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Stabilization/solidification by hydraulic binders of metal elements from landfill leachate

Mira Cheribet Drouiche[1], Karim Moussaceb[1,✉], Emmanuel Joussein[2] and Jean-Claude Bollinger[2]

[1] *Laboratoire de Technologie des Matériaux et de Génie des Procédés, Faculté de Technologie, Université de Bejaia, 06000 Bejaia, Algérie*

[2] *Université de Limoges, PEREINE EA 7500 GRESE, Université de Limoges, 123 avenue Albert Thomas, 87060 Limoges, France*

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Abstract

The objective of this work is to use stabilization/solidification (S/S) on the landfill leachates that often are heavily polluted by heavy metals and require proper treatment before discharge into the environment. The process consists of a S/S using a hydraulic binder in order to limit the solubility and mobility of the pollutants. While cement is the most used binder based on S/S values, in this study we substituted it by cement kiln dust (CKD) in two replacement ratios 25.50 and 100 %. The resulting effect on mechanical resistance and on retention of pollutants was evaluated. A metal (lead, iron and zinc) contaminated leachate from the landfill site of Sidi-Bouderham in Algeria was mixed with an amount of cement and cement kiln dust in different proportions in order to optimize our formulations. The smooth paste was obtained and a standardized test of the test specimens was analyzed for mechanical resistance after 7 and 28 d of setting. Our results show that F1P (100 % Cement) and F2P (75 % Cement + 25 % CKD) point on satisfactory mechanical strength and metal retention capacity. Our approach suggests a promising approach for remediation of polluted sites.

Introduction

Environmental protection has an important place in the national and international priority concern. With the emergence of new consumption, habits of populations, human health and quality of natural environments are threatened by the increasing amount of solid waste generated. To cope with this situation, political decisions have been made over the world to reduce the volume of waste to be stored: it must be kept at a minimum.

In Algeria, this situation poses a real problem, as it is not dealt with correctly. For this reason, a new policy of integrated solid waste management has been implemented, however its translation on the field by a sustainable environmental infrastructure program for the sound management of different types of solid waste, is still difficult to achieve as it requires improved collection and waste treatment services.

In recent years, among the techniques used nowadays, solidification/stabilization (S/S) based on hydraulic binders offered multiple advantages. The S/S process stabilizes metals by sorption; lattice incorporation and precipitation during the cement hydration (Cheng and Bishop 1992; Glasser 1997; Conner and Hoeffner 1998; Malviya and Chaudhary 2006; Paria and Yuet 2006; Chen *et al.*

✉ *Corresponding author:* articlesmoussaceb@yahoo.fr

Table 1. Chemical analyses of Portland cement CEM I from Ain El Kebira (Algeria).

Element	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O
CEM I content [%]	23.35	6.10	6.77	59.40	1.89	1.76	0.43	0.12
CKD content [%]	11.81	4.50	2.28	43.48	1.13	0.74	0.50	0.00

2009). Cement matrix materials intended for immobilization vary considerably in their formulation. It is often economically and technically advantageous to add blending agents: e.g. slag and fly ash. These affect the chemical, mineralogical and microstructural constitution of the system (Glasser 1997). Several studies have been conducted by different researchers on S/S materials (Barna *et al.* 1997; Conner and Hoeffner 1998; Halim *et al.* 2003, 2004; Mijno *et al.* 2004, 2007; Qian *et al.* 2006; Cyr *et al.* 2012; Moussaceb *et al.* 2012, 2013; Belebchouche *et al.* 2014, 2016). The characterization of the mechanism of leaching is very important to predict the long-term behavior of S/S materials, currently, the main tests used for characterization of leaching are: Tank test and pH-dependence test, and a monolithic leaching test (MLT) to follow the long-term release of chemical species and pollutants (Tiruta-Barna *et al.* 2004; Barna *et al.* 2005).
This paper deals with the solidification/stabilization carried out on the leachate of the sludge from the landfill site of Sidi-Bouderham (Algeria). On leachate and S/S materials we performed a chemical analysis (metals, sulphate, phosphate and chloride). Our study contains several objectives: (1) The characterization of leachate from the landfill site to get an idea of the pollutant nature of this waste; (2) the treatment of polluted leachate in porous S/S matrices and the release of metallic species under different leaching conditions; (3) the study of the substitution effect of cement by CKD on the mechanical resistance and retention of heavy metals.

Experimental

Sample

The leachate used in this study comes from the percolation of water onto the waste sewage mud from the landfill site of Sidi-Bouderham in Algeria, classified as class II. This leachate is then used as a liquid source for the formulation of cement pastes. The formulations investigated are based on ordinary Portland cement (OPC) CEM I from the Ain El Kebira plant (Algeria) mixed with cement kiln dust (CKD) from the Hammam-Dalaa plant (Algeria). The chemical analyses of these OPC and CKD are shown in the Table 1. The formulations realized are based on the NF EN 196-1 standard (Afnor 2016). Briefly, specimens is prepared by mixing cement and CKD together for 10 min in a mixer of 3 kg capacity (Controls 65-L0005), in the way to obtain a homogeneous mixture. Then, leachate is added and is mixed for 5 min.
The ratio (Eq. 1):

$$R = \frac{L}{C + P} \quad (1)$$

(where L (mL) refers to the amount of leachate, C (g) and P (g) is the mass of cement and CKD, respectively) was fixed in 0.32 on the way to obtain adequate hydration for a good homogeneity and a satisfactory discharge capacity; the Table 2 gives the different formulations realized. Finally, the cement mixtures are molded to prepare samples with dimensions of 4×4×16 cm^3; three replicates were done for each. Then, the molds are protected by a plastic film and they are stored in a room under a controlled temperature of (20±2 °C) for 28 d.

Table 2. Formulation of each cement pastes realized.

Nomenclature	Cement [g]	CKD [g]	Leachate [mL]
F1P	450.0	0	145
F2P	337.5	114.5	145
F3P	225.0	225.0	145
F4P	0	450.0	145

Chemical analyses and mineralogical characterization

Chemical analyses of dried solids were performed by X-ray fluorescence (XRF - CubiX PANalytical).The infrared spectra of materials are performed using an FTIR spectrometer (Shimadzu IRAffinity-1S); sample preparation was done as KBr disks. For all experimental tests (equilibrium and dynamic leaching tests), leachate solutions were previously filtered at 0.45 µm, the solution was then separated into two parts: the first was acidified at pH 2 with HNO_3 solution (65 %) for cation analysis, and then stored in the cold; the second was used for the determination of anions and of the soluble fraction. Released metals (Pb, Zn and Fe) were analyzed using graphite furnace atomic absorption spectrophotometry (Shimadzu AA-6501F;cations (Na^+, K^+ and Ca^{2+}) and anions (SO_4^{2-} and PO_4^{3-}) were quantified by UV-visible spectroscopy (Biotech Engineering Management co. ltd. (uk)) (Rodier *et al.* 2009). All these analyses were performed in triplicates, and the reported values correspond to their arithmetic mean.

Mechanical strength

The measures of compressive and flexural strength are realized after 28 d of cure onto the 4×4×16 cm^3 monoliths according to NF EN196-1 standard (Afnor 2016), using a hydraulic press of type 65-L11M2.

Leaching test according to the Toxicity Characteristic Leaching Procedure (TCLP)

The solidified/stabilized materials were assessed using the toxicity characteristic leaching procedure (TCLP) as defined by U.S.EPA (U.S.E.P.A. 1994). The sample specimens were crushed to reduce their particle size to less than 2.0 mm. The procedure involved shaking a 1 g crushed sample in 200 mL of 0.0992 M acetic acid + 0.0643 M NaOH extraction solution (1/20 Solid/Liquid ratio) with a pH of 4.93±0.05, for 18 h on a rotary at about 300 rpm. The leachate was filtered through a 0.45 µm membrane filter to remove suspended solids and stored in the cold, then analyzed.

Acid neutralizing capacity (ANC) test

The purpose of this test is to determine the solubilization of chemical species as a function of pH, as well as the ability of the material to neutralize the acidic or basic solutions to which it is subjected (Tiruta-Barna *et al.* 2004; Barna *et al.* 2005). The material was fragmented at ≤ 1 mm and immersed over a period of time in solutions containing a strong acid or base. The test itself consisted in putting in parallel various samples of the material into contact with acid or basic solutions of different concentrations. The moisture of the material is taken into account to obtain an accurate ratio L/S = 10 $mL.g^{-1}$ of dry material. One of the leaching solutions was deionized water: it will allow us to obtain the natural pH of the material. The acidic solutions were prepared using nitric acid (1 M HNO_3) and the basic solutions with sodium hydroxide (1 M NaOH).
The flasks were submitted to a mechanical agitation during the time of the test at the rate of 10 turns/min. After 7 d the leachates were filtered using filter paper of 0.45 µm pore size and their pH and conductivity were measured. These leachates were then used for the determination of metals by the AAS method, after being acidified to pH 2 with 65 % HNO_3, and for the determination of anions and the soluble fraction.

Pores water (PW) and maximum mobile fraction (MMF) tests

This method consists in the extraction of water from the pores of the solid material. Assuming that we can extract all the water by a filtration installation, the test consists by mixing finely ground material with different volumes of deionized water (Tiruta-Barna *et al.* 2004; Barna *et al.* 2005). The material was crushed at a grain size ≤ 1 mm; solid samples were then immersed of deionized water at various L/S ratio (i.e. 200, 100, 50, 10, 5, 2 $mL.g^{-1}$ of dry material) and subjected to mechanical agitation for 7 d (10 twists/min) at room temperature (23±3°C). After that, the leachates were filtered using filter paper of 0.45 µm pore size and characterized.

Monolithic Leaching Test (MLT)

The leaching behavior of monolithic waste materials is part of their fundamental characterization tests. The MLT test is intended for characterizing the mass transfer mechanisms by observing the chemical elements flow released by the porous monolithic blocks when brought into contact with a fixed volume of leaching solution. The solution is renewed periodically and the dynamics of the release of some elements is determined by the physico-chemical analysis of the obtained leachate (Tiruta-Barna *et al.* 2004; Barna *et al.* 2005).

The specimens subjected to leaching were obtained by dry cutting of undried material, cleaned with compressed air, measured and weighed: their sizes were 4×4×4 cm^3. The leaching solution (either deionized water or 50 $g.L^{-1}$ sodium sulphate) was introduced at a ratio of liquid volume/block size (L/S) of 10 $cm^3.cm^{-2}$, what is enough to ensure dynamic behavior, in a hermetically closed container to prevent air penetration (carbonation from CO_2) and water evaporation during the experiment. For each material, all experiences were performed in duplicate. Each solution where the sample is immersed was changed after 6 h, 18 h, 1 day, 2 days, 5 days, 7 days, 20 days, 28 days, giving a total of 64 days of continuous leaching. Thus, we obtained 8 solutions whose physico-chemical parameters must be measured. At each renewal of the leaching solution, it was ensured that the time spent by the specimens out of the leaching was minimized. The leached solution was recovered after shaking the bottle and filtration to 0.45 µm, the filtered solid being put back into the flask for further leaching. Thus, a new volume of leaching liquid was added, the flask was closed, and the new leaching sequence began.

Results

Because Algerian regulations for waste disposal were recently available (Official Journal of the Algerian Democratic Republic 2006) we applied their limiting values for liquid effluent discharge to assess the hazard behavior of the studied S/S wastes.

Leachate characterization

The results of the physico-chemical characterization of leachate of the landfill site of Sidi-Bouderham are shown in Table 3.

Table 3. Physico-chemical characteristics obtained of leachate from of the landfill site of SIDI-BOUDERHAM used in this study.

Element	Value	Standards
pH	7.60	6.5 – 8.5
CE [$mS.cm^{-1}$]	1.00	–
SM [$mg.L^{-1}$]	168.00	3.0
COD [$mg\ O_2.L^{-1}$]	5.00	120.0 – 130.0
Pb^{2+} [$mg.L^{-1}$]	20.00	0.5
Zn^{2+} [$mg.L^{-1}$]	40.30	3.0
Fe^{2+} [$mg.L^{-1}$]	30.23	3.0
Na^{+} [$mg.L^{-1}$]	31.83	–
K^{+} [$mg.L^{-1}$]	55.00	–
Cl^{-} [$mg.L^{-1}$]	1,775.50	–
SO_4^{2-} [$mg.L^{-1}$]	1,024.00	–
PO_4^{3-} [$mg.L^{-1}$]	20.00	–

The conductivity of the leachate is approximately 1 $mS.cm^{-1}$, which indicates a significant presence of ionic species in solution. However, the presence of metals is effective such as Pb, Cu and Fe, but Zn is the present element (40.30 $mg.L^{-1}$) which exceeds the standard value (3 $mg.L^{-1}$). According to these results, the leachate can be considered as toxic and harmful; hence the need for treatment before storage.

Characterization of solidified/stabilized materials

Feasibility and chemical properties of S/S materials

The chemical analyses of synthesized S/S materials are reported in the Table 4. As expected, the chemical composition of the formulations is rich in SiO_2 and CaO. Moreover, the use of infrared spectroscopy (Fig. 1) allows to evidence the structural characteristics of C-S-H phases (hydrated

Table 4. Major elements of S/S materials for each formulations.

Element	SiO_2 [%]	Al_2O_3 [%]	Fe_2O_3 [%]	CaO [%]	MgO [%]	SO_3 [%]	K_2O [%]	Na_2O [%]	Others [%]
F1P	21.36	5.32	6.46	56.55	1.40	1.53	0.41	0.12	6.85
F2P	21.32	4.79	5.39	54.40	1.70	1.31	0.44	0.11	11.17
F3P	17.80	4.84	4.53	51.68	0.97	1.10	0.49	0.11	13.20

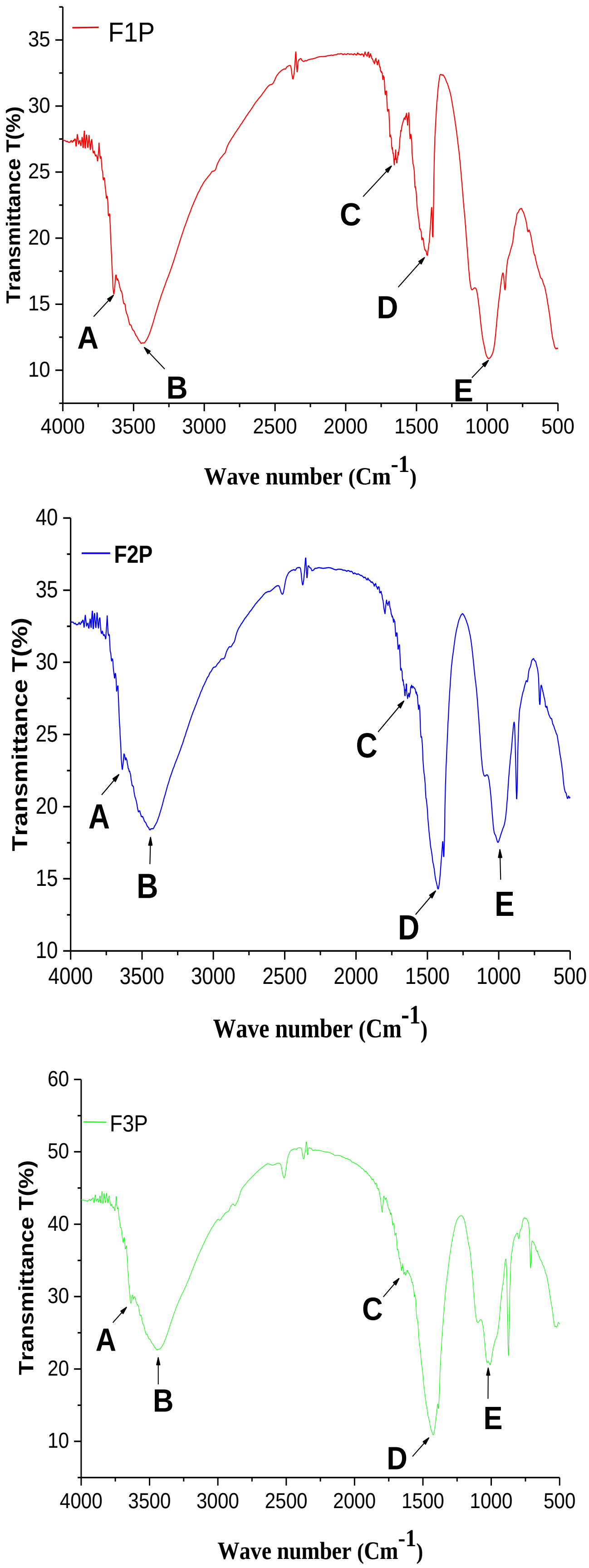

Fig. 1. Infrared spectra of materials F1P, F2P and F3P, and identification of the bands observed on the IR spectrums.

phases) by the presence of the Si-O-Ca bands near 990 cm^{-1} as well as formation of portlandite (characteristic band at 3,639 cm^{-1}), carbonates (band at 1,420 cm^{-1}) and ettringite (harmonics around 2,930 cm^{-1} and 2,853 cm^{-1} signs of the presence of clinker) during hydration period. This fact clearly explains the consolidation of the S/S materials obtained by the alkaline activation process.

Mechanical strength

Compressive and flexural strengths of solidified/stabilized materials were followed for 7 and 28 d of cure (Fig. 2). In general, materials at 28 d of curing showed higher strengths than materials at 7 d of curing.

This observation is common in studies and experiments dealing with concrete or cement strength reported in literature (Atahan *et al.* 2009; Zhao *et al.* 2013; Belebchouche *et al.* 2014). The increase of CEM 1 percentage in the formulation induces the increase of the mechanical strength (compressive and flexural) from 30 to 70 MPa in the case of the compressive one. As expected, the curing time favors such properties, which agree with the hydration process (Bediako *et al.* 2015). The observed increased compressive strength results in this study might be due to the organic content present in the Leachate, which might act as a dispersing agent, improving the dispersion of particles of cement and reducing clumping (Silva and Naik 2010). The effect of concrete mixing water has been widely investigated for different water sources other than ordinary freshwater. Silva and Naik (2010) investigated the use of partially processed sewage treatment plant water in concrete and found improvement in strength during 3 to 28 d. Taha *et al.* (2010) have studied the use of brackish and crude produced water and found better strength even at the long curing period compared to tap water. Katano *et al.* (2013) found that early and long-term strength for concrete mixed with high chloride content ingredients (seawater and sea sand) could be improved. Al-Jabri *et al.* (2011) reported no significant difference in strength for concrete mixed with car washing station waste water.

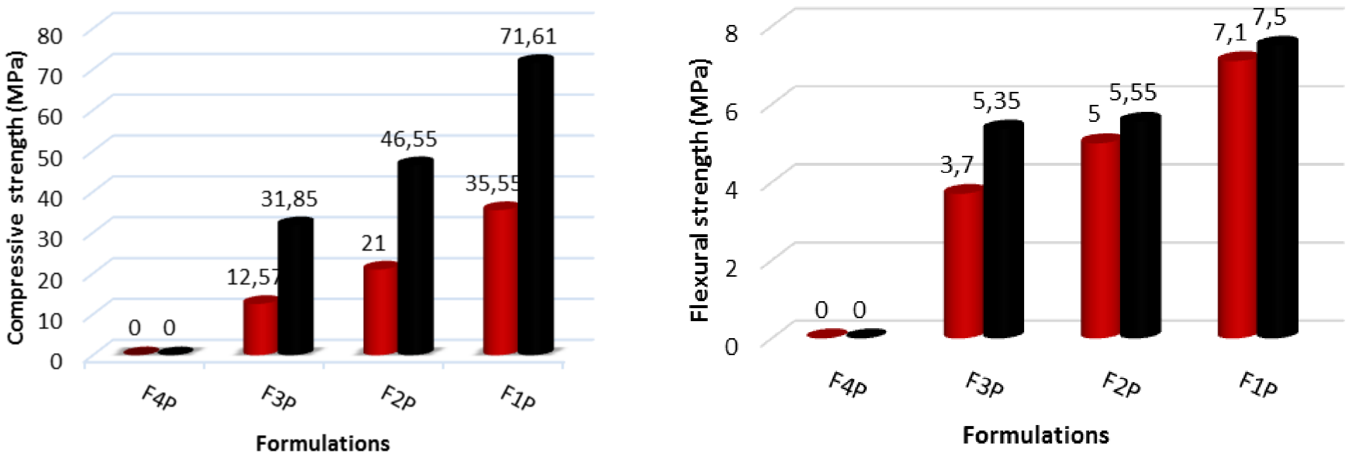

Fig. 2. Evolution of the mechanical strengths of solidified/stabilized formulations of leachates as a function of cement amount at 7 (red columns) and 28 (black columns) days.

Table 5. Element concentrations after the TCLP test of S/S materials.

Element	Concentration of leached elements [mg.L^{-1}]				Percentage of release [%]			
	Pb^{2+}	Zn^{2+}	Fe^{2+}	Cl^-	Pb^{2+}	Zn^{2+}	Fe^{2+}	Cl^-
F1P	0.25	0.10	0.16	15.79	1.25	0.24	0.52	0.88
F2P	0.42	0.11	0.33	19.80	2.10	0.27	1.90	1.11
F3P	0.87	0.20	0.38	20.60	4.35	0.49	1.25	1.12

The addition of CKD in the formulation is not satisfactory as their influence on these mechanical properties is deleterious. Several studies have been carried out to study the effect of cement substitution by CKD, Bhatty (1984, 1985, 1986) studied binary, ternary and quaternary mixtures using ordinary Portland cement (OPC), five different CKDs, two different types of fly ash (classes F and C) and slag. He observed that cements containing only CKD had reduced strength, set time and handling. The addition of fly ash to a CKD-OPC system has reduced alkali content and improved strength.

Maslehuddin *et al.* (2009) evaluated the properties of cement kiln dust (CKD) blended cement concretes. The percentages of CKD were 0 %, 5 %, 10 % and 15 %, replacing cement ASTM C 150 Type I and Type V. The results showed that the compressive strength of concrete specimens decreased with the quantity of CKD. However, there was no significant difference in the compressive strength of 0 and 5 % CKD cement concretes.

According to the data in Fig. 1 it can be deduced that the desired strength can be obtained with 100 % of CEM (F1P S/S formulation).

Leaching behavior of S/S materials according to TCLP test

To check the immobilization behavior of the solid S/S matrix against the contaminants and to determine the most efficient formulation, TCLP test is performed and results are reported in the Table 5. The concentrations of these elements are lower than those found on the leachate before treatment. The higher the cement dosage percentage, the lower are the concentrations. Whatever the formulation of the S/S materials, the leaching values from TCLP are under the regulated standards reported in Table 3.

Transfer and leaching properties of solidified/stabilized materials

According to the preceding data, it was decided to select the F1P and F2P S/S formulations (100 % and 75 % CEM, respectively) for the next tests.

Results of Acid neutralizing capacity (ANC) test

The solubilization of the chemical species contained in the F1P and F2P materials, depending

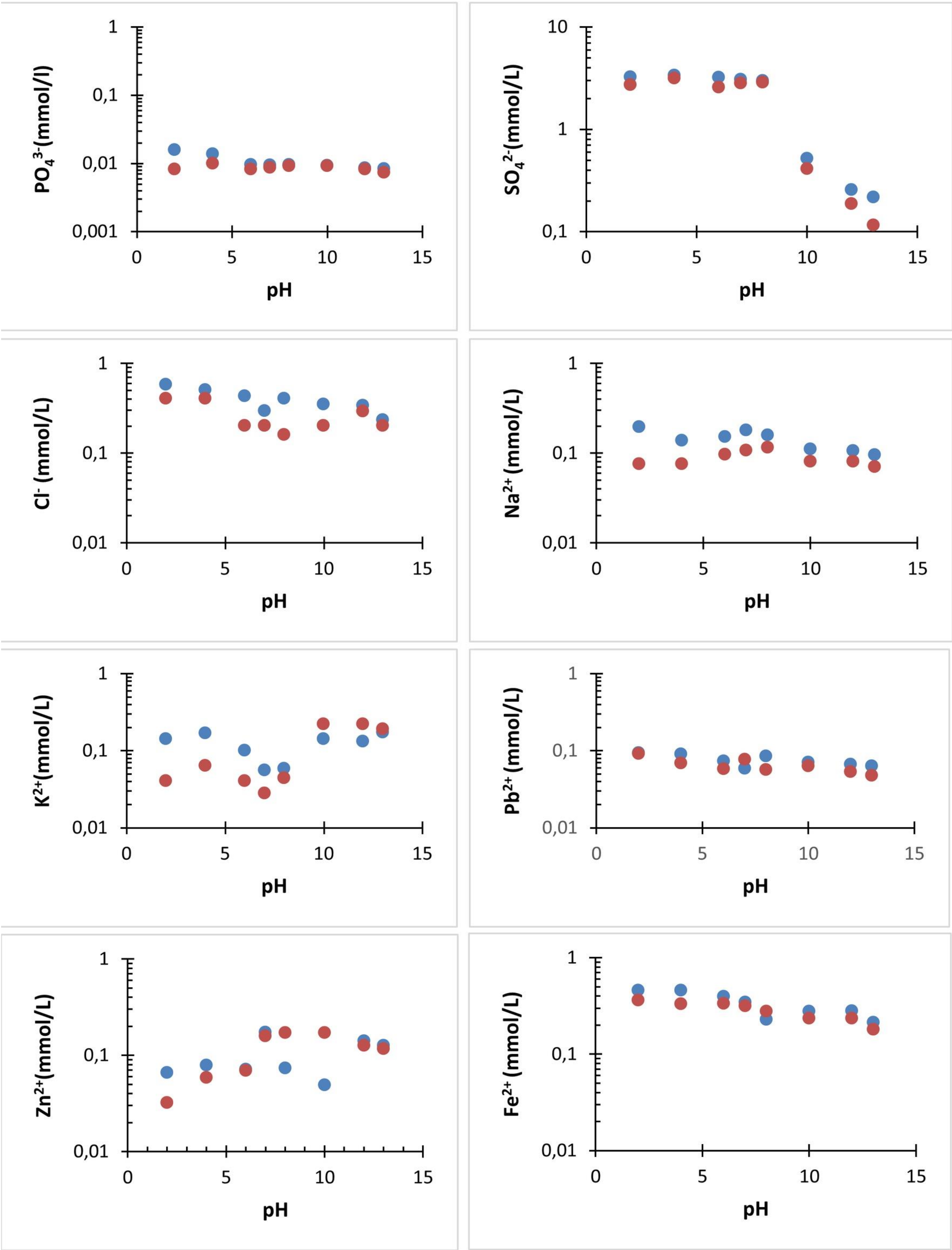

Fig. 3. Solubility of chemical species in mmol.L^{-1} as a function of pH in the case of F1P (red circles) and F2P (blue circles) materials after ANC test.

on the pH, is shown in Figs. 3 a, b, c, d, e, f, g, h. The species can be classified into two categories: pH-independent species (phosphate) and pH-dependent species i.e. chloride, calcium, lead, chloride, sodium, potassium and sulphate.

In the case of the pH-independent species, the solubilization of phosphates at the start of the acid attack, increases gradually (pH shift from 13 to 10) until it reaches a steady state concentration level.

For the pH-dependent species, the amount of solubilized chloride is almost stable at the beginning of the acid attack (pH value shifts from 13 to 6) and an increase of dissolved chloride is observed when the pH value falls under 6. The maximum amount of Cl^- extracted is obtained for pH < 4. This can be attributed to the sorption of the chlorides within the C-S-H phases. Indeed, the behavior of chloride ions may be considered as almost independent of pH. The Same behavior is obtained for the sulphate that is slightly dissolved at the beginning of the acid attack (pH between 13 and 10). When the pH decreases, solubilization increases and becomes maximum when pH < 8.

For Zn, there is an increase of the solubilization at the beginning when pH > 10. A maximum of mobilization is achieved for 6 < pH < 8, and finally a decrease is observed for highly acidic pH this result is consistent with work of Imyim (2000).

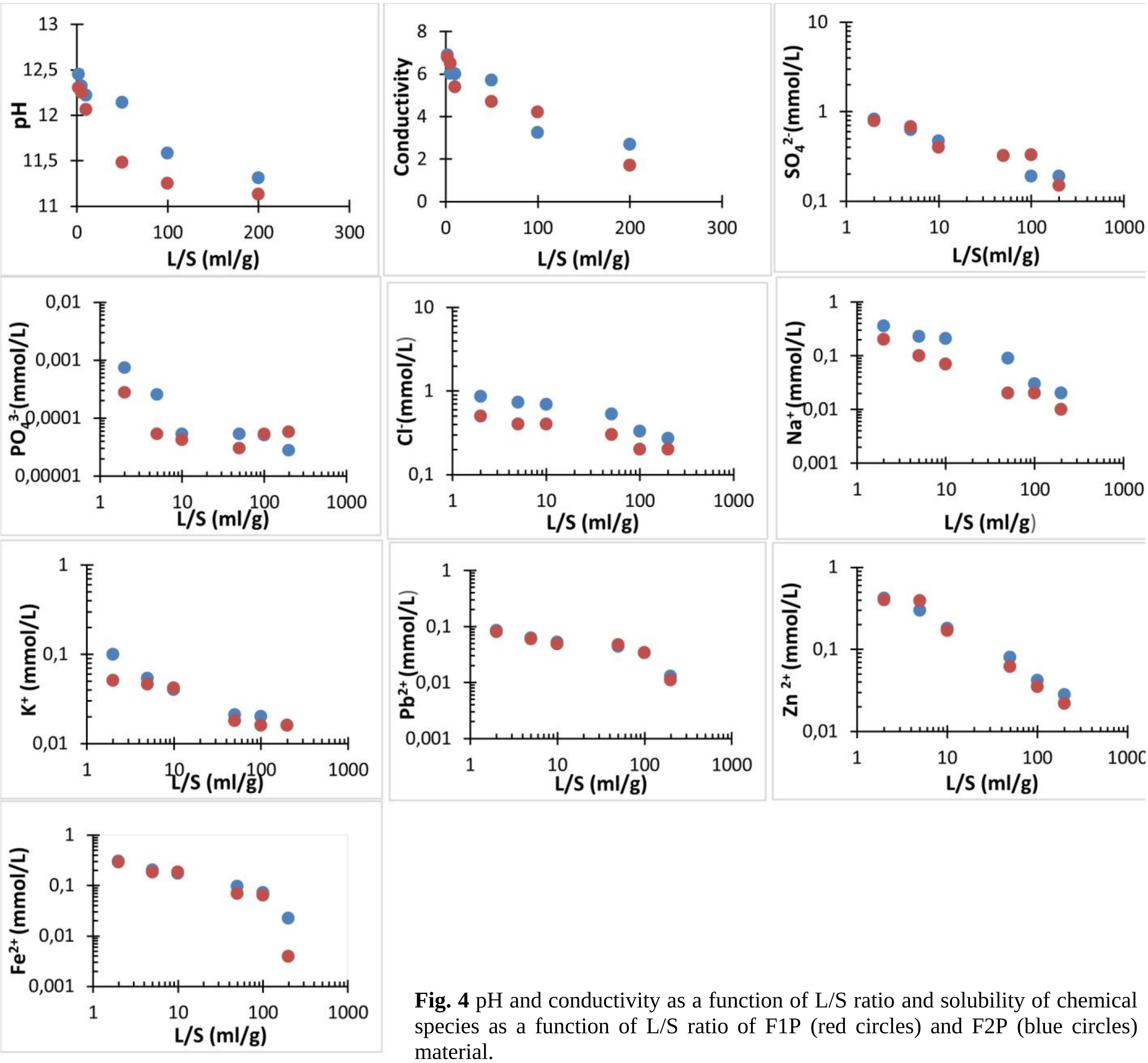

Fig. 4 pH and conductivity as a function of L/S ratio and solubility of chemical species as a function of L/S ratio of F1P (red circles) and F2P (blue circles) material.

For Pb and Fe, the solubilization regularly increases with the acidification of the medium. Concerning K, the solubilization decreases from pH less than 9 to reach a steady-state value, the amphoteric behavior of lead accounts for the solubility minimum found between pH 8 and 10 combined with sharp solubility increases in both acidic and alkaline conditions (De Windt *et al.* 2007). Catherine (2003) found a slight increase in potassium solubilization for very alkaline (greater than 11) and acidic (less than 6). For Na, it seems that the maximum values of solubilization is obtained for pH between 6 to 10 according to Catherine (2003), sodium and potassium concentrations are independent of pH.

Pores water (PW) and maximum mobile fraction (MMF) tests

The results showing the comparative evolution of pH , conductivity (Fig. 4a) and the concentration of the monitored species depending on the variation of the ratio L/S for the material F1P and F2P during PW and MMF testing are recorded in the Figs. 4b, c, d, e, f, j, h, i.

The general evolution of the pH is an increase with decreasing L/S. The initial level of basicity is 12.3. The change in conductivity exhibits the same behavior as the pH. In fact, conductivity reflects the ionic strength of the solution then the solution of the pores of cement-based material has a somewhat high ionic strength.

The maximum extracted quantities are recorded for low L/S ratios (Imyim 2000; Belebchouche *et al.* 2014). Briefly, Zn, Fe and Pb seem to have a solubility in accordance with the change in pH according to changes in L/S. The dependence of the concentration of sodium and potassium on the L/S ratio (linear with the L/S ratio in logarithmic scale) is explained by the fact that these elements come from the dissolution of highly soluble phases (Imyim 2000). The chlorides behavior is like that of Na with regular decreases when S/L increases.

Monolithic Leaching Test (MLT)

The results of the physical parameters (pH and conductivity) of MLT tests are shown in Fig. 5a and the evolution of the concentration of monitored species are shown in Fig. 5b, c, d, e, f, g, h.

Whatever of the medium used (neutral or sulphated), the shape of curves is similar. The pH is always highly basic comprised between 11 and 12, we also observe that the pH increases over time, despite the renewal of leachates through the dissolution and diffusion of the soluble species contained in the material. The conductivity is constant along time of experiment. Whatever, the elements studied, the flow decreases with time. The contents of the monitored species released into the sulphate medium are higher than those in the neutral medium; this is due to the higher ionic strength of the sulphated solution.

Discussion

Although the municipal landfill at Sidi-Bouderham (Algeria) is dedicated to domestic household waste, it is well known that such a facility surely contains several kinds of contaminants, mainly potentially toxic metals and xenobiotic organic compounds (Slack *et al.* 2005). Among the possible treatments to immobilize such hazardous chemical species, S/S within cementitious matrixes can be a good choice; this method has been intensively applied to sewage sludge (Valls and Vazquez 2000; Cyr *et al.* 2012).

Several studies have shown that the use of different ingredients added with cement, such as clays, zeolite, and fly ashes, allowed enhancing the performance of the S/S treatment (Barth 1990; Montgomery *et al.* 1991; Adaska *et al.* 1998; Balkan and Kocasoy 2004; Shi 2004; Hunce *et al.* 2012). The studied leachate mainly contains 20 $mg.L^{-1}$ Pb(II) and 1,775.50 $mg.L^{-1}$ Cl(-I) species, but also other potentially toxic metallic elements such as Zn(II) and Cu(II) and SO_4^{2-}. Although the addition of fly ash to cement commonly improves both the mechanical and physico-chemical properties of such S/S materials (Cyr *et al.* 2012), this is not the case in the present study with the addition of cement kiln dust CKD to ordinary Portland cement OPC. The TCLP test on the S/S solid materials allowed to highlight the efficiency of 100 % OPC addition (F1P sample) but the F2P sample with 25 % CKD can be another

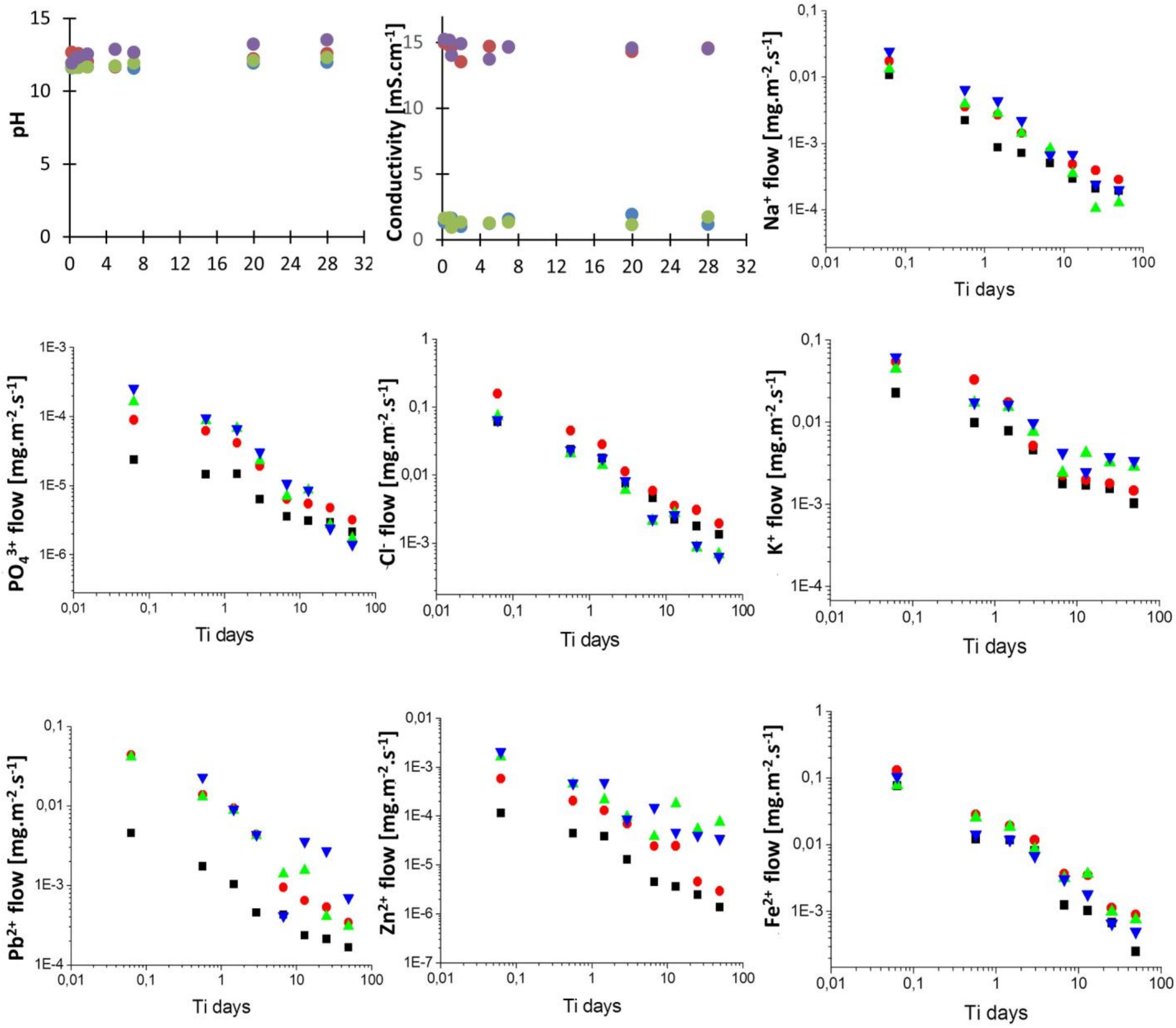

Fig. 5. Evolution of pH and conductivity as a function of time in neutral and sulphated medium, and concentration of the element flows as a function of average time (axis x; Days) for the monolithic leaching test (MLT). Indicated are data for neutral medium F1P (blue), Sulphated medium F1P (red), Neutral medium F2P (green), Sulphated medium F2P (lila).

good choice in spite of lower mechanical strength. According to the leaching test results, main pollutants in the leachate, Pb, Zn and Fe were successfully solidified and approximately, 1.25 %, 0.24 % and 0.52 % of Pb, Zn and Fe respectively released in the eluate water.

For a somewhat similar case, the cementitious S/S of landfill leachate concentrate obtained after reverse osmosis has only attracted a scarce number of studies. Hunce *et al.* (2012) added zeolite to cement, but the result was not good, they conclude that the use of zeolite as an aggregate material did not show significant differences. In the case of a Zn-rich concentrate, Kallel *et al.* (2017) have made use of clay brick waste as a substitute to coarse aggregate material in the composition of concrete mixtures; all metals but Cr were effectively immobilized in such matrixes.

Conclusions

Following this study, the "S/S" stabilization/solidification method using hydraulic binders comprising artificial Portland cement seems quite capable of treating contaminated leachates. It can retain metals pollutants in the leachate of the landfill. Applying a series of leaching tests helps determine the leaching behavior of materials and pollutants, the effect of pH of the leachate was investigated by the NAC test. EP tests and DMF provide quantities released by water leaching of the highly soluble elements such as Na, K, and Cl. The flow species released by the material in the MLT test inform about the transport mechanisms. The exploitation of the results allowed us to draw the following conclusions:

The values of the mechanical strength of the S/S materials obtained are high. However, the resistance decreases with the increase of the amount of the substituted CKD, for the formulation F4P (100 % CKD) the resistance is lower than that recommended by the standard XP X 31-212, so it is not satisfactory for the manufacture of test pieces.

It turns out that the ratio R = 0.32 is interesting. The latter has given satisfactory results by retaining almost completely the heavy metals from where the concentrations of the analyzed elements in the leachate of materials S/S return effectively within the range of Algerian standards. The retention of heavy metals decreases with the increase in the amount of CKD whose release percentages of Pb, Zn and Fe are respectively 1.25, 0.24 and 0.52 for F1P and 2.1, 0.27 and 1.09 for the formulation F2P.

Conflict of Interest

The authors declare that they have no conflict of interest.

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