

Evaluation of the Permeability of Clay-Lime Mixtures for Hazardous Waste Insulation Barriers, Case of Southern Algeria (Adrar)

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Abstract

Containment of hazardous waste is one of the methods that can be used today for its disposal worldwide, especially in landfills. In order to preserve groundwater, landfills are constructed on a natural or reconstituted clay layer of low permeability, known as a passive safety barrier.

This paper presents an experimental laboratory study that aims to upgrade local materials for use as a passive barrier in the bottoms of hazardous waste storage facilities (HWSCs). The basic materials are clays from the Adrar region in southern Algeria. An experimental study based on physical-chemical characterization and mechanical tests was performed on 2 samples of clayey soils (AA, Adrar clay, and AB, Bouda clay). Physico-chemical and mechanical characterization of Adrar clay (AA) mixtures with lime was conducted to minimize swelling and improve the studied soil. The permeability criterion allowed the adoption of an optimal mixture of Adrar clay + 6 % lime for a permeability lower than 10^{-12} m/s.

Keywords: Clay, Adrar, Passive barrier, Lime, Permeability

Introduction

Hazardous waste has one or more properties that can be solid, organic (solvents, hydrocarbons, Etc.), mineral (acids, metal hydroxide sludge, Etc.), or gaseous, according to the French National Federation of Pollution Control and Environmental Activities (FNADE) [1]. Hazardous waste can cause significant damage to our health and the environment. The quick technologic development and the population's demands are causing to the production of growing quantities of hazardous waste. Globally, the United Nations Environment Programme (UNEP) has estimated that more than 400 million tons of hazardous waste are universally produced yearly, or nearly 13 tons per second, mainly by industrialized countries (The World Counts 2023). In a single generation, the production of artificial chemicals has increased by 40,000 %, from 1 million to 400 million tons annually [2] World Counts 2023). In southern Algeria, rapid urban development and the growth of the petroleum industry are starting to create large quantities of hazardous waste, which pose a fundamental problem in managing and controlling pollution risks of the groundwater table, one of the solutions is the confinement of this special waste in waste storage centers.

The modes of hazardous waste treatment include different channels, such as recycling and recovery, physical-chemical, biological, thermal or storage treatments, FNADE [1]. The solution to this research's contamination problem is storage; it allows enclosing the waste in specific places, called waste storage centers, using insulating barriers (passive barriers). These barriers can be artificial (geomembranes) or natural like compacted layers such as mixtures of sand and bentonite [3-5]; mixtures of sand, bentonite and organophilic clay [6-9]; bentonite, calcareous sand and tuff [10,11]; geosynthetic clay (bentonites and geotextiles) [12] artificially prepared in the laboratory [13]; silty soils [14]; compacted clays [12,15,16]; clayey sand [17]; soil mixtures and cement kiln dust (CKD) [18]; lime-treated soil mixtures [19,20]; lime-softening sludge [21]; dam sediments and bentonite [22]; clay and bentonite [23,24]; red mud (a waste product from the refinery) [25]; red mud, clay and cement [26]; clay waste and bentonite [27,28].

Many authors have studied the permeability of the passive layer of hazardous waste disposal sites, such as permeability on modified bentonites for crude oil leachate Fathollahi *et al.* [29], $K = 5.2 \times 10^{-9}$ and 9.56×10^{-11} m/s with water; mixtures of 70 % sand and 30 % sodium bentonite ($S_{70}B_{30}$) Heidarzadeh and Parhizi [6], $K = 1.16 \times 10^{-11}$ m/s; with a mixture of 85 % sand and 15 % bentonite ($S_{85}B_{15}$) for Gueddouda *et al.* [3], $K = 4.58 \times 10^{-11}$ m/s; a mixture of 30 % sepiolite by weight of zeolite exposed to Copper and Crome waste [30], $K = 1.17 \times 10^{-8}$ m/s; [23] $K = 6.83 \times 10^{-11}$ m/s to water and 7.8×10^{-9} m/s to crude oil. Green *et al.* [31] compared the water permeability of 3 types of soils such as 13.5×10^{-9} m/s for Fire clay (soil) and 220×10^{-9} m/s for Kosse Kaoline and $K = 38 \times 10^{-9}$ m/s for Ranger Shale; Dabska [21] obtained a value of 6.5×10^{-9} cm/s to achieve a compacted lime softening sludge lining.

Permeability depends on pore volumes, such as density, initial water saturation degree, and pore size distribution. Other parameters that affect the permeability value are mineralogy, particle size distribution, plasticity index, liquid limit and compaction [17,21,32-36]. Alkaline materials such as lime can also have an effect on soil permeability [37-40]. Alkaline chemicals can modify not just the physical properties but as well the chemical properties of clays. Our experimental study clarifies this issue; it investigates the effect of mixing lime to clay to improve the barrier characteristics of landfills. These characteristics include in particular changes in permeability, heavy metal capture, and puncture resistance of the passive barrier [19]. The choice of lime is justified by the pozzolanic reaction that changes the pore size distribution to finer ones [38]. The total porosity is reduced [37] and if the content of lime is adequate, this reaction can be predicted to lead to a decrease in permeability [38,39]. But Only flocculation of the clay particles occurs, when the lime content is insufficient for the pozzolanic reaction (pH below 10), increasing permeability. Other research [39,41], have also shown that moisture content can significantly affect the permeability of lime-treated soils. It can be indicated that lime can give a double benefit to the pozzolanic reaction, firstly lime creates an environment with high pH which solubilizes silica and alumina, and secondly, lime gives residual free calcium, which combines with the silica and alumina provided by the clay to create the pozzolanic reaction. This transformation continues by forming cementitious products: Lime stabilization of fine-grained soils results in an increase in bearing capacity and a decrease in swelling and shrinkage potential [42]. As a result, stabilized soil has higher strength, lower permeability, and lower compressibility than natural soil [43].

In order to be effective for insulation, passive barriers must meet certain specifications, namely: [3,44].

1) The permeability in the saturated state is between 10^{-8} and 10^{-10} m/s [7,45,46].

2) Material physical stability in contact with water [47].

3) The swelling properties must be such as to ensure good contact with the host rock, They make it possible to fill in existing cracks that would otherwise develop in the future [48].

Another environmental and vital interest of the study area of southern Algeria contains an extensive reservoir of groundwater, the water sheet of the Albian [49]. This aquifer is the largest freshwater reserve in the world and extends between Algeria, Tunisia, and Libya. Approximately 70 % of the aquifer is located in the Algerian territory southeast of the country, **Figure 1**. This particularity requires urgent protection in front of the progression of pollution sources.

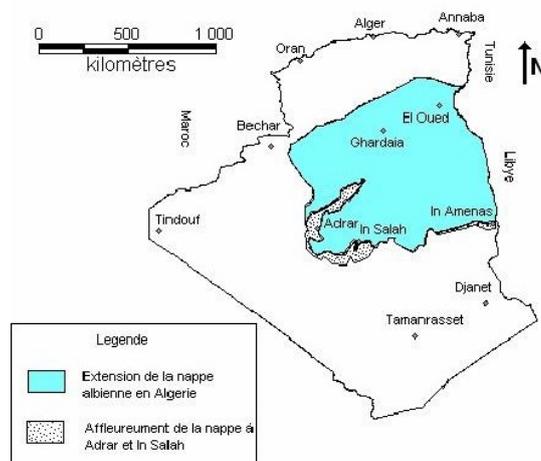


Figure 1 Geographical extension of the Albian nappe in Algeria [50].

The essential deposits in the Adrar region in southwestern Algeria are considered locally available natural and economical materials and without environmental risks. Within the framework of sustainable development in front of this problem in a vast and arid region, we propose that we have devoted our work to improving engineered barriers (passive layer) of clay which will be intended for the realization of the funds of the waste storage centers (WSC) and which brings us to a purely ecological solution. The material is approved based on the following criteria, its nature, permeability, workability and adequate mechanical parameters.

The study in question presents a description of laboratory testing techniques. It examines the experimental results of clay and lime blends, suggesting an appropriate and optimum combination to satisfy the regulatory criteria and the technical-economic aspects of the design in a severe climatic environment [51,49]. Two measurement techniques have been used for a reasonable estimate of permeability: the exploitation of the odometer test and the use of the triaxial cell (GDS). The adequate lime addition quantity that satisfies the permeability requirements will be determined in this case. Without forgetting to point out that the research is purely concerned with the technical improvement of the insulation layer and the protection of the water table from hazardous waste; our study strongly contributes to the containment solution and not the treatment of this waste. This study directs us towards several avenues for improving the passive layer with respect to leachates from different sources such as crude oil, crude oil, municipal leachates etc.

Materials and method

Location site

Geological context

The local area is located in the Lower Cretaceous, commonly called “intercalated continental”. It consists of fine sandstones weakly cemented and clays. The Cambrian formation is represented by pebbly sandstones, ferruginous quartzites and sandstone-clay shales, and locally low-grade sands and anhydrite) (Adrar Mines Directorate, see **Figure 2**).

The 1st sample (AA) is known as Adrar clay and it comes from a deposit located a few kilometers north of the city of Adrar (the eastern part of the RN6). It was taken at a depth of 5 to 10 m (photo AA in **Figure 3**). This material is red. The 2nd sample (AB) is named Bouda clay. It is located west of Adrar; it was taken at a depth of 0.5 to 1 m (photo AB of **Figure 3**). According to the code of MUNSELL (Soil color cats, revised edition 1994), the Adrar clay is classified between 10R 4/6 and 10R 6, while the Bouda clay is classified between 10R 6/4 and 10R 6/6. The 3rd material used is lime from the Ghardaia region in central Algeria. The clay has been treated with lime with several percentages for stabilization and improving its geotechnical properties, namely the permeability subject of this research is a critical factor for use in waste storage centers.

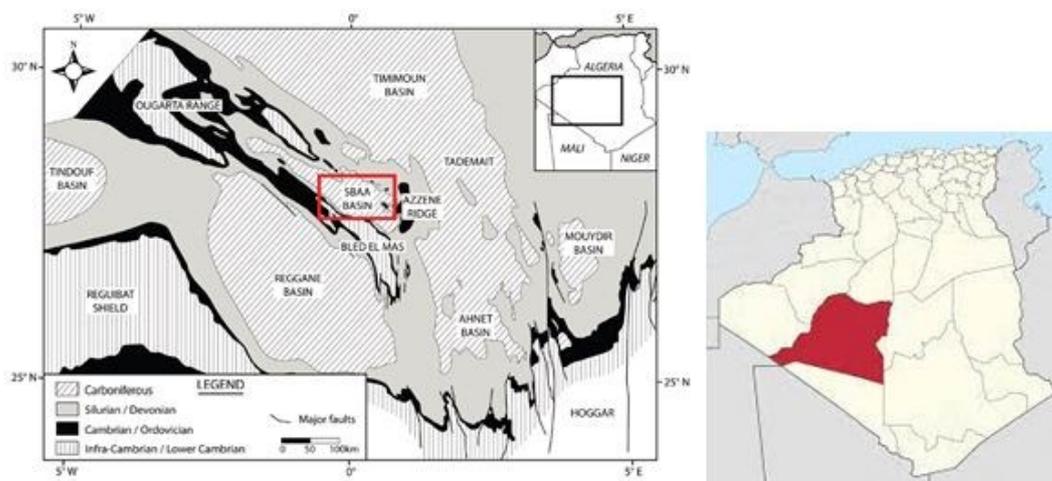


Figure 2 Subcrop geological map of Algeria from WEC Algeria 1996 (Schlumberger / Sonatrach) in [52].

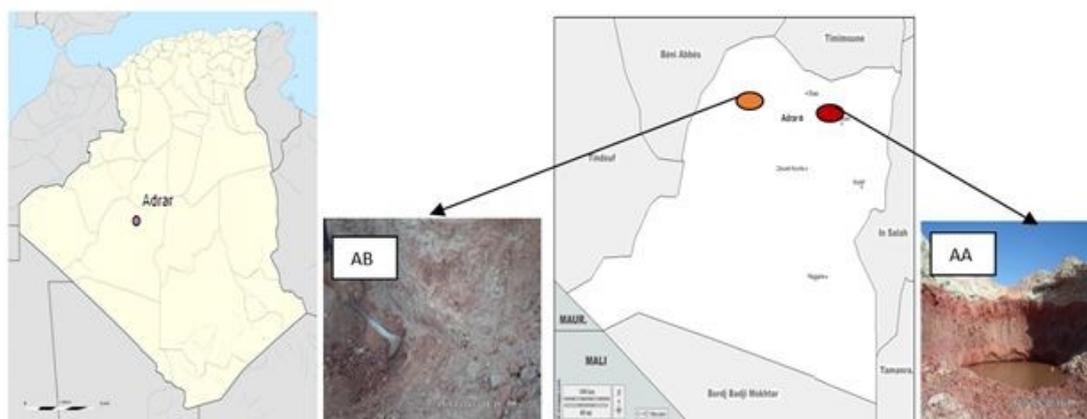


Figure 3 Location of Adrar AA and Bouda AB sites.

Sample preparation

In this study, several mixtures of Adrar clay (AA) and lime (C) were considered to achieve the desired soil combination: 2 % L and 98 % AA, 4 % L and 96 % AA, finally 6 % L and 94 % AA. Determining the minimum proportion of clay to lime should result in an appropriate permeability (10^{-8} to 10^{-10} m/s) or less [7]. Consolidation and compaction tests were performed. The collected natural clay was ground into fine particles that could pass through a 4mm sieve for the Proctor tests and the 2 mm for the triaxial and pH tests after oven drying the samples at a temperature of 105 °C. The quantities of lime and water were calculated in percentage of weight. To each mixture was added the amount of water necessary to reach the optimal water content of the Modified Proctor Test (MOP) [7,17,22] according to NFP 94-093. The Samples were compacted to maximum dry density on compaction curves. All mixtures were prepared manually and carefully to obtain remarkable homogeneity.

Compaction test

For each mixture, the modified Proctor optimums were obtained graphically from the curves as a function of the percentage of lime. The results are summarized in **Figure 8**.

Free swelling tests

The tests were conducted at the laboratory of the University of Oran ENSET. The free swelling test is performed on natural and treated samples. The static compaction test with the characteristics of the Proctor optimum reconstituted the latter. Static compaction consists of subjecting the sample to a static pressure under a slow speed of 1mm/min, and the soil is compressed in a mold with the help of a press. The dimensions of the samples are as follows: Height $H = 20$ mm, diameter $D = 50$ mm [53,54].

The free swelling tests are carried out in a conventional odometer according to XP P94-091. First, the sample is prepared (crushed, sieved, and dried), followed by static compaction at Proctor optimums. In a cylindrical cell, the sample is placed between 2 porous stones; after it is smoothed and leveled at the top and bottom, under a very low load (of the order of the weight of the piston), then the sample is saturated for several days until stabilization. The swelling rate under odometer conditions is the ratio between the variation of the height of the specimen and its initial height.

The total swelling noted G (%) is determined according to the following formula:

$$G \% = (\Delta H/H_0 \text{ in } \%); \quad (1)$$

G : Variation in the height of the sample.

With: $\Delta H = H_f - H_0$; H_0 : initial height; H_f : final height after stabilization.

Permeability coefficient

Indirect measurements of permeability

Indirect methods of evaluating permeability k (m/s) are adopted by the results of the odometer test on samples reworked using graphical results (compressibility and consolidation curve). The permeability k is determined using the consolidation coefficient C_v (m^2/s) obtained by the Taylor and Casagrande method and the volume compressibility coefficient m_v (m^2/kN). This approach defines the C_v and m_v coefficients

from the compressibility and consolidation curves. The Taylor and Casagrande methods allow estimating the value of the permeability (k) as a function of the parameters according to the following formula:

$$k = C_v \cdot \gamma_w / E' \quad (2)$$

C_v : Coefficient of consolidation; E' : Oedometric modulus; γ_w : Volume weight of water.

Direct measurements of permeability

The Sample is prepared under optimum modified Proctor conditions by static double-piston compaction. Static compaction is chosen because it produces for obtaining more homogeneous samples. After the sample has been placed inside the triaxial cell, a confining stress of $\sigma'_3 = 100$ kPa is 1st applied to maintain the sample in place and to ensure that there is no space between the membrane and the sample. Application of a low back pressure at the base of the sample ($u_e = 20$ kPa) to eliminate air bubbles between the membrane and the sample (u_e : Base pressure). Gradual increase in the confining stress and back pressure (always at the base) to allow the release of air bubbles in the connection tubes ($u_e = 30, 40$ kPa) while keeping the mean effective stress constant and keeping the outlet valve open ($u_s = 0$ kPa), (u_s : Back pressure) the interstitial pressure at the top of the sample. The vertical deformation of the specimens was recorded as a function of time and the specimens were considered saturated when the piston displacement became constant (less than 10 %), (less than 0.01 mm in 24 h) [46]. After the saturation phase, the desired confining stress is applied, which is always greater than the back pressure applied to the base of the sample $\sigma'_3 = 250$ kPa (cell pressure). A back pressure is applied at the sample outlet equal to that at the base ($u_e = u_s = 100$ kPa). Keeping u_s constant, we increase u_e , in our case $u_e = 200$ kPa. The permeability was calculated according Darcy's law, which explains the relationship between the quantity of water flowing through a cross-section for a specific time. The following expression gives it:

$$v = Q/A = k i \quad (3)$$

v : Darcy flow (m/s);

Q : Flow rate (m³/s);

A : Section of the sample;

i : The hydraulic gradient is calculated by the following expression;

$$i = \Delta u / \gamma_w H$$

where Δu is the pressure difference between the 2 ends of the sample ($u_e - u_s$) (Kpa).

H : Height of the sample (m).

γ_w : water density (kN/m³).

To determine the saturated permeability, and satisfying Darcy's law, the following steps are followed:

- A time interval is set between t_1 and t_2 ($\Delta t = t_1 - t_2$), t (s).
- Determine the change in volume in this time interval ($\Delta V = V_e - V_s$), V (mm³).
- The flow rate is determined ($Q = \Delta V / \Delta t$), Q (mm³/s).
- The velocity of the flow is determined ($v = Q/A$), where A is the cross-sectional area of the sample.
- The value of k is determined from Darcy's law:

$$v = k i \quad (4)$$

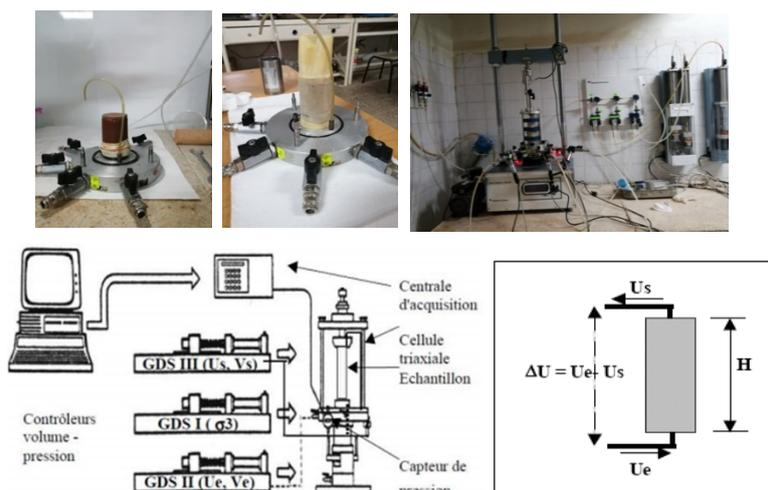


Figure 7 triaxial cell and its pressure-volume control system (GDS) and HP Agilent 34970A acquisition system.

Results and discussion

Clayey soil identification

XRD analysis

The X-ray diffractometer was used in this study to determine the different proportions of the minerals that make up our soils, (see **Figure 4**). The diffractograms reveal that the AA soil presents a significant mixture of muscovite/Illite ($KAl_3Si_3O_{10}(OH)_2$), as well as quartz (SiO_2 -illite) and Kaolinite with the presence of iron (geotite FeO_2 , hematite Fe_2O_3 and magnitite Fe_3O_4). These irons have a high density, they contribute to the strengthening of the mechanical properties of the clays [55], while the soil AB presents a mixture of quartz SiO_2 , phengite (Ph), Sapphirine, as well as some traces of magnitite (Fe_3O_4). Finally, the lime presents a mixture of lime, portlantite and calcite.

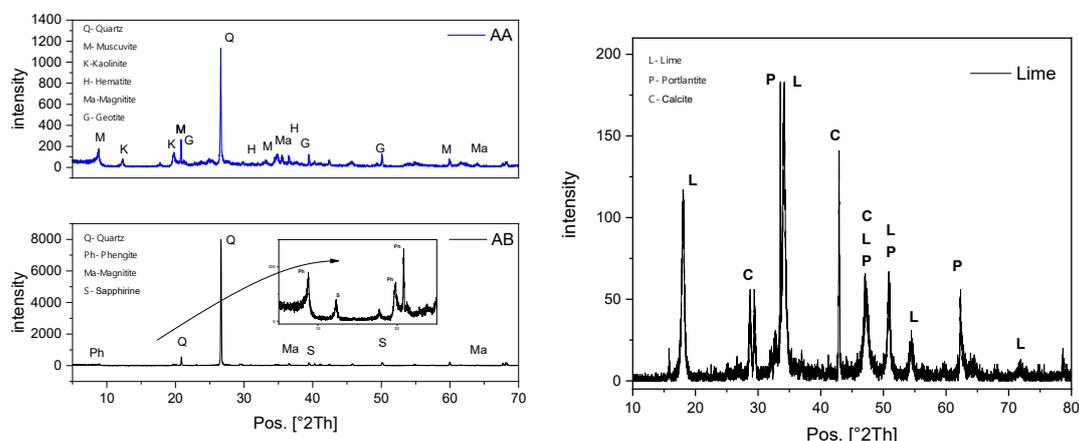


Figure 4 Mineralogical analysis of materials used.

SEM analysis

Scanning electron microscopy (SEM) allows us to observe the clay sample's texture and characterize the mineralogical assemblages. The SEM images of the 2 samples, AA (a) and AB (b), are shown in **Figure 5**. The clayey particles appear as clusters of fine aggregates and rod-shaped platelets with irregular contours. This is a morphology encountered as well for Illites/muscovite. The images of **Figures 5(a) - 5(b)** agree with what we obtained by XRD and the presence of carbonates in the AB clay and Iron in the AA clay. The carbonates (Calcite $CaCO_3$) are present as well-visible aggregates and Iron. On the other hand, the Quartz SiO_2 is present in the form of small size grains. The EDX spectrum of **Figures 5(c) - 5(d)** highlight for both types of clay the presence of the main constituents of the clay, such as silicon and aluminum, with a constant

quantity of potassium and an essential quantity of Iron for the AA clay, on the other hand, the AB clay presents some traces of Iron, magnesium, and calcium. % Atomique (At) (massique)

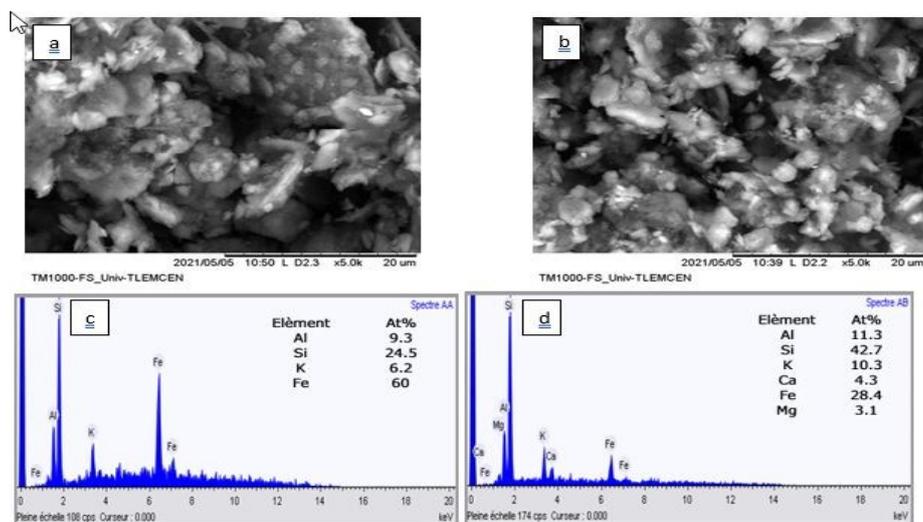


Figure 5 Scanning electron microscope SEM photographs and EDX analysis carried out AA and AB clays.

Chemical's characteristic

Chemical analyses of the 2 clay soils with the 2, 4 and 6 % AA lime mixtures are summarized in **Table 1**.

Table 1 Chemical analysis of clayey soil.

Parameters	Denomination (%)	AA (%)	AB (%)	Lime (L)	AA + 2 % L	AA + 4 % L	AA + 6 % L
Silicon oxide	SiO ₂	51.23	59.23	10.56	40.59	30.50	21.60
Aluminum oxide	Al ₂ O ₃	16.89	10.46	0.49	14.63	11.06	7.99
Titanium dioxide	TiO ₂	1.40	1.07	0.07	0.80	0.77	0.70
Ferric oxide	Fe ₂ O ₃	17.45	7.99	0.26	8.06	6.80	5.60
Calcium oxide	CaO	0.06	4.66	52.88	11.86	23.42	34.73
Magnesium oxide	MgO	0.82	0.11	15.70	4.83	7.90	10.36
Sodium oxide	Na ₂ O	0.24	0.24	0.24	0.23	0.24	0.23
Potassium oxide	K ₂ O	4.10	3.17	0.12	3.20	2.36	1.40
Phosphorus oxide	P ₂ O ₅	0.03	0.04	0.02	0.03	0.02	0.02
Sulfates oxide	SO ₃	0.04	0.13		0.09	0.20	0.22
	PH (25 °)	7.80	8.74	0.25	12.35	12.62	12.82

The main mineralogical constituents of the 2 soils are silica and alumina. These 2 elements constitute the structure of the 2 clays. It appears from the examination of the chemical composition of the 2 clays that they present a silicate with an aluminic tendency with a high content of SiO₂. The low content of CaO for AA indicates that this clay contains a low percentage of calcium carbonate (calcite); contrary to the clay of Bouda AB, the presence of calcite CaO is noticed. The ratio of Alumina / Silica tells us about the material's permeability to moisture. The greater this ratio, the greater the permeability [56,57]. In our case, this ratio is small for AA = 0.32 and for AB = 0.26, which implies that these clays have low permeability.

The ratio SiO₂/Al₂O₃ = 3.03 for AA and 3.86 for AB; this variation shows the existence of free Quartz in the clay fraction in elevated proportion [57-59]. In addition, the high content of oxides (MgO, K₂O, and Na₂O) compared to the contents of other oxides probably shows that the clay contains exchangeable cations such as Mg²⁺, K⁺ or Na⁺ [60]. Furthermore, it is noted that the percentage of SiO₂ for both clays is less than

60 %, which allows us to conclude that these soils tend to swell; it is noted that non-swelling clays have a silica content greater than 80 % [61]. In addition to these constituents, it is noted that there are average proportions of Iron for the AA soil due to the presence of geotite, hematite and iron cations in substitution of illite [62]. This explains its great capacity for water absorption and its high swelling capacity.

Geotechnical properties

Identification tests were performed on reworked samples. The results of the tests are shown in **Table 2**. According to Casagrande's abacus and the USCS (Unified Soil Classification System) classification, the AA soil is classified among the clayey and very plastic soils (CH); the AB soil belongs to the same plastic clayey soil groups. According to the results of Skempton Ac [63], we can get an idea about the predominant mineral in the clay matrix. Illite is the predominant mineral in the AA and AB samples [64]. Based on the interval of $0.75 < A_c < 1.25$, we can conclude that our 2 soils have regular activity. The values of the total specific surface area SST are deduced from the methylene blue test (Vbs) by the formula of [65]. The blue value is carried out according to the standard test NFP 94 - 068; the specific surface is high, around 0.172 km²/kg for AA and 0.129 km²/kg for AB, it presents minerals close to calcic montmorillonite and Illite according to Holtz and Kovacs [64] and Lautrin [66]. The cation exchange capacity calculated by the blue test indicates that the 2 mineralogical clays are classified as Illites according to the classification of [64,67-69] $10 < CEC < 40$; we are in the presence of kaolinites for the AB clay and Illites for the AA clay, according to Truche [62]. According to the results of the test of Calcium carbonate content, it is 3.78 % (for AA) and 8.65 % (for AB), which indicates that we are in the presence of clay or silt ($0 \% < \% \text{CaCO}_3 < 10 \%$), according to the classification of [70]. The work of Williams and Donaldson [71] confirms that the clay of Adrar has a very high swelling potential against that of Bouda, it has a high swelling potential. According to (the classification of soils based on the compression index Cc and the index of voids e₀ obtained by oedometric tests [72], the AA soil is classified among the very compressible soils, while the AB soil is moderately compressible.

Moreover, according to the classification of [73], based on the compression index Cc, the AA soil is classified as medium clay and the AB soil is stiff clay. **Table 2** also shows the results of permeability calculated by the odometer test. It can be concluded that AA clay is very impermeable compared to AB clay. Therefore, the clay maintained in the rest of our study is AA.

Table 2 Physical characteristics for clayey soil.

References	Norms	Denomination	AA	AB
[74]	NF P 94-054	Density of solids grains (g/cm ³)	28.7	27
[75]	NF P 94-051	Liquid limit, LL (%)	79	64.5
	/	Plastic limit, PL (%)	36.74	26.92
	/	Plasticity index (%) (PI = LL - PL)	42.26	37.58
	XP P 94-060	Shrinkage limit SL (%)	12.9	14.9
	/	Shrinkage index (%) (SI = LL - SL)	66.1	49.6
[76]	NF P 94-05	Percentage of elements smaller than 2 μm, F2 (%)	46	41
	/	Activity (PI/F2)	0.92	0.91
[77]	NF P 94-055	Organic matter content OM (%)	0.086	0.086
	ASTM D4373	Calcium carbonate content (%)	3.78	8.65
[78]	NF P94-068	Value of methylene blue (%)	8.2	6.16
	/	Cation exchange Capacity = (VBSx1000) /374 (m ² /g)	21.92	16.47
	/	Total specific surface area = 21VBS (m ² /g)	172.2	129.36
[79]	NF P 94-093	Max dry density at MOP (KN/m ³)	17.7	19.6
	/	Optimum water content (%)	20.68	12.41
	XP P 94-091	Void indices e ₀	0.637	0.380
		Compression index Cc	0.289	0.107
		Cc / 1+ e ₀	0.176	0.077
		Cv (cm ² /s)	0.171.10 ⁻³	0.720.10 ⁻³
		K (m/s)	4.10.10 ⁻¹⁰	1.07.10 ⁻⁹

Lime

The quicklime used for stabilization has a low concentration of oxide elements such as silicates SiO_2 and aluminates Al_2O_3 (see **Table 1**). According to the standard test NF P18-554-555, the apparent density of the treated lime has the value $\rho_{\text{app}} = 1.7 \text{ g/cm}^3$ and absolute $\rho_{\text{abs}} = 2.34 \text{ g/cm}^3$.

Determination of the percentage of lime

The initial lime consumption gives the minimum amount of lime for cation exchange or soil affinity Bell [80]. This induces, as a consequence, a significant change in the properties of the treated soil in the short term. The literature proposes several methods to estimate this amount of lime, such as pH, swelling pressure, change in plasticity, mineralogical composition, and free lime content in the short term. In our study, we used the pH. The results of the pH variation with the different percentages of lime additions are shown in **Figure 6**. The 1st method suggested by Eades and Grim [81] saying that activating the pozzolanic reaction between lime and soil requires a minimum PH of 12.4. Soil alkalinity increases with the addition of lime. The pozzolanic reaction increases with pH, contributing to have better flocculation. Expansive clay is subjected significant transformations in its structure when it is mixed with lime. The test for measuring the pH of untreated and treated clay at different percentages of additions was performed according to ASTM D 4972. Treating Adrar clay samples with lime increases the pH of the mixture. These results are in agreement with those of several researchers [81-93]. The pH increases from 7.8 for natural clay to 12.35 after adding 2 % lime and stabilizes from 12.62 to 12.82 for 4 and 6 %. According to this method, a minimum of 3 % of lime is required to obtain a pH value of 12.4, which corresponds to the Lime Fixation Point (LFP); [82,90,94-97]. The increase in pH values is ascribed to the decrease in the relative amount of clay mineral content in the treated sample (see **Table 1**), 2, 4 and 6 %) due to the reaction of lime with clay, allowing the consumption and dissolution of clay minerals and possibly the creation of new minerals with linking characteristics [82,83, 87,88,91,98-100].

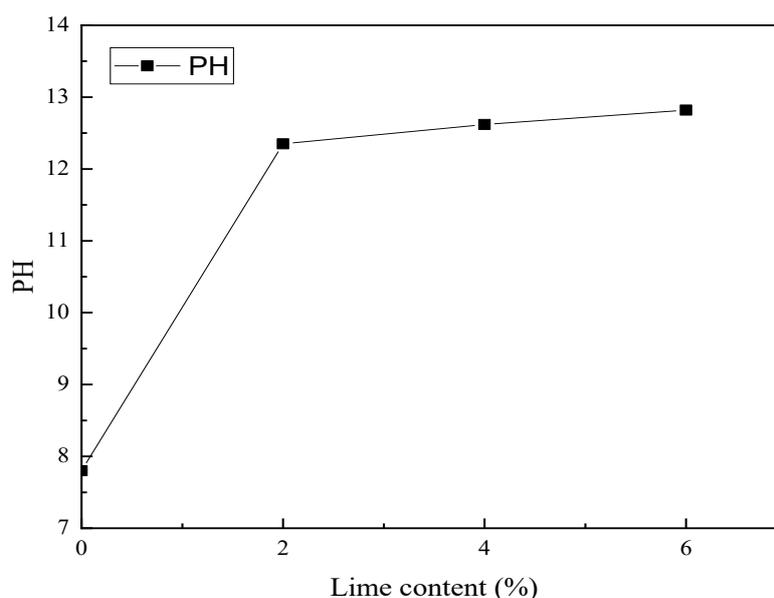


Figure 6 Effect of lime on pH.

The 2nd method suggested by Hilt and Davidson [101] is called the minimum percentage of lime or “lime fixation” or “lime fixation point L_m ” method. Which is the percentage of quicklime necessary to produce a saturated solution of lime in a clay-in-water suspension and thus fully satisfy ion exchange. Adding quicklime above this point is necessary to cause stabilization reactions [82,90].

$$L_m = [\text{clay content} (< 2 \mu\text{m})/35] + 1.25; L_m = 2.56$$

Considering the high efficiency of quicklime, an addition of 3 to 4 % was chosen. It was decided to treat the soil with 4 % lime based on the above methods.

Compaction tests and Atterberg's limits

The results of the modified Proctor test of lime-treated soil are shown in **Figure 8**. The curves show the relationship between maximum dry density (MDD) and optimum moisture content (OMC) of compacted clay mixtures with different lime contents. The OMC values increase with lime content and vary from 20.68 to 22.2 %; the MDD values of raw clay are much higher than those obtained for lime-treated clay. All the MDD values range from 17.7 to 17 KN/m³. This result is perfectly consistent with previous works [43,83,97,102-108]. This decrease can be explained by the fact that lime tends to absorb water to complete its hydration, as well as the pozzolanic reaction [20,80,96,108]. The increase in optimum water content is due to pozzolanic reactions between clay particles and lime in the soil; this process increases the water affinity of the soil. In addition, the immediate reactions are represented by flocculation and agglomeration due to the reorganization of clay particles in the soil into agglomerates. This trend allows for an increase in the value of OMC and a decrease in the values of MDD [55]. The magnitude of the change in these 2 parameters is related to the types of soil treated. The effect of lime content on the Atterberg limits is shown in **Figure 9**.

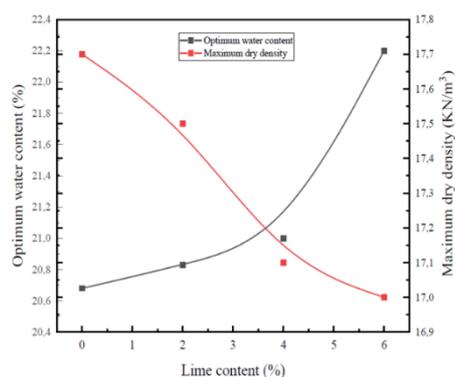


Figure 8 Variation of maximum dry density and the optimum water content versus lime of AA.

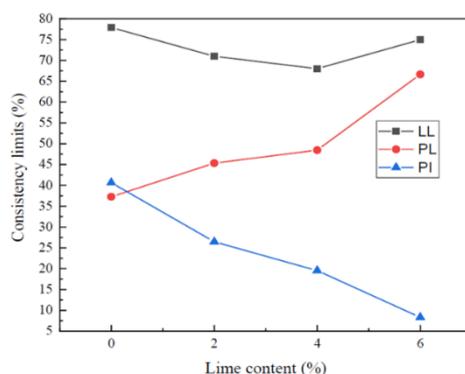


Figure 9 Effect of lime on Atterberg limits of and the optimum water content versus lime of AA stabilized clay.

It is noted that the addition of lime leads to a modification of the limits of Atterberg. It is noted as an immediate improvement of the workability and mechanical resistances, translated by a reduction of the plasticity marked by a lowering plasticity index (PI). This result seems identical to the data in the literature [43,81-83,93,102,105,107,109-112]. This decrease is due to a pozzolanic reaction that occurs when the pH of the soil solution becomes higher which is caused by the addition of (OH)⁻ ions to the soil [102,113]. Some authors note that the extent of plasticity changes depends not on the curing time but on the amount of lime added [93,114,115]. The reduction in plasticity can be ascribed to the increasingly granular nature of soils with lime [115].

Free swelling tests

The curves in **Figure 10** represent the relative variation of the free swelling height as a function of the logarithm of time ($\Delta H/H = f(\log(t))$) for the clay/lime mixtures. The course of the curve of the evolution of the free swelling in time is the same for all the mixtures; we distinguish 2 phases of swelling, primary

swelling, which occurs very early. Its end is marked by a point of inflexion, allowing for a modify in the course of the curve, and a secondary swelling characterized by a linear part of the curve [64].

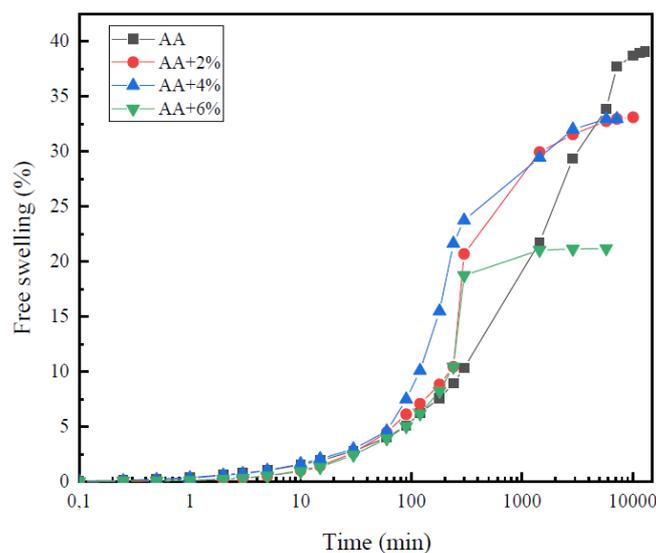


Figure 10 Free swelling evolution of untreated and treated samples versus the time.

Figure 11 shows the effect of different lime amounts on the swelling potential of treated samples. The figure shows that the expansive soil has a high swelling potential of 39.05 % untreated. The results obtained show that adding 2, 4 and 6 % lime reduced the swelling potential to 33.3, 32.95 and 21.35 %, respectively. This shows that increasing lime content significantly decreases the swelling potential of the expansive soil [43,83,102,105,106,109,110]. This decrease is mainly attributed to the reduction of the calcium-saturated clay's water absorption capacity due to the cationic exchange, which is more accentuated as the percentage of lime is essential. Also, it is due to the development of a cementitious matrix capable of resisting expansion [115,116]. These results agree with the Atterberg limit test values, which have shown that lime treatment of an expansive soil reduces the water absorption potential. This behavior can be ascribed to the pozzolanic reaction between lime (Ca^{2+}) and silica (SiO_2) that occurs after the separating silica from clay minerals. Overall, based on the swelling test results, it can be concluded that the stabilization of the expansive clay with lime will significantly improve the swelling properties of the clay. These results agree with the SEM micrographs' observations, which will be described in the following sections, **Figures 16(c)** and **Figures 12** shows the free swelling of the soil in its natural state. An increase in swelling equally to 7 mm in height is recorded, which confirms the hypothesis deduced from the chemical tests.

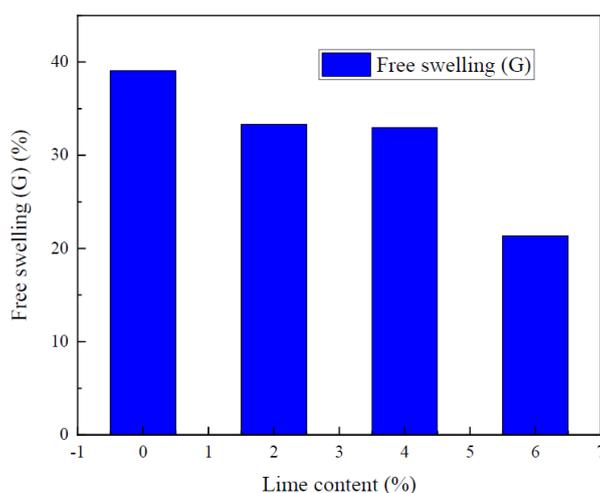


Figure 11 Improvement in free swelling due to mixing the clay with lime.



Figure 12 Free swelling for natural soil clay with lime.

Permeability coefficient

Indirect measurements of permeability

Figure 13 shows the odometer curves of compressibility of untreated and treated clay with different percentages of lime. Comparing these curves globally shows a difference in the void indices. From the consolidation curves shown in **Figure 14**, we can determine the compressibility index C_c and the swelling index C_s .

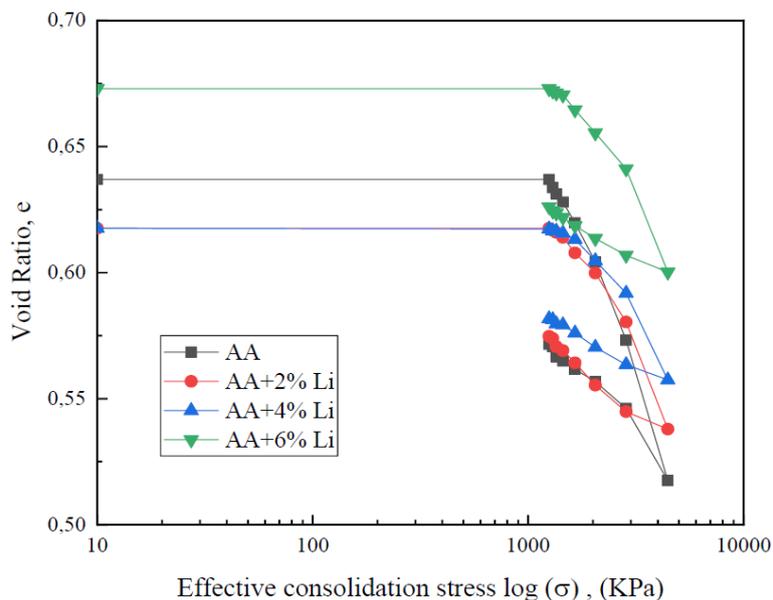


Figure 13 Compressibility curves versus lime.

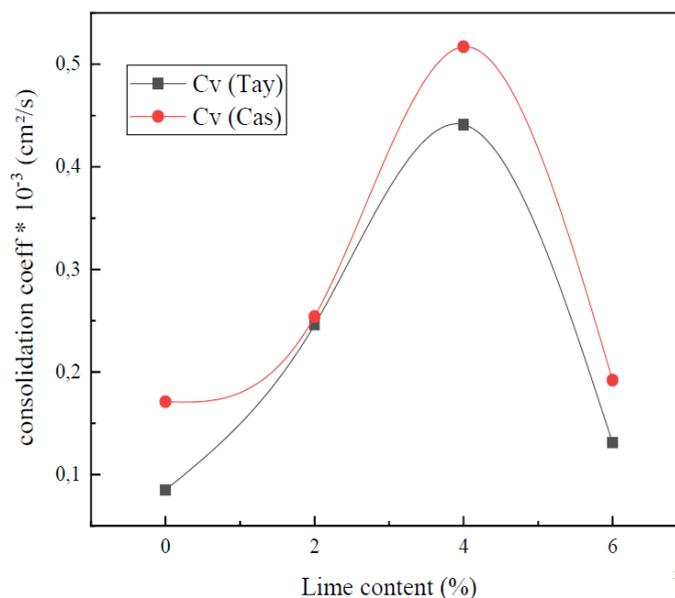


Figure 14 Cv by Taylor and Casagrande methods versus lime.

The variation of compressibility and swelling indices with the percentage of lime added are presented in **Table 3**. Improving the treated clay's mechanical characteristics is translated by low compressibility, equivalent to more resistance and less settlement and deformation following a loading. Similarly, the swelling index, which in turn undergoes a significant reduction, is mainly due to the cation exchange reaction. Indeed, the free calcium of the lime is exchanged with the adsorbed cations of the clay mineral, allowing the clay particles to move closer to each other, leading to a flocculation/agglomeration of the clay particles, which transforms the clay into a more granular material [83,114,115,117,118].

Table 3 Variation in compressibility index (Cc) and swelling (Cs) of untreated and lime-treated clay samples.

Designation	AA	AA + 2 % Li	AA + 4 % Li	AA + 6 % Li
Compression index Cc	0.3	0.22	0.17	0.2
Swelling index Cs	0.15	0.036	0.03	0.03

It can also be concluded, according to the classification of [73], that the AA soil is a medium clay, from which the mixtures can be described as stiff clays ($0.10 < C_c < 0.25$). The results of the consolidation tests are used to determine the coefficient of consolidation (Cv). This parameter was used to determine the permeability k. **Figure 14** shows the effect of lime on the coefficient of consolidation deduced by the Taylor and Casagrande method of treated and untreated clay. The values of the coefficient of consolidation for most fine soils are between 10^{-2} and 10^{-5} cm²/s. This range perfectly agrees with the results obtained for the Adrar clay, which is 10^{-3} cm²/s. According to this figure, the consolidation coefficient increases slightly with the proportion of lime included, which is in with the results of several researchers [83,94]. The increase in the consolidation coefficient is related to the processes of cationic exchange and flocculation/agglomeration that result from the reaction between lime and clay [95,117,119-121]. Also, the soil becomes more granular, which increases permeability [115,121,120]. Above 4 % of lime, the coefficient of consolidation underwent a reduction.

The results of the permeability variation are shown in **Figure 15**. The odometer test is carried out under vertical consolidation conditions, whereas the triaxial test is carried out under isotropic consolidation. **Figure 15** shows the saturated permeability values of the mixtures for different percentages of lime measured indirectly with a maximum vertical stress of 3,200 kPa by both the Taylor and Casagrande methods. It can be concluded that there is a slight increase in permeability up to 4 % of lime; after this interval, a decrease in permeability is noticed; The results obtained by the odometer tests (Taylor and Casagrande methods) are almost identical.

Direct measurements of permeability

The permeability variation of the clay mixtures before and after treatment with different percentages of lime measured directly is shown in **Figure 15**. The findings indicate that after the introduction of lime to the expansive soil, the permeability increases slightly, but after 4 % lime, there is a sharp drop in this hydraulic parameter in % (1.6×10^{-9} to 1.1×10^{-12} m/s⁴) in the case of lime addition from 0 to 6 %, respectively. As for the effect of the amount of lime added, according to [38,39,111,117,121,122] the permeability of soils treated with lime increases until the addition of a certain amount of lime corresponding to the Lime Fixation Point (LFP) which is the minimum amount necessary to promote pozzolanic reactions in the long term [121,101,81] beyond this threshold, the permeability of soils stagnates or even decreases. Explanations are put forward by Locat *et al.* [38], McCallister and Petry [39], Brandl [111], Nalbantoglu and Tuncer [117] and McCallister [122] to explain these variations. Thus, they suppose that when the lime content is lower than the (LFP), the increased permeability observed is linked to the less good aptitude of the soil to be compacted and thus to a progressive decrease in its dry density. In the case of lime dosages higher than the (LFP), they assume that the decreases in density are counterbalanced by the significant precipitation of cementitious products that block the circulation of water and thus reduce the permeability of the soil, contrary to Runigo [123]. According to McCallister and Petry [39] and McCallister [122], the permeability decrease phase is based on the increasing formation of cementitious products during the percolation time, which blocks the pores. The following authors concluded that with the curing time exceeding 7 days, the permeability of the stabilized soil decreases with the curing time [19,100,124,125].

Figure 15 also compares the results of the three methods (Taylor, Casagrande and triaxial). The results of the permeability tests acquired by the odometer tests are higher than those obtained by the triaxial cell in 6 % lime mixture, the permeability is vastly significant by the triaxial method (1.1×10^{-12} m/s²) than that of the Taylor method (2.45×10^{-10} m/s²). The target permeability values for contaminants have to range between 10^{-8} and 10^{-10} m/s, which can be obtained at a lime content of 6 %. A permeability value of fewer than 1.1×10^{-12} m/s under an applied vertical pressure greater than 250 kPa is a vital result worth noting.

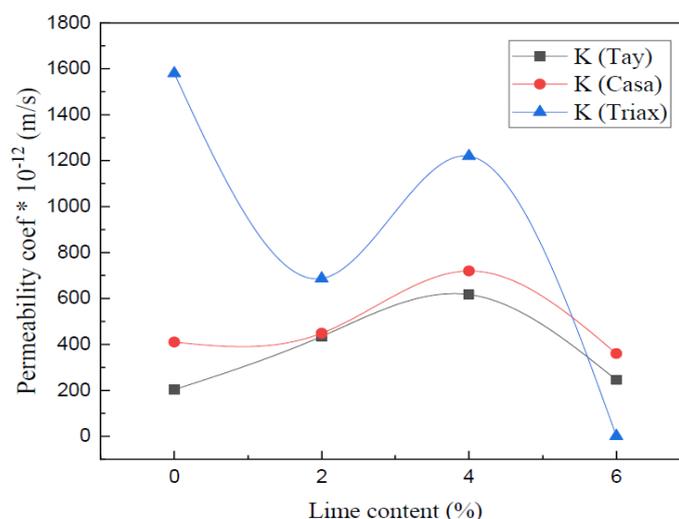


Figure 15 Permeability of clay-lime mixture (by Taylor, Casagrande, and triaxial methods).

In order to obtain an accurate result and to further evaluate the interactions between the lime particles and the clay particles in the 6 % mixture, SEM scanning electron microscopy tests were performed on the untreated raw **Figure 16(a)** and milled, **Figure 16(b)** and milled (6 %) treated samples **Figure 16(c)** and XRD test in **Figure 17**. From the SEM images in **Figure 16** and XRD in **Figure 17**, the soil becomes more granular with an increasing percentage of lime [112,115]. The formation of calcium alumina silicate hydrated (C-A-S-H) [38,80,81,87,108,115], which is mentioned by circle in **Figure 16(c)** and new peaks in the interval from 20 to 40 for **Figure 17** same results as [36,38] this formation is produced due to the pozzolanic reaction, which contributed to the improvement of mechanical properties [38,102,103,126,127]. The EDS of **Figures 16(d) - 16(e)** compares the spectra of the raw and treated clay at 6 % and we notice a reduction of the peaks attributable to the clay fraction (Al, Si, K). The variation in the percentage of the chemical elements obtained by the microanalysis results is noticed by a reduction of these elements and an increase in the percentage of Calcium.

The increase passage was 0.8 % for raw clay to 12.6 % for the mixture with 6 % of lime, same remark in [44,107,108].

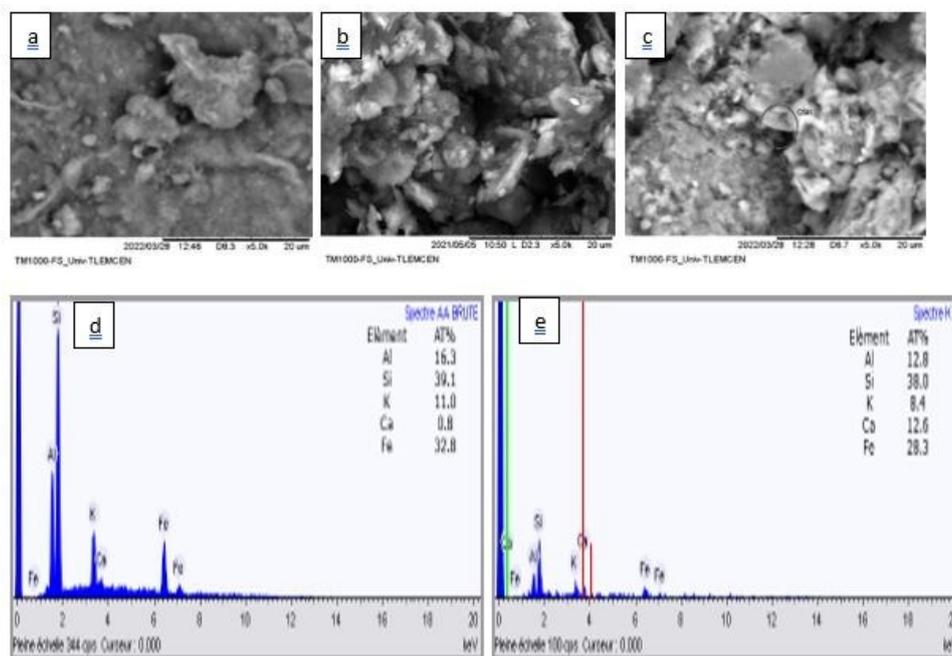


Figure 16 SEM and EDS test. a untreated clay unground (raw), b ground, c clay treated with 6 % lime, d EDS of raw clay, e EDS of clay treated with 6 % lime.

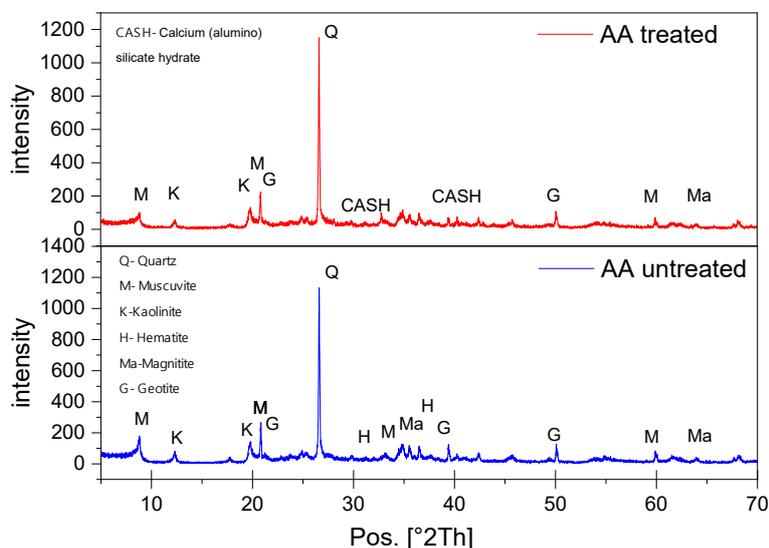


Figure 17 XRD test for untreated clay and treated with 6 % lime.

To show the significance of the results found in this research, **Table 4** summarizes the positioning of the permeability of the clay-lime mixture in previous work on other mixtures. This treatment had importance; hence, the research work was and still is updated and is scattered on all continents where we find diversity in natural or artificial additions. The value of permeability mentioned in the table of previous works varies between 4×10^{-9} and 4.58×10^{-11} ; in our case, we used a mixture of (94 % clay and 6 % lime) which gave a permeability of order of $1.1 \times 10^{-12} m/s$; comparing this value to other research gave an improvement rate of 98 %. Finally, this mixture is chosen because of its easy, economical application and because the primary component of this mixture (clay) is abundant in the Adrar region.

Table 4 Permeability comparison.

Author(s)	Year	City country	Optimal mixture	k-value s in m/s
[29]	2015	Tahran Iran	Bentonite and hexadecyl trimethyl ammonium hexadecyl trimethyl ammonium quaternary ammonium salt (HDTMA) Ordinary bentonite	$k = 5.2 \times 10^{-9}$ cm/s to crude oil $k = 9.56 \times 10^{-11}$ cm/s with water
[6]	2020	Kashan province, Iran	30 % sodium-bentonite and 70 % Sand	$k = 1.16 \times 10^{-11}$ m/s with phenol solution
[3]	2016	Maghnia (B) South Algeria (S)	15 % bentonite and 85 % Sand	$k = 4.58 \times 10^{-11}$ m/s à l'eau
[30]	2016	S: Eskisehir, Turkey Z: Balikesir, Turkey.	Mixture 30 % sepiolite in weight of zeolite (S/Z = 0.3)	$k = 1.17 \times 10^{-8}$ cm/s au mélange de cuivre (Cu) et de chrome (Cr)
[23]	2019	Al- Nahrawan, Iraq.	Soil and 6 % Sodium bentonite	$k = 6.83 \times 10^{-11}$ cm/s à l'eau $k = 7.8 \times 10^{-9}$ cm/s au pétrole brut
[71]	1981		Ranger Shale Fire clay Kosse Kaoline	$k = 38 \times 10^{-9}$ cm/s à l'eau $k = 13.5 \times 10^{-9}$ cm/s à l'eau $k = 220 \times 10^{-9}$ cm/s à l'eau
[21]	2019	Poland	Compacted lime softening sludge	$k = 4.0 \times 10^{-9}$ cm/s à l'eau $k = 6.8 \times 10^{-11}$ cm/s aux lixiviats municipaux
Our study		Adrar South Algeria	94 % clay and 6 % lime	$k = 1.1 \times 10^{-12}$ m/s à l'eau

Conclusions

This article aims to present a methodology based on the recovery of local materials in the region of Adrar for use in storage centers of hazardous waste. Tests were carried out on the influence of adding lime to a swelling clay according to several parameters, such as compaction, swelling potential, and permeability. The following results from this research:

- The addition of lime to clay soils leads to an immediate improvement in workability and mechanical strengths, which is reflected in a reduction of plasticity marked by a lowering of the plasticity index (PI), consequently, a substantial increase in the plasticity limit (w_p), a minor increase in the liquidity limit (w_L) were recorded. This reduction of (PI) is due to a pozzolanic reaction that occurs when the pH of the soil solution becomes higher which is caused by the addition of $(OH)^-$ ions to the soil.

- With the increase of the lime percentage from 2 to 6 %, the maximum dry density decreases, and the optimum water content increases. The reduction of the maximum dry density due to the reorganization of the clay particles is caused by the phenomenon of flocculation and agglomeration, while the increase of the optimum water content is due to the need for more and more water for hydration and pozzolanic reaction.

- The swelling potential decreases with increasing lime percentage. This is due to the reduction of adsorption. This gradual change in swelling potential is due to the pozzolanic reaction between lime (Ca^{2+}) and silica (SiO_2), alumina (Al_2O_3) that occurs after the separating silica and alumina from clay minerals,

requiring a longer time for the pozzolanic compounds to develop and bind the particles together and the formation of calcium alumina silicate hydrated of (C-A-S-H).

- The effect of a lime percentage added on the consolidation coefficient C_v deduced by the Taylor and Casagrande method increases slightly. This increase is related to cationic exchange and flocculation/agglomeration, which results from the reaction between lime and clay. Above 4 % of lime, the consolidation coefficient underwent a reduction. The results obtained by the odometer tests (Taylor and Casagrande methods) are almost identical.

- The variation of the permeability of the AA clay mixtures with different percentages of lime by the direct method (triaxial GDS) and indirect (Taylor and Casagrande) presents a slight increase of the permeability with the increase of the lime. After the 4 % of lime, we have a brutal fall in the permeability. This behavior can be explained by the permeability of lime-treated soils, which increases until a certain amount of lime is added, corresponding to the Lime Fixing Point (LFP), which is the minimum amount of lime necessary to promote pozzolanic reactions in the long term. Beyond this threshold, the permeability of treated soils stagnates or even decreases. This reduction is explained by the decrease in density, which is neutralized by the high speed of the cementitious products reducing water circulation in the soil and thus decreasing the permeability.

- From the SEM images and XRD tests, an increase in the lime percentage in the soil becomes more granular. The formation of hydrated calcium alumina-silicate of (C-A-S-H) is mentioned for the chosen mixture (AA + 6 % lime). It occurred due to the pozzolanic reaction, which contributed to improving the mechanical properties. The EDS illustrates the increase in the percentage of Calcium for the 6 % lime mixture and a decrease in the percentages of other constituents, such as Potassium. In conclusion, the flocculating agents Ca^{2+} and Mg^{2+} increase with increasing lime, while the deflocculating agents Na^+ and K^+ decrease with increasing lime.

Based on these results, we can validate the percentage of 6 % of lime to ensure the waterproofing with the good mechanical behavior of the waterproof layers of the hazardous waste storage centers. We recommend this economical local material combining clay and lime. This mixture can be used effectively in liner systems for industrial and municipal hazardous waste.

The results of this study provide an essential starting point for designing of an improved landfill liner. However, further field tests and trials should be required before this technology can be proven and implemented.

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