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Evaluation and optimization of Solidification/Stabilization treatment of the exhausted oil- based drilling sludge (Southern Tunisia)

D. Louati [a, b,*], S. Selmani [b, c], M. Choura [a, b]

[(a)] *Research Unit of Environmental Geotechnical and Civil Materials, Sfax, Tunisia*
[(b)] *National School of Engineering of Sfax, University of Sfax, Soukra road Km 4, BP 1171, 3000 Sfax, Tunisia*
[(c)] *Technical Center for Building Materials, Ceramics and Glass (CTMCCV), Tunisia*

**Corresponding author. E-mail address: dhouhaloua@hotmail.fr, +216 22 681 039 +216 52 681 059*
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Abstract

Solidification/Stabilization treatment of the exhausted oil-based drilling sludge using hydraulic binders, followed by a compaction at the paste phase, was used to reduce the mobility of the pollutants while improving the physical properties of the sludge. The treatment involved mixing the exhausted sludge with required quantities of hydraulic binders such as: Portland artificial cement, air quicklime and water, corresponding to the formulations tested. The obtained smooth paste was subjected to the compaction tests using a triaxial press for the production of standard specimens, in which, leaching tests, chemical analysis and compressive strength measurements were carried out after 30, 60 and 90 days curing. Results revealed that the compaction at the paste phase resulted in optimizing the amount of hydraulic binders and water and improving the retention capacity of pollutants in a cementing matrix compared to previous works. Furthermore, it led to the improvement of the mechanical properties of the stabilized/solidified sludge.

Keywords: *Oil-based drilling sludge; Compaction; Hydraulic binders; leaching tests; Compressive strength.*

1. Introduction

Environmental protection and waste management play a crucial role in national and international priority concerns [1]. Nowadays, human health and environment quality are threatened by the increasing quantity of the generated solid waste [2]. It has been stated that a perfect sustainable society should not generate waste exceeding its own capacity of mainly treating and disposing of it according to [3].

Different solid waste treatment technologies were developed such as: containment, remediation technologies, thermal treatments, physico-chemical treatments, Solidification/ Stabilization (S/S) treatment, etc [4]. S/S using hydraulic binders is the most promising and the most frequently selected treatment due to its versatility, efficiency, time and cost to dispose [5- 6]. S/S has been extensively used since the 1950's. In recent years, S/S is a widely applied treatment for the management/disposal of broad varieties of wastes; principally those classified as hazardous [7-8]. S/S is expected to continue to be an essential tool in hazardous and radioactive waste management [9]. The Environmental Protection Agency has identified (S/S) as best demonstrated available treatment technology for at least 50 commonly produced industrial hazardous wastes. This Agency has selected (S/S) treatment for over 20% of its Superfund site source control remediation projects.

The (S/S) technique was used in order to decrease the surface area through which transfer of pollutants can occur and thus reduce the solubility and the availability of the contaminants for leaching or decreasing the leaching rates of hazardous materials (Stabilization) while improving their physical properties (solidification) [10- 11]. (S/S) treatment includes adding certain binders or additives to the

waste to make chemical bonds change in the materials, resulting in confining the waste. Hydraulic binders commonly used are Portland artificial cement, limestone, lime, lime kiln dust (LKD), fly ash, slug, etc.
Previous work has identified that the (S/S) is a reliable and an appropriate technology for the treatment of drilling sludge [12-13]. Within this framework, the objective of the current study is to evaluate and optimize the S/S treatment, using hydraulic binders, followed by a compaction at the paste phase of the oil-based drilling sludge consists mainly of heavy and alkali metals and organic constituents. Portland artificial cement (PAC) was applied as hydraulic binder in order to investigate the leaching properties and the compressive strength of the Solidified/Stabilized sludge. It has been revealed that using (PAC) in (S/S) of heavy metals containing waste was effective in reducing the leaching of these elements [14]. Furthermore, air quicklime was considered, in this current research, as a degreasing agent.

2. Material and methods

2.1. Sludge sampling

A sampling campaign of exhausted oil-based drilling sludge was conducted directly from the oil site of EDAM of the company of AGIP BV, located in the region of Oued Zar in the south of the region of EL Borma (Southern Tunisia) (Figure 1). Outdoor drilling sludge samples were collected 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 [15]. Sludge samples were taken at several points into sludge drain pan.

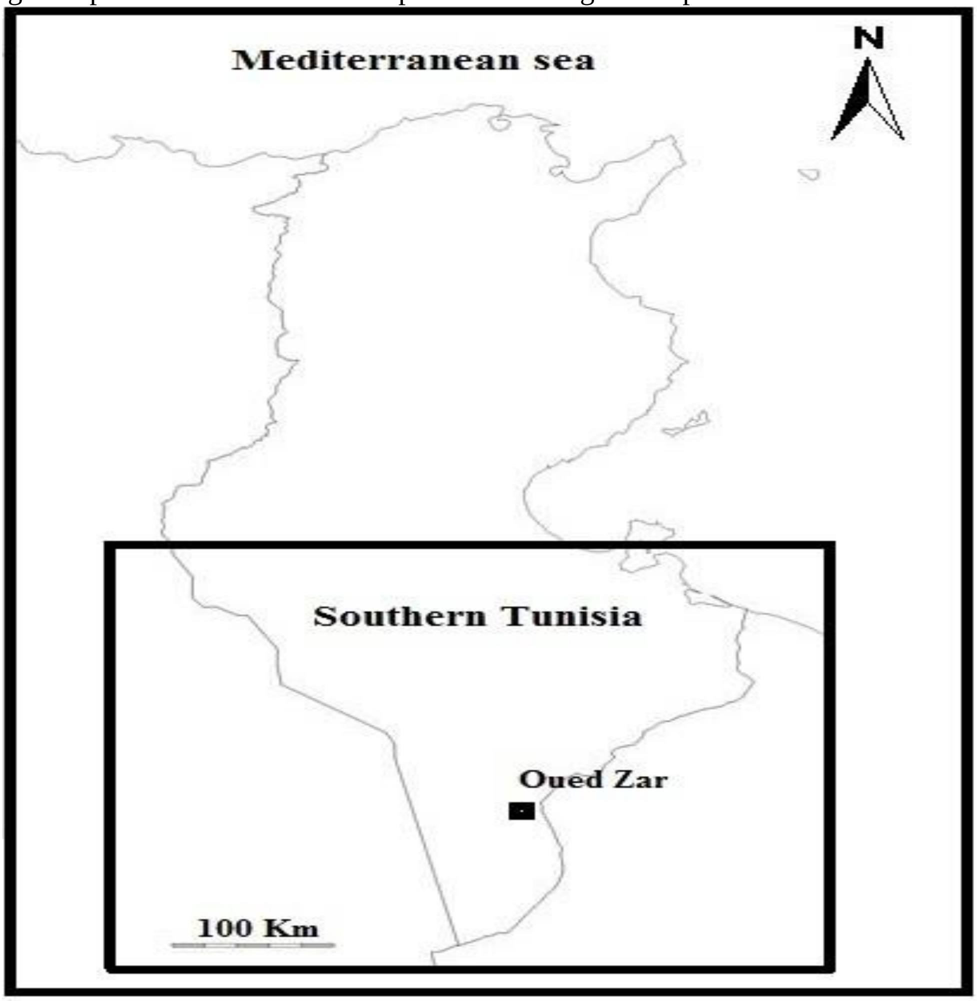

Figure 1. Sampling site: (Oued Zar, Southern Tunisia)

2.2. Chemical characterization of exhausted oil-based drilling sludge

2.2.1. Leaching tests and chemical analyses

Leaching tests were performed on exhausted sludge, containing heavy and alkali metals, according to the AFNOR NF X31-210 French standards. The leaching test was determined by putting in contact, during 24 h of continuous agitation, about 50 g of the sludge sample and 500 ml of distilled water. The solution was separated from the solid residual fraction by filtration through 0.45 µn filter. The filtrated leachates were then subjected to chemical analysis. Heavy metals (Cu^{2+}, Fe^{2+}, Cr^{3+}, Zn^{2+}, Ni^{2+}, Pb^{2+} and Mn^{2+}) were measured using an atomic absorption spectrometry, alkali metals (Na^{+}, Mg^{2+}, Ca^{2+} and K^{+}) were determined by flame photometry.

2.3. Solidification/Stabilization treatment

2.3.1. Hydraulic binders

(S/S) treatment involves mixing a binding reagent into the contaminated media or waste [7]. Different types of additives or hydraulic binders were used in order to immobilize physically and chemically hazardous components initially present in waste and thus to obtain monolithic products [10-16-17]. In this study, two types of hydraulic binders were used: (PAC) and air quicklime. The (PAC) was mainly used for pollutants chemical stability. It is the most common hydraulic binder in use today because of its availability and its reduced price [18-19]. The air quicklime was used as a degreasing agent capable of absorbing large amounts of organic compounds and retaining volatile pollutants.

2.3.2. Test specimens preparation

The treated sludge specimens were prepared for various formulations of binders, according to the AFNOR NF X31-211 French standard (diameter = 4 cm; height = 8 cm). The cement, air quicklime, exhausted sludge sample and water were put into the container and were mixed for 4 min, using magnetic mixer in order to achieve a uniform admixture [20]. The sample weight was fixed at 1.5 kg. In fact, two series of proportioning were performed. One by fixing the air quicklime percentage at 50% while varying the cement proportioning from 0 to 40%. The other by fixing the cement percentage at 10% and varying that of the air quicklime from 60 to 80%. The ratio (R) was fixed at 0.36 (Table 1). This ratio was given by the Equation 1:

$$R = \frac{Water}{PAC + Air\ quicklime} \qquad (1)$$

This ratio was chosen in order to obtain sufficient hydration for a good homogeneity and satisfactory flow capacity [21].

Table 1. Testing protocol for each mixture type

(PAC) proportioning (%)	Air quicklime proportioning (%)	Water quantity (ml)	Specimens number
10	50	324	15
20	50	378	17
30	50	432	15
40	50	486	16
0	50	270	20
10	60	378	23
10	70	432	16
10	80	486	20

The obtained smooth paste was subjected to compaction tests using a triaxial press (Figure 2) at a constant strain rate of 0.01 MPa for the production of standard specimens (Figure 3) in which were carried out after 30, 60 and 90 days curing, leaching tests, chemical analyses and compressive strength (CS) measurements. It should be noted that these specimens were stored within vacuumed plastic bags in order to avoid a change in water content and were cured in the laboratory at a temperature of 25°C.

Figure 2: (a): Triaxial press (pressure = 40 Bar); (b): Triaxial compression test for each specimen

Figure 3. Standard cylindrical specimens of Solidified/Stabilized sludge

Leaching tests were performed in accordance with the AFNOR NF X31-211 French standards by putting in contact, during 24 h of continuous agitation, a specimen and 500 ml of distilled water. The solution was filtered through 0.45 µn filter (GF/C). Chemical analysis of heavy and alkali metals were determined using the same techniques as for the exhausted drilling sludge in order to assess the short and medium-term behavior of these elements in the Solidified/Stabilized sludge.

3. Results and discussion

3.1. Exhausted sludge chemical characterization

Results of heavy and alkali metals analysis carried out on exhausted sludge leachates are shown in table 2.

Table 2. Heavy and alkali metals content in exhausted sludge leachates

Element		(N.T106-002)	Content (ppm)
Heavy metals	Cu^{2+}	0.50	0.39
	Fe^{2+}	1	ND
	Cr^{3+}	0.50	ND
	Pb^{2+}	0.10	0.20
	Zn^{2+}	5	1.59
	Ni^{2+}	0.20	0.22
	Mn^{2+}	0.50	ND
Alkali metals	Ca^{2+}	500	654
	Mg^{2+}	200	5.66
	Na^{+}	300	349
	K^{+}	50	82

N.T106-002: Tunisian standard of wastewater discharge; ND: Not detected

These results indicated that the zinc (Zn) is the dominant element in comparison with other heavy metals. Its concentration is of 1.59 ppm. Nickel (Ni) and lead (Pb) concentrations exceeded the recommended threshold. They were slightly higher than the maximum levels allowed by the Tunisian Standard of wastewater discharge (N.T106-002). Iron (Fe), chromium (Cr) and manganese (Mn) were not detected. Alkali metal concentrations, except for magnesium (Mg), are higher than those defined by the Tunisian Standard (N.T106-002) requiring, thereby, a pretreatment of the exhausted sludge before discharge into the environment.

3.2. Treated sludge chemical characterization

Results of heavy and alkali metals analysis carried out on treated sludge leachates by the PAC (air quicklime = 50%) and by the air quicklime (PAC = 10%) are shown in tables 3 and 4 respectively.

In general, an increase in the retention of heavy and alkali metals according to the proportioning of (PAC) and air quicklime and according to time was observed. Table 3 showed that the concentrations of potassium (k) and sodium (Na), after 90 days curing with a cement proportioning equal to 40%, were less important than those observed in exhausted sludge leachates and those defined by the Tunisian standard. Only a tiny amount of magnesium (Mg) is presented during three months curing. Its maximum concentration of about 0.07 ppm remains very less than that detected in exhausted sludge leachates and that presented by the Tunisian standard.

Table 3. Variation in heavy and alkali metals content in treated sludge leachates by the (PAC)

Element		Day	(PAC) proportioning (%)				
			0	10	20	30	40
Heavy metals (ppm)	Cu^{2+}	30	0.070	ND	ND	ND	ND
		60	0.090	0.080	0.06	0.060	0.030
		90	0.080	0.060	0.06	0.040	0.030
	Ni^{2+}	30	0.015	0.015	0.013	0.011	0.011
		60	ND	ND	ND	ND	ND
		90	ND	ND	ND	ND	ND
Alkali metals (ppm)	Mg^{2+}	30	0.070	0.060	0.03	0.020	0.010
		60	0.060	0.040	0.03	0.010	0.010
		90	0.050	0.040	0.04	0.020	0.010
	Na^{+}	30	340	292	266	256	128
		60	340	260	228	227	116
		90	318	258	220	220	107
	K^{+}	30	252	157	139	89	68.6
		60	250	120	115	60	55
		90	250	100	102	52	40

This table also showed a total retention of calcium (Ca) during three months. Regarding heavy metals, a total retention of manganese (Mn), iron (Fe), zinc (Zn), chromium (Cr) and lead (Pb) was observed. A nickel (Ni) is present in very small amount. Its maximum concentration of about 0.015 ppm remains very less than that detected in exhausted sludge leachates and that presented by the Tunisian standard. After 60 and 90 days curing, a total retention of this element was observed. Copper (Cu) is completely retained for (PAC) proportioning ranging from 10 to 40%. After 60 and 90 days curing, this element is present in very small amount especially in the absence of cement while keeping values less than those detected in exhausted sludge leachates and presented by the Tunisian standard. [13] treated the exhausted oil- based drilling sludge, using the classical technique that uses a simple vibration of sludge at the paste phase and the high sulphate resistant cement (HSR) instead of the (PAC). Results indicated that the copper persists in small amounts (0.04, 0.03 and 0.02 ppm), respectively, for (HSR) proportioning ranging from 40 to 60% and R= 0.4. Therefore, (PAC) has a more significant effect on the retention of heavy metals, including copper, than the (HSR). Several researchers have used different hydraulic binders such as (PAC) and fly ash for Stabilization and Solidification of electroplating waste [22]. They reported that the cement has shown better results. After 90 days curing, the concentrations of potassium (k) and sodium (Na) in the (S/S) treated sludge leachates by the air quicklime were less important than those observed in exhausted sludge leachates and those defined by the Tunisian standard (table 4). Besides, a total retention of magnesium and calcium was reported during three months curing. Regarding heavy metals, a total retention of copper (Cu), iron (Fe), manganese (Mn), zinc (Zn), chromium (Cr), Nickel (Ni) and lead (Pb) was recorded after 30, 60 and 90 days curing.

Table 4. Variation in alkali metals content in treated sludge leachates by the air quicklime

Alkali metals (ppm)	Day	Air quicklime proportioning (%)		
		60	70	80
Na^{+}	30	288	284	254
	60	259	210	240
	90	243	186	220
K^{+}	30	148	137	121
	60	121	116	70
	90	100	85	45

Obtained results confirm those found by researchers who applied the compaction of sludge at the paste phase, by using (PAC) [8]. In fact, these results indicated that the compaction at the paste phase of this treated sludge resulted in up to 35% enhancement of the retention of pollutants, in a cementing matrix, as compared with the classical technique that uses a simple vibration of sludge at the paste phase. Therefore, (S/S) treatment of exhausted sludge using (PAC), which varies only from 0 to 40% and with R = 0.36, resulted in satisfactory results. Compared with the classical technique, the implemented technique led to an improvement in the compactness of the sludge, and thus assured a better retention of heavy and alkali metals in response to the leaching of this treated sludge. Thus, compaction at the paste phase of the Solidified/Stabilized sludge resulted in optimizing the amount of hydraulic binder and water (40% water gain) and improving the retention capacity of pollutants in a cementing matrix.

3.3. Compressive strength evaluation

Compressive strength (CS) measurements were conducted on obtained specimens in order to examine the changes of resistance characteristics. The evolution of compressive strength (CS) with time curing for different (PAC) proportioning (air quicklime = 50%) and different air quicklime proportionings (PAC = 10%) are presented in Figures (4 and 5) respectively. Figure 4 shows that the (CS) evolved toward increasing according to (PAC) proportioning and over the time curing, which agrees with the results of previous studies indicating that the (CS) increased with an increase of time curing and with cement content [20-23-24]. In fact, after 30 days curing, the CS increased gradually from 1.7 to 4.5 MPa according to (PAC) proportioning. After 60 days curing, it increased considerably of about 1 MPa for (PAC) proportioning of 20 and 40%. Passing from 60 to 90 days, the increase in the (CS) according to (PAC) proportioning did not exceed 1 MPa in comparison with that observed previously. This could be due to the slow carbonation process of quicklime. Thus, the (CS) of 1 MPa was obtained for a (PAC) proportioning = 10%, while, this value was obtained for a (PAC) proportioning = 40% in the study carried out by [13] indicating that the compaction at the paste phase of the treated sludge resulted in better cohesion and resistance of stabilized/solidified specimens by improving the mechanical properties of this sludge.

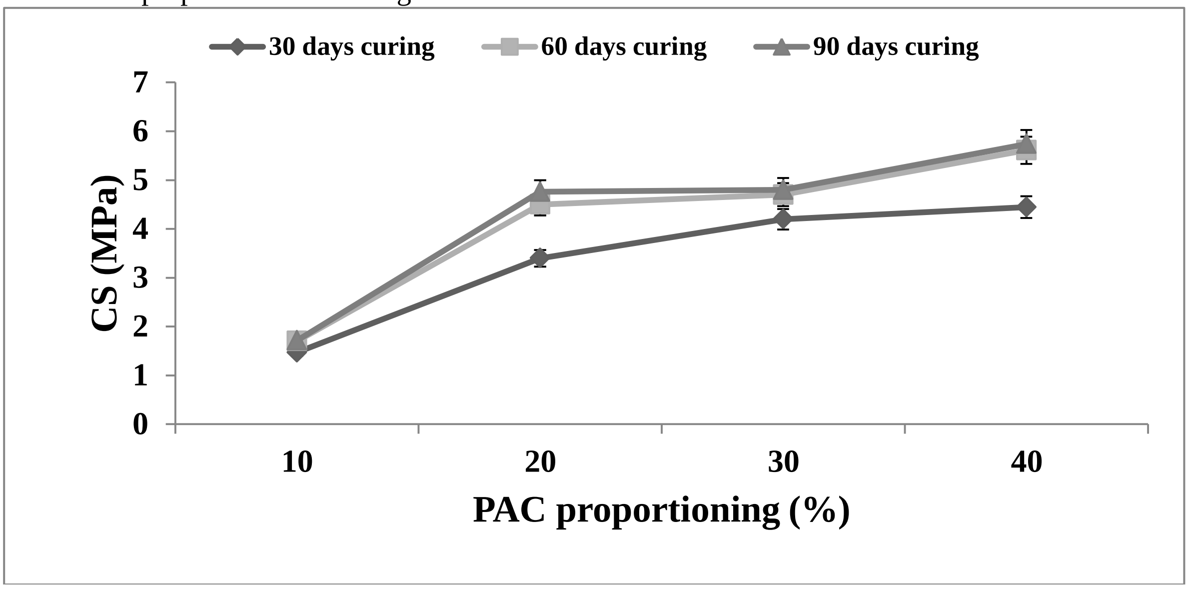

Figure 4. (CS) evolution according to (PAC) proportioning and according to time [25]

After 30 days curing, the CS evolved linearly from 1.47 to 2.24 MPa; according to air quicklime proportionings (Figure 5). After 60 and 90 days curing, lime proportioning led to an increase in the (CS) of the treated sludge up to 70%. Beyond this limit, a significant decrease in the (CS) was recorded. This could be due to the slow carbonation process of quicklime. Thus, (PAC) has a more significant effect on the cohesion and the resistance of the stabilized/solidified specimens, than the air quicklime.

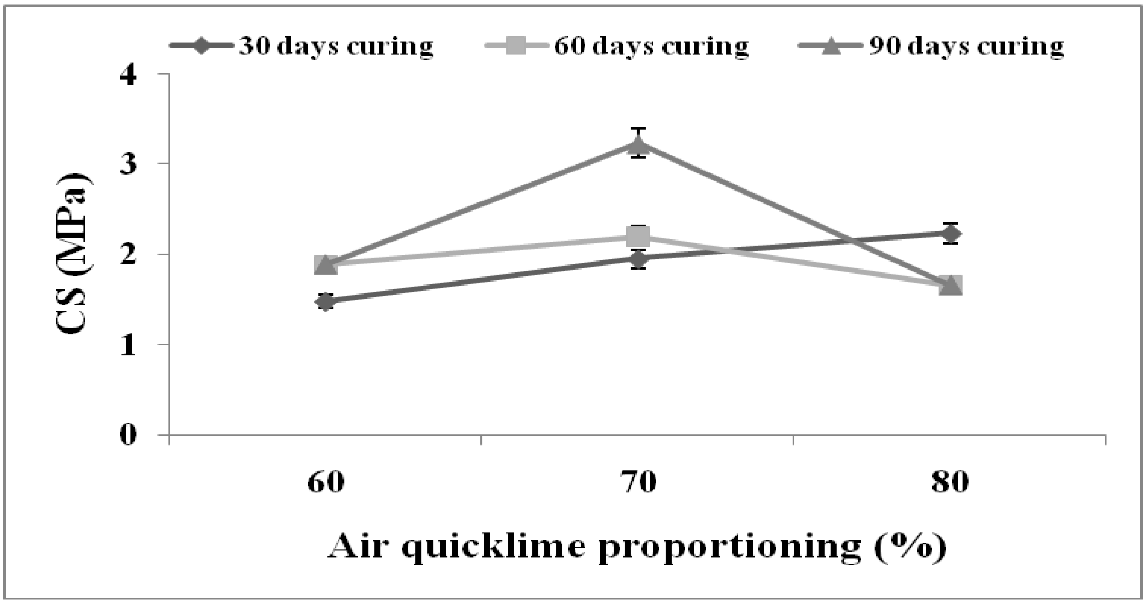

Figure 5. (CS) evolution according to air quicklime proportioning and according to time [25]

4. Conclusion

The (S/S) treatment by compaction at the paste phase is used to treat the oil-based drilling sludge as a hazardous waste for disposal and in the remediation of contaminated lands. This treatment was able to treat this sludge, by reducing the leaching level of pollutants from cementing matrix, even at low proportioning of binders, especially for the case of the (PAC). Indeed, a total retention of the majority of the heavy metal was observed. Regarding alkali metals, the retention in these elements increased according to binders proportioning and according to time. Besides, Retention of alkali metals was more important for the case of (PAC). The use of air quicklime, as a hydraulic binder, is not satisfactory for the confection of specimens and the retention of pollutants. This could be due to the slow carbonation process causing a weakening of the structure of specimens during days curing. In this study, the air quicklime was used as a degreasing agent capable of absorbing large amounts of organic compounds. The compressive strength of standard specimens depends largely on the presence of (PAC) even at low quantities. Therefore, (S/S) treatment using cement as hydraulic binder is not applied just for this particular waste. This treatment is applicable for the treatment of all types of hazardous wastes containing heavy metals especially those from industrial units.

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