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ORIGINAL PAPER

Comparison of the effectiveness of different binders in solidification/stabilization of a contaminated soil

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Abstract

In the current research, solidification/stabilization (S/S) treatment of the contaminated soil using hydraulic binders and additives was used to (1) reduce the mobility of organic and inorganic contaminants and (2) compare the ability of various binders in fixing contaminants. The samples were collected from Franco-Tunisian Petroleum Company, located in Sidi Litayem, Sfax (Southern Tunisia). Leaching tests were performed on contaminated soil, containing metallic elements, and hydrocarbons. Calcium aluminate cement (CAC), ordinary Portland cement (OPC), and ground-granulated blast-furnace slag (GGBFS), additives especially the bentonite and water, were used for S/S treatment. The obtained standard specimens were subjected for treating after treatment the leachability of pollutants, compressive strength (CS), and XRD analysis. The results of analysis conducted on contaminated soils showed that concentrations of metallic elements were in the range of 9.08–427 mg/kg and 15,520 mg/kg of organic compound. Next, 10% of the used binder improved the immobilization of pollutants and gave a satisfactory CS exceeding 1 MPa. Thus, the CAC is more effective in reducing the leachability of metal contaminants than OPC + GGBFS and produces much higher strength, which was of the order of 2.41 MPa. The mechanical characterization was confirmed by XRD analysis. The lowest values of organic compounds are presented in mixtures treated by 10% of used binder, indicating the effectiveness of those with the presence of 10% of bentonite. This work shows that 10% (OPC + GGBFS) + 10% bentonite improved the immobilization of metallic elements and hydrocarbons, thus proving its efficiency due to its low cost.

Keywords Contaminated soil · Solidification/stabilization · CAC · GGBFS · Compressive strength · Leaching tests

Introduction

Nowadays, with the inevitable rise in population and industrialization, the continuous production and consumption of energy has led to large quantities of waste. The latter is growing in volume and in toxicity, and the problem will be further aggravated. Increasingly, our everyday waste production consists mainly of organic constituents, such as hydrocarbons and toxic chemical elements, namely alkali and metallic elements and these toxic products are combined with a plethora of other chemicals, which eventually impact public human health and environment quality (Qian et al. 2006). It has been stated that a

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perfect sustainable society should not generate waste exceeding its own capacity of waste treatment (Stren et al. 1992). So, the question is: how can we change the way we consume so as to produce less waste, while using all waste as a resource? The sustainable management of waste is, therefore, and above all, the responsibility of states and territory governments, which manage waste in accordance with their respective legislation, policies, and programs.

Different waste treatment technologies such as remediation technologies, containment, thermal treatments, physicochemical treatments, solidification/stabilization (S/S) treatment, and recycling and composting (Ji et al. 2004) were developed. Not only is S/S treatment using hydraulic binders the most promising (Al-Ansary and Al -Tabbaa 2007), but also a remedial technology that has attracted widespread use around the world. Besides, S/S processes have emerged as an efficient method for the treatment of wastes polluted with potentially toxic metals due to its versatility, efficiency (Falciglia et al. 2012).

This treatment involves mixing hydraulic binders into the waste to transform it into a solid material with stable pH, low mobility, and leachability of pollutants (Zhou et al., 2006). According to Van der Sloot et al. (2007), this will create the minimum threat to the environment by transforming hazardous wastes into non-hazardous or less-hazardous wastes.

Previous research works have found that the S/S treatment is a reliable and an appropriate technology for the treatment of contaminated soils (Scanferla et al. 2009; Voglar and Lestan 2011; Jin et al. 2016). It is a commonly used land remediation method that aims to improve the mechanical properties and the leaching resistance of the contaminated soil (Jin et al. 2016). The range of operating conditions that result in acceptable treatment of some commonly used binders has been investigated. It was shown that about 20% binder dosage can satisfy most leaching criteria (Kogbara et al., 2014).

The US Environment Protection Agency (EPA) has recognized that the use of ordinary Portland cement (OPC) as a binder is the best demonstrated available technology. It has also been reported that the OPC is the most widely used binder in the S/S technique (Vandeperre et al. 2008). According to Gu et al. 2015, the treatment of contaminated soil using the OPC is widely used in road, rail, and airport construction. Navarro et al. (2011) have examined the effectiveness of this cement in contaminated soil by using leaching experiments and geochemical modeling. The viability of the cement-based S/S technology was evaluated on 40 site representative soil samples from Cinkarma brownfield using OPC (Volgar and Lestan 2010). The introduction of OPC and cement kiln dust as hydraulic binders has improved the properties of arsenic-contaminated soils especially strength, durability, permeability, and volume stability (Tariq and Yanful, 2013). Furthermore, Barth et al. (1989) have revealed that using OPC in the S/S of metallic elements containing waste is effective in reducing the leaching of these elements. Laforest and Duchesne (2005) have used the ground-granulated blast-furnace slag (GGBFS) and OPC to evaluate the retention of hexavalent Cr.

These authors proved that GGBFS was more efficient than OPC in fixing Cr at lower concentrations. However, with concentrations above 2000 mg/l, OPC was more efficient. Jin et al. (2016) demonstrated that although the treatment of the in situ solidified/stabilized soil using novel MgO-bearing binders alone provided negligible strength to the soil, it improved the immobilization capacities on both organic and inorganic contaminants. Furthermore, the substitution of MgO by 90% of GGBFS increased the strength and decreased the permeability remarkably (Wang et al. 2015). Special cements such as calcium aluminate cement (CAC) (Conner 1990; LaGrega et al. 1994) have also been studied. The CAC pastes containing Pb, Cu, and Zn and the leaching tests presenting 99.9% retention were studied by Navarro-Blasco et al. (2013). Regarding the immobilization of Cr, the lower pH and the higher content of Fe present in CAC could improve its ability to immobilize Cr^{6+} to Cr^{3+} since Cr^{6+} is more soluble than Cr^{3+} (Navarro-Blasco et al. 2013).

In the current research work, OPC was selected as the most used cement to investigate the leaching properties and the unconfined compressive strength of the solidified/stabilized soil. GGBFS was used both to improve the mechanical behavior of the soil, including the unconfined compressive strength in combination with the cement and reduce the environmental impact of OPC (Wang et al. 2015). Al Ansary and Al Tabbaa (2007) showed that the combination of 20% of GGBFS-OPC and 30% of lime-OPC with low oil content led to the immobilization of the pollutants and the reduction in oil concentration. CAC was applied in view of its rapid hardening. Bentonite was used as the soil binder, additive desiccant due to its absorption properties and a degreasing agent (Carmody et al. 2007). Economically, it was used to optimize the amount of hydraulic binders and water in a cementing matrix.

The main objectives are (1) comparing the ability of various binders in the fixation of contaminants and (2) assessing their effects on the properties of S/S materials and curing conditions.

Material and methods

Contaminated soil sampling

A sampling campaign of contaminated soil was conducted directly from the oil site of the Franco-Tunisian oil company located in Sidi Litayem (SIT 11) of the region of Sfax (Southern Tunisia) (34° 53′ 13.82″ N 10° 31′ 52.41″ E) (Fig. 1). Outdoor contaminated soil samples were taken from the surface (0–30 cm), during the period extending from

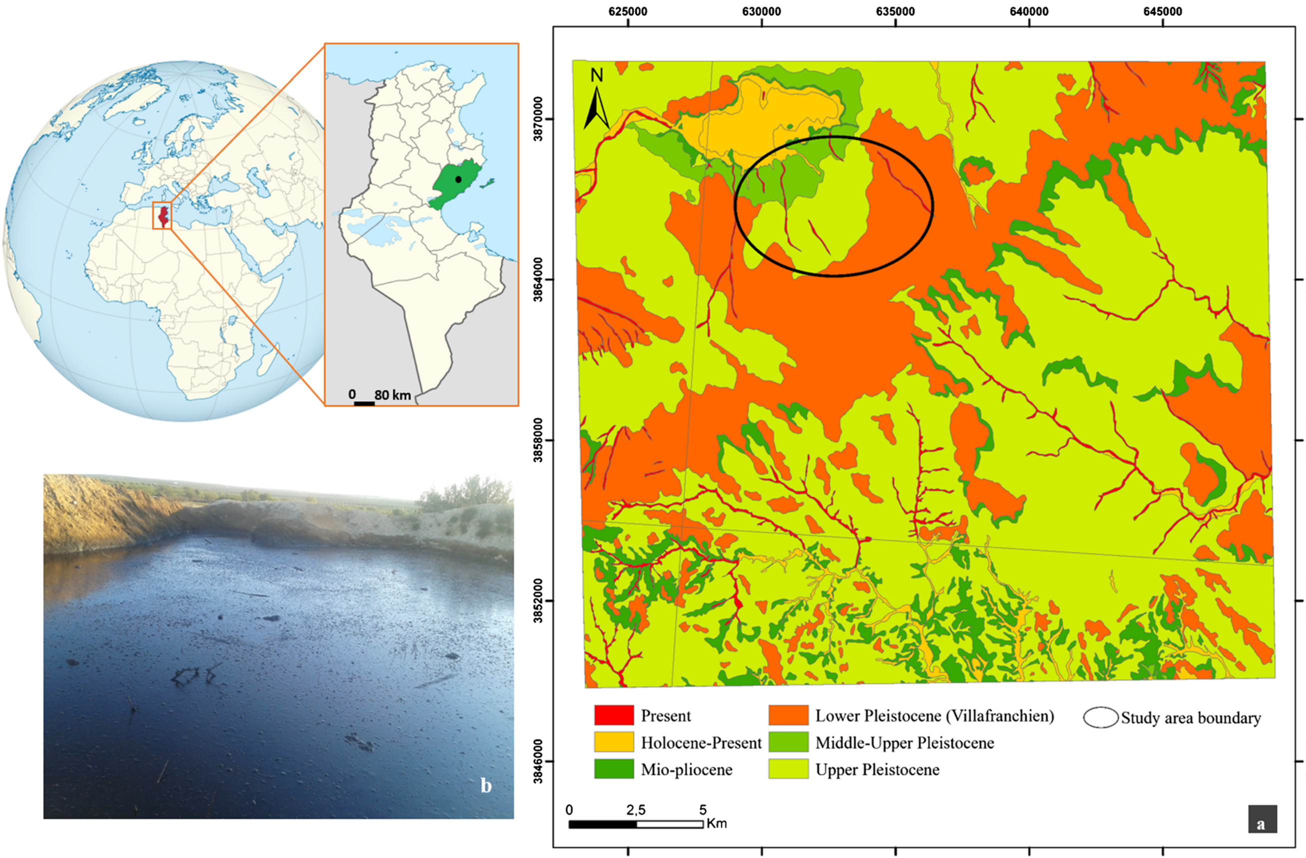

Fig. 1 **a** Geological map of Tunisia with a zoom of the study area. **b** Overview of contaminated site

December 2015 to January 2016. These samples, containing metallic elements and salts, and rich in organic constituents such as petroleum hydrocarbons, have a dark brown color. They were uniformly mixed before further experimental work. The studied soil before treatment is sand-colored clayey (S0). Sampling was carried out using a manual coring of cylindrical specimens (diameter = 4 cm; height = 8 cm) according to the AFNOR NF X31-210 French standard relative to the characterization of ultimate industrial stabilized water (Recueil 1994). In fact, sampling was conducted in control soil named (SIT11-2) (34° 53′ 15.61″ N 10° 31′ 49.16″ E) and polluted soils (SIT11-1: P1, P2, P3, and P4) (34° 53′ 15.35″ N 10° 31′ 51.34″ E) (Fig. 1). Then, contaminated soil samples were air-dried at ambient temperature and sieved at 2 mm, for the

Table 1 Testing protocol for each mixture type

Mixing binders	Soil (%)	Cement/bentonite (%)	Water (%)	Tween 80 (%)
M1: CAC	73	5:0	22	0.025
M2: CAC	68	10:0	22	0.05
M3: CAC + bentonite	68	5:5	22	0.05
M4: CAC + bentonite	63	10:5	22	0.075
M5: CAC + bentonite	63	5:10	22	0.075
M6: CAC + bentonite	58	10:10	22	0.1
M7: 60% OPC + 40% GGBFS	73	5:0	22	0.025
M8: 60% OPC + 40% GGBFS	68	10:0	22	0.05
M9: 60% OPC + 40% GGBFS + bentonite	68	5:5	22	0.05
M10: 60% OPC + 40% GGBFS + bentonite	63	10:5	22	0.075
M11: 60% OPC + 40% GGBFS + bentonite	63	5:10	22	0.075
M12: 60% OPC + 40% GGBFS + bentonite	58	10:10	22	0.1

CAC calcium aluminate cement, *OPC* ordinary Portland cement, *GGBFS* ground granulated blast furnace slag

Table 2 Metallic elements and organic compound contained in contaminated soil

Element		Cr	Cu	Pb	Zn	Cd	Ni
Contaminated soil	Concentration in soil (mg/kg)	210.79	128	96.7	427.59	9.08	206.8
	Aqueous concentration in the leachate (mg/l)	0.6	0.792	0.335	1.52	0.1	0.402
(N.T106-002)		0.50	0.50	0.10	5	0.04	0.20
pH		8.26					
Organic compound (mg/kg)		15,520					

*N.T106-002: Tunisian standard of wastewater discharge

analysis of chemical and physical properties in the laboratory of Research unit in Geotechnical Environmental and Civil Materials.

Contaminated soil characterization

For the analyses of the total copper (Cu), cadmium (Cd), chromium (Cr), zinc (Zn), nickel (Ni), and lead (Pb) concentrations in contaminated soil samples, about 2 g of the dried soil were digested in a mixture of concentrated acids (nitric acid, fluorhydric acid, and perchloric acid) according to the EPA 3052 guideline (EPA 1996). The digested samples were then diluted to 50 ml with double distilled water, and filtered through Whatman filter paper into acid-washed polyethylene sample bottles. After filtration, the samples were determined for Cu, Cd, Cr, Zn, Ni, and Pb using an atomic absorption spectroscopy AAS with the air-acetylene flame (Analytik Jena model ZEEnit 700PC). The detection limits (mg/l) were 0.015 for Cd, 0.01 for Pb, 0.002 for Zn, 0.001 for Ni, 0.004 for Cu, and 0.01 for Cr.

Leaching tests were performed on contaminated soil, containing metallic elements, and hydrocarbons, according to the AFNOR NF X31-210 French standards. The leaching test was determined by putting in contact about 50 g of the contaminated soil sample and 500 ml of distilled water, during 24 h of continuous agitation. Non-ionic detergent polyoxyethylene 20 sorbitan monooleate Tween 80 was chosen as an additive (Voglar and Lestan 2011) to make the organic matter containing in the soil more hydrophilic (Table 1). The solution was separated from the solid residual fraction by filtration through 0.45-μm filter. The filtrated leachates were then subjected to chemical analysis of different parameters in the laboratories of the Faculty of Sciences of Sfax (Tunisia) and the Olive Tree Institute of Sfax (Tunisia). Metallic elements Cu, Cd, Cr, Zn, Ni, and Pb were measured in triplicate using an atomic absorption spectrometry. The contaminated soil pH was measured in a soil/distilled water ratio of 1/2.5 by the electrometric method using a pH meter equipped with a glass electrode (Louati et al. 2017). Soxhlet extraction apparatus was used for the extraction of organic compounds from the contaminated soil (Kumar and Kothiyal 2011).

The mineralogical analysis of prepared mixtures is determined by X-ray diffraction. The estimate of the percentage of each mineral is affected by the integration of the area of the most intense peak (Mosbahi et al. 2014; Mosbahi et al. 2017).

All XRD data were collected under the same experimental conditions using an X'pert HighScore plus PANalytical diffractometer. Philips PW type diffractometer PANalytical X Pert Pro MPD (θ–2θ) system, equipped with a copper anticathode, was used CuKα radiation source under 40 kV/40 mA and an angular range of 3–70° 2θ for bulk rocks and 3–30° 2θ for fine fraction, a step size of 0.017° 2θ, and a counting time of 10 s/step to determine mineral composition.

Tested formulations

The treated soil specimens were prepared for various formulations of hydraulic binders and additives, according to the AFNOR NF X31-211 French standard (diameter = 4 cm; height = 8 cm) (Table 1). Indeed, hydraulic binders, additives, contaminated soil sample, and water were put into the container and mixed for 4 mn, using magnetic mixer in order to achieve a uniform admixture (Wang et al. 2014). The obtained smooth paste was subjected to compaction tests using standards cylindrical tube models with an inside diameter of 4 cm and a height of 8 cm for the

Table 3 pH variation in treated soil leachates

pH	Mixture											
	M1	M2	M3	M4	M5	M6	M7	M8	M9	M10	M11	M12
28 curing days	10	10.9	9.8	9.4	8.8	10.5	8.6	9.8	8.6	9.5	8.9	10.1
60 curing days	9.8	11.1	8.7	8.9	8.6	11	8.7	10.4	8.6	9.8	8.8	10.3
90 curing days	9.9	11.3	8.6	9	8.7	11.2	8.7	10.9	8.7	10	8.8	10.4

Table 4 Concentration of leached metals in treated soil

Element (mg/l)	Day	M1	M2	M3	M4	M5	M6	M7	M8	M9	M10	M11	M12
Cr	28	0.310 ± 0.02	0.249 ± 0.00	0.391 ± 0.00	0.340 ± 0.01	0.253 ± 0.00	0.150 ± 0.00	0.277 ± 0.00	0.150 ± 0.00	0.360 ± 0.00	0.182 ± 0.01	0.280 ± 0.01	0.150 ± 0.01
	60	0.190 ± 0.00	0.127 ± 0.01	0.335 ± 0.00	0.310 ± 0.00	0.219 ± 0.01	ND	0.282 ± 0.00	0.147 ± 0.01	0.381 ± 0.01	0.162 ± 0.01	0.291 ± 0.01	0.125 ± 0.04
	90	0.182 ± 0.00	0.135 ± 0.01	0.311 ± 0.00	0.250 ± 0.05	0.175 ± 0.01	ND	0.261 ± 0.00	0.119 ± 0.00	0.355 ± 0.01	0.175 ± 0.01	0.225 ± 0.01	0.118 ± 0.09
Cu	28	0.299 ± 0.01	ND	0.374 ±0.00	0.271 ± 0.00	0.305 ± 0.01	ND	0.320 ± 0.01	0.279 ± 0.01	0.273 ± 0.01	0.089 ± 0.01	0.213 ± 0.00	0.120 ± 0.00
	60	0.273 ± 0.01	ND	0.315 ±0.00	0.258 ± 0.01	0.256 ± 0.01	ND	0.325 ± 0.01	0.221 ± 0.00	0.264 ± 0.01	0.098 ± 0.00	0.299 ± 0.01	ND
	90	0.260 ± 0.03	ND	0.318 ±0.01	0.222 ± 0.00	0.147 ± 0.00	ND	0.350 ± 0.00	0.125 ± 0.00	0.267 ± 0.00	0.09 ± 0.01	0.191 ± 0.01	ND
Pb	28	0.101 ± 0.02	0.05 ± 0.00	0.197 ± 0.01	0.143 ± 0.01	0.290 ± 0.02	0.022 ± 0.02	0.290 ± 0.01	0.068 ± 0.00	0.300 ± 0.00	0.105 ± 0.27	0.190 ± 0.01	0.068 ± 0.00
	60	0.129 ± 0.01	ND	0.210 ±0.00	0.128 ± 0.01	0.231 ± 0.00	ND	0.270 ± 0.01	ND	0.316 ± 0.01	0.080 ± 0.22	0.182 ± 0.01	0.020 ± 0.00
	90	0.197 ± 0.01	ND	0.190 ± 0.01	ND	0.280 ± 0.01	ND	0.201 ± 0.01	ND	0.290 ± 0.00	0.050 ± 0.25	0.101 ± 0.01	ND
Zn	28	0.653 ± 0.02	ND	0.690 ±0.00	0.689 ± 0.01	0.669 ± 0.01	ND	0.793 ± 0.00	ND	0.837 ± 0.01	0.198 ± 0.01	0.818 ± 0.01	ND
	60	0.639 ± 0.01	ND	0.641 ± 0.00	0.617 ± 0.01	0.601 ± 0.00	ND	0.785 ± 0.01	ND	0.814 ± 0.00	0.150 ± 0.00	0.774 ± 0.01	ND
	90	0.612 ± 0.01	ND	0.630 ±0.00	0.600 ± 0.01	0.516 ± 0.05	ND	0.751 ± 0.00	ND	0.807 ± 0.01	0.040 ± 0.01	0.581 ± 0.01	ND
Cd	28	0.090 ± 0.01	ND	0.055 ±0.00	0.040 ± 0.01	0.100 ± 0.00	ND	0.09 ± 0.01	0.05 ± 0.00	0.070 ± 0.00	0.052 ± 0.00	0.1 ± 0.01	ND
	60	0.066 ± 0.02	ND	0.051 ±0.00	ND	0.09 ± 0.01	ND	0.05 ± 0.01	ND	0.050 ± 0.00	0.045 ± 0.01	0.1 ± 0.01	ND
	90	0.063 ± 0.028	ND	0.040 ±0.00	ND	0.09 ± 0.00	ND	0.05 ± 0.01	ND	0.095 ± 0.01	0.045 ± 0.01	0.05 ± 0.00	ND
Ni	28	0.323 ± 0.01	ND	0.253 ±0.00	0.050 ± 0.00	0.210 ± 0.01	ND	0.230 ± 0.01	ND	0.221 ± 0.00	0.094 ± 0.01	0.245 ± 0.01	ND
	60	0.323 ± 0.01	ND	0.245 ±0.01	0.022 ± 0.01	0.214 ± 0.00	ND	0.275 ± 0.01	ND	0.213 ± 0.00	0.061 ± 0.00	0.216 ± 0.01	ND
	90	0.263 ± 0.012	ND	0.218 ±0.00	0.022 ± 0.01	0.161 ± 0.00	ND	0.289 ± 0.01	ND	0.201 ± 0.00	0.057 ± 0.01	0.110 ± 0.01	ND

ND not detected

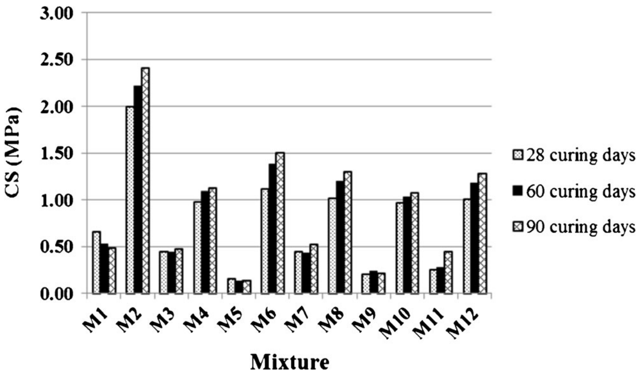

Fig. 2 Compressive strength (CS) evolution according to binders proportioning and according to time

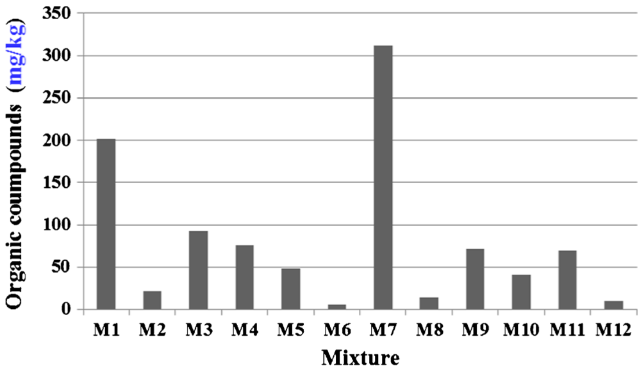

Fig. 3 Variation in organic compound concentrations in the different treated mixtures after 90 curing days

production of standard specimens in which leaching tests, chemical analyses, compressive strength (CS) measurements, and mineralogical analysis by X-ray diffraction were carried out after 28, 60, and 90 curing days. In fact, CS were determined in triplicate using MATEST CYBER-TRONIC. Leaching tests were performed in accordance with the AFNOR NF X31-211 French standards by putting in contact a specimen and distilled water using liquid to solid ratio (L/S) of 10:1, during 24 h of continuous agitation. The solution was filtered through 0.45-μn filter (GF/C) and tested for pH using a pH meter of HACH. Metallic element concentrations were analyzed using an atomic absorption spectrometry (Tromp et al. 2012). The rest of liquid from the batch-leaching test was transferred into a 1000-ml separatory or conical bottomed funnel for organic extraction, which consists in adding 30 ml of dichloromethane (DCM) and shaking it for 2 min. The complete extracted sample was then poured into a container, whose weight was recorded before pouring, for DCM evaporation at 48 h. Finally, the mass of the residual was recorded (Ouellet-Plamondon 2011). Table 1 presents the list of the tested cementations formulations for soil S/S.

Results and discussion

Contaminated soil chemical characterization

The results of chemical characterization carried out on contaminated soil are shown in Table 2.

The results of the analysis carried out on the acid attack solutions of soil indicated that the zinc (Zn) is the dominant element in comparison with other metallic elements. Its concentration is 427.59 mg/kg followed by chromium (Cr) 210.79 mg/kg, nickel (Ni) 206.8 mg/kg, Copper (Cu) 128 mg/kg, lead (Pb) 96.7 mg/kg, and cadmium (Cd) 9.08 mg/kg.

The results of the analysis conducted on soil leachates indicated an alkaline soil with a pH average value equal to 8.26. Furthermore, except for Zn, the concentration of metallic elements is higher than the maximum levels allowed by the Tunisian standard of wastewater discharge (N.T106-002). Thus, the concentrations of these elements exceeded largely the recommended threshold, thus requiring a pretreatment of the contaminated soil before discharge into the environment. Table 2 also shows that the mean concentration of organic compound in the contaminated soil is in the order of 15,520 mg/kg, which prevents the intake and the hydration of the mixture and presents large difficulties for the treatment.

Treated soil chemical characterization

The results of analysis conducted on treated soil leachates are shown in Tables 3 and 4 respectively. The pH values for the 12 mixtures varied from 8 to 11 with an average of about 9, indicating an alkaline soil. Besides, these values increased in comparison with those observed in soil before treatment, which could be due to the cements' hydration reaction (CAC and OPC + GGBFS). It could also be explained by the fact that the leaching phenomenon of the chemical species of the solid phase is less intense (Table 3).

In general, an increase in the retention of most metallic elements according to the proportioning of different binders and according to time was observed. Table 4 shows that the mean concentrations of Cr, Cu, Pb, Zn, Cd, and Ni for 3 months were less important than those observed in contaminated soil leachates. In addition, the observed leachate concentrations of different elements, except those of Pb, Cd, and Ni, were less important than those presented by the Tunisian standard. Table 4 also reveals a total retention of Cu for the mixtures M2 and M6 after 28, 60, and 90 curing days and of Zn, Cd, and Ni for the mixtures M2, M6, and M12 during 3 months.

Moreover, the retention of metallic elements was more important in mixtures M6 and M12, indicating that the use of 10% of CAC and 10% of GGBFS + OPC as shown in Table 2 plays an important role in stabilizing these elements, especially after 28 and 90 curing days where

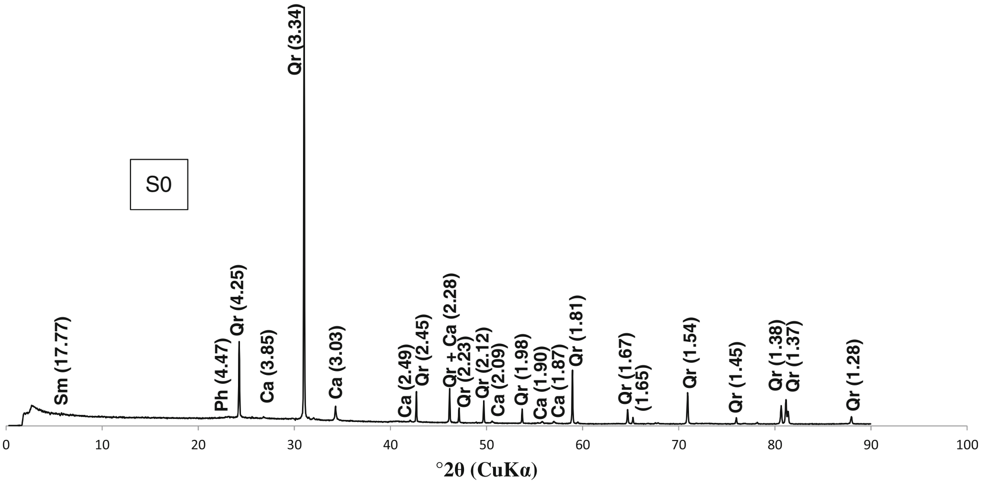

Fig. 4 X-ray diffractogram of the S0 sample. Sm, smectite; Ph, phyllosilicate; Qr, quartz; Ca, calcite. X-ray diffraction analysis of S0 showed that it is very rich in quartz, some calcite, and with low peak of phyllosilicates. NB: S0: contaminated soil

most elements were undetected. Furthermore, it should be noted that the leachate concentrations of metallic elements for the mixture M6 (10% CAC with 10% bentonite) were less than those detected in the mixture M12 (10% OPC + GGBFS with 10% bentonite). This could be explained by the fact that the CAC reduced the leachability and the diffusion of metal contaminants more efficiently than OPC even in the presence of the GGBFS. On the other hand, the addition of 10% bentonite plays an important role in the absorption of metallic elements in the cementing matrix (M'leyeh et al. 2002). Voglar and Lestan (2011), who tested the efficiency of these two binders (CAC and OPC) in stabilizing the metallic elements in a cementing matrix, confirmed our results. Indeed, these authors found that the metallic element concentrations in the soil treated by the OPC were more important than those treated by the CAC, especially for the case of Cu. It was of the order of 3.41 ppm (> 0.5 ppm) in the case of soil treated by the OPC, while it was undetected in the case of the soil treated by the CAC (Voglar and Lestan 2011).

Compressive strength evaluation

Compressive strength (CS) measurements were conducted on obtained specimens in order to examine the changes of strength characteristics (Louati et al. 2017). The evolution of the CS with time curing for different binder proportioning is presented in Fig. 2. This figure shows, except for the mixtures M1, M5, and M9, an increase in the compressive strength with time. An increase in the CS with cement content (CAC or OPC + GGBFS) was also observed. In fact, calcium silicate hydrate (C-S-H) is the main product of the hydration of OPC and is primarily responsible for the strength in cement-based materials, and when GGBSF was added, $Ca(OH)_2$ was able to react with SiO_2, Al_2O_3. Therefore, this produced more C-S-H gel (Wang et al. 2014). The decrease in the CS according to the bentonite proportioning (CAC = 5%) for the mixtures M3

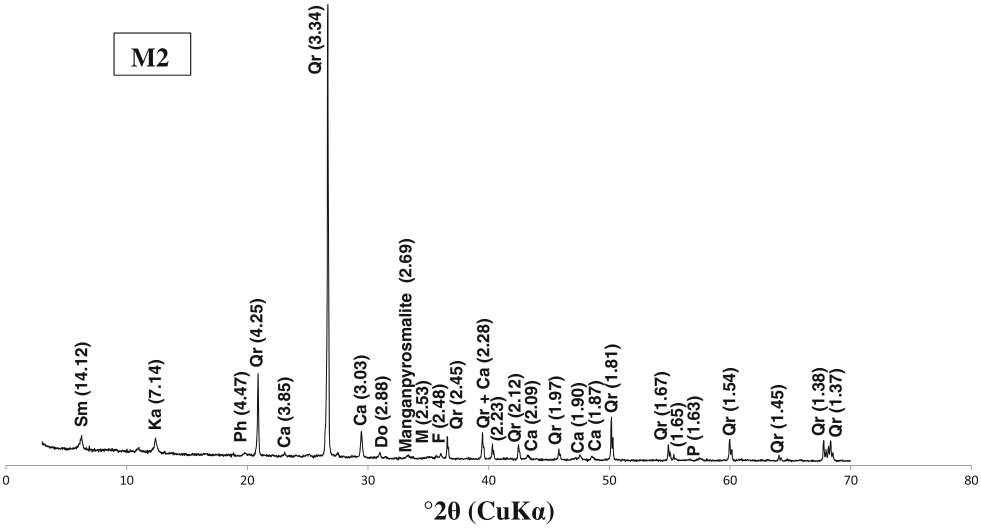

Fig. 5 X-ray diffractogram of M2 mixture treated with S/S. Sm, smectite; Ka, kaolinite; Ph, phyllosilicate; Qr, quartz; P, portlandite; Ca, calcite; Do, dolomite; F, fayolite; M, magnetite

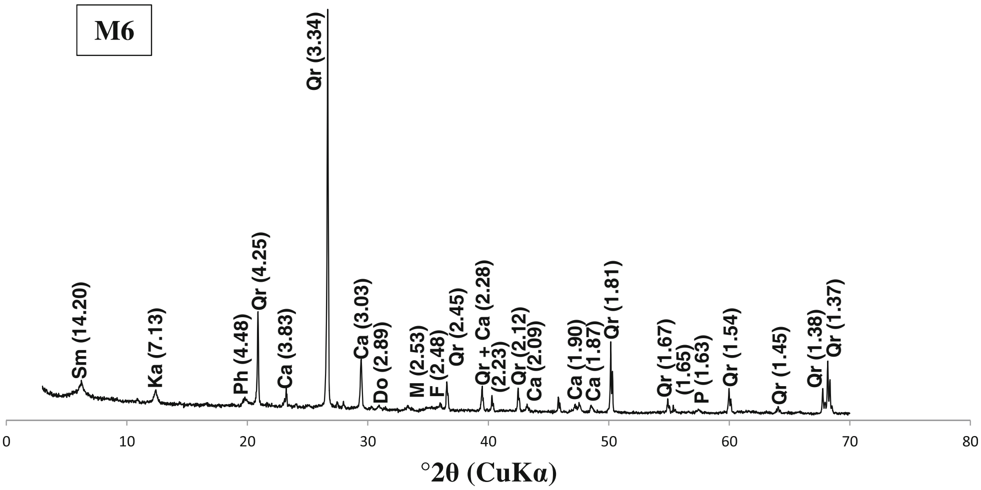

Fig. 6 X-ray diffractogram of M6 mixture treated with S/S. Sm, smectite; Ka, kaolinite; Ph, phyllosilicate; Qr, quartz; P, portlandite; Ca, calcite; Do, dolomite; F, fayolite; M, magnetite

and M5 could be explained by the fact that the bentonite can produce a negative impact on strength development. Actually, the bentonites which are foliated clay inflate with water that is why the CS of cement decreases. However, heating bentonite loses water and resists to compression which may be used as a result in future research work. Thus, the use of bentonite is not investigated on strength development but it has an important role in metallic element retention as mentioned previously. The CS for the mixtures M1, M3, M5, M7, M9, and M11 did not exceed 1 MPa standard for landfill disposal of class 3 industrial wastes treated in comparison with the other mixtures. This could be due to the low amount of used cement (5%), even in the presence of 5% of bentonite. Except for mixtures M5 and M9, all CS values exceeded 0.4 MPa. According to Malviaya and Chaudhary (2006), the minimum desired compressive strength for stabilized material should be estimated based on the design loads. For example, the US EPA considered the CS value at 0.35 MPa as a satisfactory compressive strength for materials placed on it in a landfill. Moreover, in the UK, the desired 28-day compressive strength is 0.7 MPa, but the value that is as low as 0.35 MPa is also considered acceptable, depending on the test specimen (Hills and Pollard 1997). The CS values observed in mixtures M8 and M12 are lower than those observed in mixtures M2 and M6 (Fig. 2). Furthermore, the highest values of the CS are measured in the mixture M2 (10% CAC), especially after 90 curing days where the mean value is about 2.41 MPa. Voglar and Lestan (2011) have shown that the CAC is more effective in reducing the leachability and diffusion of metal contaminants than OPC, thus producing much higher strength.

Organic compound leachability

To assess the effectiveness of S/S in immobilizing organic compounds, the leachability of these elements is also a key factor (Wang et al. 2015). Figure 3 and 4 reveal that the organic compounds detected in different treated soil mixtures are very weak in comparison with those observed in contaminated soil (15,520 mg/kg). Indeed, the values varied between 6.12 and 312 mg/kg, where mixtures M1 and M7presented the

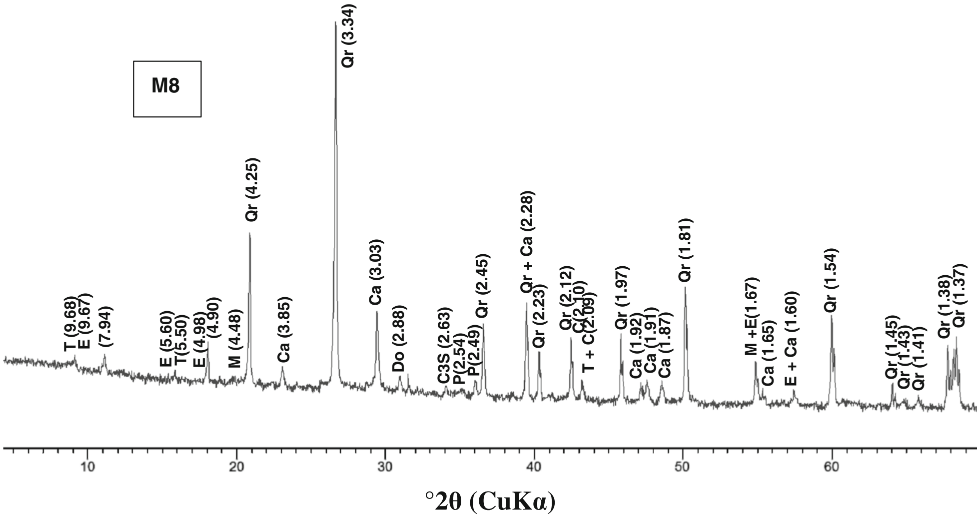

Fig. 7 X-ray diffractogram of M8 mixture treated with S/S. Sm, smectite; Ka, kaolinite; Ph, phyllosilicate; Qr, quartz; P, portlandite; Ca, calcite; Do, dolomite; E, ettringite; F, fayolite; M, magnetite; T, thaumasite

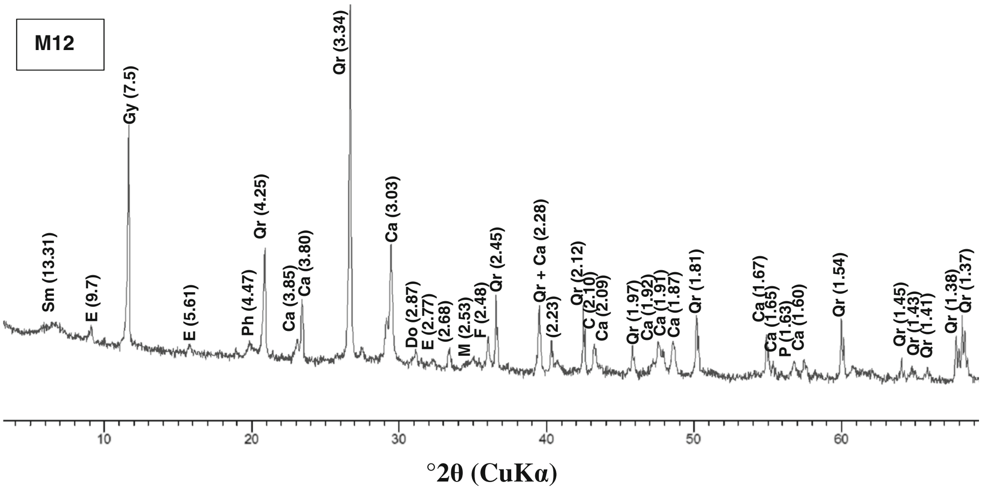

Fig. 8 X-ray diffractogram of M12 mixture treated with S/S. Sm, smectite; E, ettringite; Ph, phyllosilicate; Qr, quartz; P, portlandite; Ca, calcite; Do, dolomite; F, fayolite; M, magnetite; C, cementite

highest values in the order of 200 and 312 mg/kg, respectively, mainly explained by the less percentage of the used binders (CAC and OPC + GGBFS) in the order of 5%. The lowest values are presented in mixtures M6, M12, M8, and M2, respectively, indicating the effectiveness of the used cements (CAC and OPC + GGBFS) with the presence of 10% of bentonite. This is because of the high absorption ability of organic compounds by cements and, especially, by the bentonite (Gillman 2005). Furthermore, bentonite was considered, in the current research, as a degreasing agent.

Treated soil mineralogy

This current research was evaluated by the treatment of the soil using the XRD analysis. X-ray diffraction was important in order to know the mineralogical composition of the treated soil in comparison with contaminated soil. In fact, the mixtures M2, M6, M8, and M12 were selected. These mixtures gave satisfactory results regarding compressive strength and metallic element immobilization. The mineralogical analysis detected in M2 revealed the presence of clay minerals, such as smectite and kaolinite, as well as accessory minerals, such as quartz and calcite (Fig. 5). For M6, the mineralogical analysis has shown that this mixture is very rich in quartz, calcite, clay mineral, such as smectite and kaolinite (Fig. 6). M8 showed the presence of ettringite and thaumasite (Fig. 7). M12 revealed the abundance of quartz and gypsum and the presence of calcite (Fig. 8).

The presence of the quartz and the detection of phyllosilicates in mixture M2 proved the natural origin of these elements and the addition of bentonite in M6. The polluted soil (S0) is clayey sand (Fig. 4). Furthermore, new mineral phases compared to contaminated soil S0 like magnetite, portlandite, manganpyrosmalite, and fayolite appeared (Figs. 5 and 6). These minerals ensure the durability of mixtures, which is confirmed with compressive strength results. The presence of ettringite and thaumasite in M8 reduces the mixture durability in comparison with the mixture M2, which shows the higher strength (Fig. 7). The high proportions of gypsum in M12 could be explained by the high proportion of cement (OPC + GGBFS) of the order of 10%. Besides, the abundance of ettringite compared to fayolite, magnetite, portlandite, and cementite was detected in this mixture. These minerals contained iron oxides emanating from the slags (metallurgy) used in the mixture which is rich in irons (Fig. 8).

Conclusions

In this work, the physical and chemical performance of the ex situ S/S-treated soil using novel binders (CAC and GGBFS) and additives have been investigated. Even at low proportioning of binders, this remediation retained the organic compound (like hydrocarbon) very well with the retention efficiencies of 97–99%. However, although the concentrations of most metal were lower than the standards and showed a decrease between 28 and 90 curing days, those of Pb, Cd, and Ni were higher in some mixtures. In this case, it is recommended to increase the proportioning of binders. We conclude that the retention of the total organic compound is better than that of metals. In fact, the use of 10% of CAC and 10% of OPC + GGBFS gave satisfactory results when the compressive strength exceeded 1 MPa (desired resistance). However, the result showed that CAC reduced the leachability of metallic elements and produced a higher compressive strength that is more efficient than OPC + GGBFS. Similarly, although adding 10% of bentonite to 10% of CAC or to 10% OPC + GGBFS decreased the strength, it improved the immobilization capacities on both inorganic and organic contaminants more than the use of binder alone. Therefore, bentonite has a great power of retention of the total organic and inorganic compounds. Although the use of CAC as a binder in the S/S gives better results for the strength and retention of the metal elements, it is very expensive. Hence, the use of OPC + GGBFS is satisfactory, thanks to its availability and its reduced price compared to the CAC, giving results that are not more aloof than those of CAC whether for the resistance or retention of metals. Finally, the XRD analysis in this research work confirmed the mechanical characterization by the formation of new minerals compared to contaminated soil.

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