usually formed during the hydration of supersulfated cements such as the sulfoaluminate clinker used in the present formulation F3. This type of ettringite is not expansive. Conversely, ettringite of type 2, presenting small rod-like crystals (1 μm to 2 μm long) is considered as expansive because of large amounts of these crystals have been found in deteriorated concretes under sulfate attacks. Additionally, some observations have shown that microcrystalline ettringite is capable of adsorbing large amounts of water on the surface, causing volume changes (Mehta, 1973). Type 2 ettringite is assumed to be the ettringite observed in formulation F2.

In contrast, significant volume expansion was not expected on samples treated with the "Clinker Y" binder (formulation F3) because the main hydration product of sulfoaluminate clinkers is ettringite, which usually precipitates during the consumption of the calcium sulfate after 1 to 2 days of hydration (Winnefeld and Lothenbach, 2010). However, we think that, in this treatment, the volume expansions can be explained by a delay in hydration due to the unavailability of water at the beginning of the hydration process. It should be noted that the water to solid ratio (W/S) of formulation F3 was about 0.15, meaning that not enough water was available to hydrate the binder during the first few days of curing. As explained previously, volume expansion tests were conducted at 7 days of curing and samples were immersed in water at 40 °C for 7 days. Under these conditions, the additional water provided to the system could lead to the precipitation of secondary ettringite and consequently the expansion of the samples.

4.2. Sulfate stabilization with GGBS binders

As presented in Table 4 and Figure 1, treatments of sulfate-spiked soil with GGBS binders (formulations F4 and F5) showed a large decrease in sulfate concentration in solution after leaching tests. It was calculated that of about 89% of sulfates were immobilized in the solid. Reduced species of sulfur present in GGBS as sulfides and native sulfur (Allan and Kukacka, 1995; Chaouche et al., 2017) could play an important role to decrease the leachable sulfate concentration in F4 and F5 treatments. However, it should be noted that the amount of sulfur provided to the treatment by the addition of GGBS binder is about 0.009%wt, which seems very low to significantly decrease the leachable sulfate concentration. Additionally, leachable heavy metal concentrations were well within the established limits. For F4 and F5, pH values in solution were lower than pH measured in formulations F2 and F3. This condition could have decreased the release of anionic heavy metals such as chromate, which is strongly pH dependent (Alloway, 2013; Bourg and Loch, 1995; Leisinger et al., 2014). As mentioned previously, GGBS has reducing properties due to reduced sulfur species. It has

been shown that Cr(VI) can be reduced to Cr(III) when GGBS is added to OPC mixtures. Thus, leachable Cr concentration decreases after leaching tests (Allan and Kukacka, 1995; Kindness et al., 1994).

SEM/EDS and XRD analyses showed that sulfates were partially consumed by the formation of ettringite phase (cf. Figure 4). These experimental observations were confirmed by modeling. Numerical calculations showed that ettringite was the most stable phase at the equilibrium of the system. In this study, the precipitation of ettringite was defined as one of the mechanisms dominating the immobilization of sulfates in the GGBS-treated soil. It should be noted that, in the geochemical model, a hydration level of 30% for the GGBS binder was assumed, which enabled good agreement between numerical and experimental results. The low hydration level of GGBS binder at 28 days can explain the difficulty in experimentally identifying the phases containing sulfates in formulation F5 and also other possible mechanisms of sulfate stabilization. Wild et al., (1993) studied the stabilization of sulfates in sulfate-rich clays treated with GGBS-lime binders. They proposed that the amorphous phase C-A-S-$\bar{S}$-H was initially formed, which seems to be a precursor of ettringite formation. Similarly, Wild et al. (1999) suggested that, in lime-slag systems, gypsum is consumed but part of the products formed during the reaction are not crystalline. This can be another explanation of the difficulty in identifying sulfate-bearing phases in the GGBS formulations F4 and F5.

Unlike treatments F2 and F3, formulations F4 and F5 did not reveal ettringite formation to be a source of large volume expansions. GGBS-treated soils had volume expansions lower than 5%, which demonstrated the ability of GGBS binders to stabilize sulfates without producing significant swelling. This finding reinforces previous studies reporting the usefulness of binders other than lime or OPC in the treatment of sulfate-rich soils (Celik and Nalbantoglu, 2013; Kogbara and Al-Tabbaa, 2011; Puppala et al., 2003; Tasong et al., 1999; Wild and Tasong, 1999; Wild et al., 1998). Celik and Nalbantoglu (2013) found that soils containing sulfates at a concentration of about 10000 mg/kg of dry mass that were treated with only lime showed swelling of about 8% with respect to the initial volume of the sample. However, when GGBS was added, the swelling was reduced to 1%. Similarly, Wild and Tasong, (1999) showed a significant reduction in expansion when clays containing sulfates where stabilized by using 83% GGBS and 17% lime. They measured little expansion (< 2%) in the samples during the first 7 days of moist curing. Wild et al. (1999) explained that, in a GGBS-lime soil system, the proportion of GGBS influenced the magnitude of expansion. Higher GGBS content led to low expansions (< 5%) in sulfate-rich soils. The reduction of expansion in this type of system can be explained by the consumption of lime, which usually precipitate in the form of portlandite in clinker-rich binders, by the GGBS to be activated (Wild et al., 1999). In this case, GGBS reaction became the dominant reaction and the amount of lime is considered insufficient to enable the precipitation of large amounts of expansive phases (Wild et al., 1999). As mentioned previously, Wild et al. (1999) explained that, in a sulfate-rich soil treated with mixtures with low lime and high slag content, gypsum is consumed and no crystalline ettringite can

be formed. The authors of the present article think that the low hydration rate of the GGBS binder used in formulation F5 led to small volume changes over the 7 days of immersion test.

Treatment of sulfate-rich soils with GGBS binders appears to be useful for the stabilization of sulfates. However, there are several factors that require further investigation. The influence of leachant pH on the leachability of sulfates from sulfate-spiked soil is not determined by EN 12457-2 procedure. In order to understand the sulfate immobilization as a function of leachant pH other procedures could be used such as the method presented by the European standard EN 14429. On the other hand, leaching tests are usually applied to treated soils at short-term hydration (28 days). However, in the case of soils treated with GGBS binders, this curing can underestimate the capacity of such binders to stabilize sulfates due to the low rate of hydration. It is suggested that a higher immobilization of sulfates (or other pollutants) could be observed in soils treated with GGBS binders if leaching tests were carried out at long-term. Further investigation can be also addressed to the evaluation of long-term durability of GGBS-treatments as mentioned by Wild et al. (1999).

Finally, as mentioned previously, in this study, leaching tests were carried out in accordance with the European standard NF EN 12457-2 as required by French law (Legifrance, 2014). This leaching method is presented in the literature as a dynamic test that simulates the worst case of leaching behaviors, mainly because of the L/S (10 L/kg) and the particle size used (below 4 mm) (Vitkova et al., 2011). It should be noted that NF EN 12457-2 procedure does not require the characterization of the particle size below 4 mm. As reported in the literature, this condition led to discrepancies between several results from an identical waste source (Lackovic et al., 1997; Vitkova et al., 2011; Zandi et al., 2008). Zandi et al. (2008) explained that concentrations in eluates are underestimated when the dust fraction (<0.5 mm) is discarded from the leaching tests. Conversely, results are overestimated if only the finer fraction is considered because larger proportion of fine grain fraction leads to larger concentrations of elements in leachates. Therefore, further experimental investigations are needed to estimate the disparity between results coming from similar wastes by performing this leaching test. Characterization of particle size below 4 mm is highly recommended in order to increase the reliability of this leaching test.

5. Conclusions

Sulfate immobilization mechanisms were studied in four different formulations. Treatments of sulfate-spiked soil with ordinary Portland cement of type CEM I and with an alternative clinker mainly composed of ye'elimite and belite resulted in a high immobilization of sulfate (>80 %) in the solid. However, chromium (Cr) was released into solution in excess of the limit established by French law (0.5 mg/kg of dry mass of soil). Moreover, neither of the treatments was appropriate because volume expansions exceeded the guideline value (5%) established by French standard NF P 94-100. Large volume expansions (> 5%) in these treatments were explained by the precipitation of expansive

ettringite and because of delayed hydration due to the unavailability of water during the first 7 days of curing. On the other hand, sulfate-spiked soil treated with binders having a high GGBS content resulted in satisfactory immobilization of sulfates in the solid while avoiding the release of heavy metals into solution. In addition, GGBS-treatments decreased the swelling potential of the sulfate-spiked soil, which was demonstrated by measured volume expansions lower than 5%. Identification of sulfate-stabilization mechanisms in GGBS-treatment was a difficult task. However, mineralogical analyses enabled the identification of ettringite, which did not produce volume expansion in the treated soils. The precipitation of ettringite in the GGBS treatments was also verified by modeling calculations. The formation of ettringite was found to be one of the mechanisms controlling sulfate solubility. Further experimental investigations are needed to identify other possible mechanisms of sulfate stabilization in soils treated with GGBS.

The current work was restricted to a soil artificially contaminated with sulfate. Nevertheless, this system may be expected to behave in a similar manner to a natural sulfate-rich soil where the main source of sulfates is gypsum.

Acknowledgements

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