

Original Research Paper

Effect of Natural Pozzolan on the Stabilization of Two Swelling Soils from the Western Region of Algeria

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ABSTRACT

In recent times, a large number of studies have been carried out on swelling soils. It has been revealed that the swelling phenomenon, which still remains uncontrollable, is causing major disorders in several civil engineering structures around the world. The present study aims primarily to study the stabilization of two swelling clays. The first one comes from the region of M'zila that is located in the Wilaya (Province) of Mostaganem, while the second is from the region of Bourmadia in the Wilaya of Relizane. This article also presents the results of geotechnical investigations that were carried out on two swelling soils that were treated with natural pozzolan (NP) from the town of Béni-saf that is situated in north-western Algeria. The geotechnical parameters, such as consistency limits, as well as the proctor compaction test parameters and direct shear test parameters, along with the oedometer swelling test result analysis, were recorded and examined to evaluate the effects of adding different proportions of NP to these swelling soils. The results of the tests showed that several key properties of the above mentioned soils were significantly enhanced after the introduction of NP. For instance, the rectilinear shear test at the Casagrande box showed that the cohesion coefficients of Bourmadia and M'zila soils containing 10% NP improved by 93.3% and 47.1%, respectively. In addition, the results of the oedometer test to determine the swelling potential indicated that NP was capable of inhibiting the swelling of both clay soils. Overall, the outcomes suggest that NP from the region of Béni-saf has remarkable potential for improving the geotechnical properties of expansive soils that are often encountered during the implementation of many civil engineering structures and construction projects.

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INTRODUCTION

The phenomenon of soil swelling has been known for about as long as the field of geotechnical engineering has been practiced. As construction increased in arid and humid regions, problems associated with swelling of poor quality soils began emerging and gaining increasing attention. Subsequently, a large number of studies started focusing on expansive soils (Hachichi and Fleureau, 1999; Al-Rawas, 2006; Nowamooz and Masrouri, 2007; Srikanth and Sai, 2024; Tsiampousi et al., 2024). The swelling character of some clay soils is primarily associated with their mineralogical composition. Other factors, such as particle size, plasticity, organic matter content, salt (sulfate) content, and cation exchange capacity, can also affect, in a significant manner, the swelling pressure or deformation of clays (Djedid et al. 2001, Amri et al. 2019, Hamid et al. 2024).

Just like many other countries throughout the world, Algeria is fully concerned about the problem of swelling or expansive soils in different regions of the country. These soils have caused significant damage to numerous oil refineries and homes. Examples include the In Amenas hospital in the southeast of the country (Hachichi and Fleureau, 1999), several buildings in the Wilaya of Tlemcen (Belabbaci, 2014), in Ksar El Boukhari and Boughezoul in the Wilaya of Medea (Amri et al., 2019), as well as In N'gaous where several buildings and roads show several signs of degradation (Benyahia et al., 2020). It is widely acknowledged that the swelling phenomena are the cause of many disorders both for structures built on the surface (buildings, superficial foundations, retaining structures, embankments, etc.) and for underground structures (tunnels, piles, buried pipes, deep foundations, etc.). These soil swellings often cause significant disturbances that require very costly rehabilitation work; their implementation is generally not easily controlled (Hachichi and Fleureau 1999, Benyahia et al. 2020).

Several buildings were constructed on expansive soils in the region of Bourmadia, in the Wilaya of Relizane (North-West of Algeria). The numerous field trips and observations made on construction areas in Bourmadia indicated that the soil shrinkage-swelling phenomenon has led to enormous irreparable damage that is characterized by cracks in walls, subsidence affecting pavements and sidewalks, as well as differential settlements that generally result in very clear movements at the joints in blocks.

The stabilization of expansive soils is a branch of research that is attracting more and more interest among researchers in the field of geotechnical engineering. Moreover, many materials, which are a priori unusable, can be improved. This approach may help to avoid replacing the existing land with a noble material that is becoming increasingly rare. The significant financial implications of swelling have motivated researchers to define an efficient and economical stabilization process, while taking into account the nature of soil to be treated, the duration of the operation, the availability of the materials to be used, as well as the environmental conditions (Belabbaci, 2014).

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The improvement of soil properties by the addition of mineral binders, also called chemical soil stabilization, is a technique that was introduced so many years ago, with the main aim of making the treated soils capable of meeting the requirements of some specific technical projects. This approach is widely used in many applications in the field of civil engineering, such as the construction of embankments, railways and roads, foundations, the backfilling of bridge abutments and retaining walls, and others (Gueddouda et al. 2013, De Windt et al. 2014, Guidobaldi et al. 2017, Nishimura et al. 2017, Mrabent et al. 2017, Bakaiyang et al. 2021). It should be pointed out that this technique is beneficial in many ways. It actually makes it possible to:

- Improve the strength properties, including shear strength and compressive strength;
- Mitigate and reduce the volume and swelling potential, and control shrinkage, as well as diminish the plasticity index (Ip);
- Reduce permeability and compressibility of treated soils, as well as decrease soil deformation and settlement when the clayey and silt particle percentage declines;
- Enhance durability to withstand adverse environmental conditions such as freeze-thaw or wetting-drying cycles, erosion, and bad weather (Le Runigo et al., 2009, Al-Mukhtar et al. 2010, Cuisinier et al. 2011, Quang and Chai 2015, Liu et al. 2023, Elsayed et al. 2024, Yasir et al. 2024).

Nowadays, many binders are employed in soil stabilization. However, it has been reported that the stabilization technique that utilizes lime and/or cement is the most widely used. It is important to know that these stabilizing agents have their own drawbacks. Indeed, these stabilizing elements have some environmental impacts, such as greenhouse gas emissions, energy consumption during their production, high production costs, and depletion of natural resources (Guidobaldi et al. 2017, Mrabent et al. 2017, Por et al. 2017, Larouci et al. 2020, Hasan et al. 2023, Darapu and Vindula, 2024). In addition, although lime and cement have the ability to improve, to some extent, the engineering properties of soils, they can also have an adverse effect on some other properties. For example, these additives could cause swelling problems in the presence of sulfate. The replacement of these conventional stabilizing agents by new and less expensive products, with a low environmental impact, is considered an important issue today. Hence, several studies have been conducted in this connection by several researchers on the use of cementitious additives such as fly ash, silica fume, blast furnace slag, rice husk ash; cement dust, etc. ...(Vakili et al. 2016, Cheshomi et al. 2017, Indiramma et al. 2020, Almuaythir et al. 2023, Utkarsh et al. 2024, Hamed et al. 2024). Nevertheless, to the best of our knowledge, few investigations have been conducted to date to investigate the role of NP in soil stabilization.

It is widely known that natural pozzolans are rocks that are made up of volcanic projections (pyroclastites) with a scoriaceous and alveolar texture. Their formation may result from the rapid expansion of magma on the surface, accompanied by degassing or by an interaction between the magma and water. Natural pozzolans are different from basalts which have a more massive appearance. Indeed, pozzolanic materials are porous, and their rapid cooling, like quenching, causes the appearance of an amorphous phase (Senhadji et al. 2014). Algeria

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has a very abundant quantity of pozzolanic materials of volcanic origin which is modestly exploited by some cement plants in the west of the country as a cement additive. In this regard, several researchers have demonstrated the very high pozzolanic activity of NP coming from the town of Béni-saf, in northwestern Algeria (Senhadji et al. 2012, Chihaoui et al. 2016). They found out that the use of this finely ground volcanic rock as cement additive acts directly on the physicochemical properties of concrete. This pozzolan also offers many advantages, such as improving the durability of concrete, and reducing the quantities of gases emitted into the atmosphere during the production of clinker, and greenhouse gases (Senhadji et al. 2014, Elbar et al. 2018, Chihaoui et al. 2022). Working towards this same objective, it was shown that natural pozzolan has the potential to reduce GHG emissions when used for the stabilization of expansive soils.

The present contribution is highly interesting. It essentially aims to valorize local materials from the western region of Algeria using an experimental approach for soil stabilization in order to avoid the swelling phenomenon. Thus, the objective lies in highlighting the impact of natural pozzolan, which is a very abundant volcanic material in Béni-saf, a large town in the western region of our country, on the swelling potential of soils of Bourmadia (Relizane) and M'zila (Mostaganem).

In a first step, the materials used were identified and characterized by means of physical and classical geotechnical tests, and by chemical and mineralogical analyses. These different processes made it possible to establish a classification of the swelling potentials of each of these soils, using various approaches. Next, in a second step, this study sought to assess the effect of the different natural pozzolan percentages (0%, 2%, 4%, 6%, 8%, and 10%) on the consistency limits, the parameters of the compaction test and direct shear test, and the parameters of the swelling oedometer test.

2. EXPERIMENTAL PROGRAM

2.1 Materials used

2.1.1 Origin of the soils studied

The region of Bourmadia was first considered in this work. It is located in the southeastern part, about 6.60 kilometers from the center of the Wilaya of Relizane, as shown in Figure. 1. The geographical coordinates of this site are 35 ° 42 '34" North and 0 ° 34' 18" East. The soil sample under study was extracted from an urbanized site. It is important to note that the soil of Bourmadia has, in the recent past, been the cause of many very destructive disorders in several constructions. For this reason, it was decided to choose this soil for the purpose of studying its characteristics and its behavior during humidification, in order to propose the most adequate solution capable of reducing its swelling. It should be noted that the soil sample under study was taken at a depth of 2 m. It was also made sure that the sample was as homogeneous as possible, and all large rocky elements were eliminated. The color of Bourmadia soil is dark brown. The second region studied in this work is that of M'zila that is located 40 km northeast of the center of the Wilaya of Mostaganem in an area where the soil is

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known for its high swelling, as depicted in Figure 1. The geographic coordinates of this site are 35° 56′ 23.4168″ North and 0° 5′ 23.1612″ East. The surface area of this site is 369 Hectares. It is worth noting that the M'zila deposit belongs, from the geological point of view, to the Cheliff sedimentary basin. The soil sample was collected at a depth of 4.5 m, from numerous spots of the two sites, using a shovel. The main constituent is gray clay. After extraction, both soils were placed in plastic bags and transported to the laboratory. A series of geotechnical identification and characterization tests were then carried out.

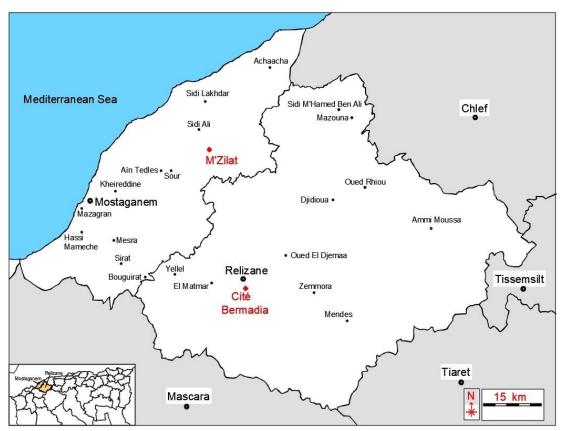


Fig.1: Geographic location of the soils studied.

2.1.2 Natural pozzolan from Béni-Saf

The natural pozzolan used in this study comes from the region of Bouhamidi that is 2.5 km from Béni-saf, in western Algeria. This region extends over a length of 160 km, from the coast of Oran to the Algerian-Moroccan border. The rock was thus extracted and then finely ground to obtain fine particles, with less than 80 μ m, after drying at 105 °C, as illustrated in Figure 2.

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Fig. 2: Natural pozzolan of Béni-saf

2.2 Experimental procedure

In the first part, the two expansive soils from the two sites of Bourmadia and M'zila, both in the western region of Algeria, were characterized with regard to their chemical and mineralogical composition, their physical characteristics as well as their geotechnical characteristics. The recorded data were used to establish a classification of the swelling potential of each of the two soils, using different identification approaches. It is worth specifying that the test methods were carried out based on the procedures and interpretation criteria specified in the AFNOR standards.

In addition to the soil identification tests and the determination of soil class, a second experimental program, intended for the stabilization of the two soils by the NP from Béni-Saf, was also initiated. This experimental program consisted in preparing mixtures of clayey soils/pozzolan (Bou-NP and Mzi-PN). The first step of this program was the treatment procedure which, first, consisted in drying samples of the two soils of Bourmadia and M'zila, in an oven at 60 °C, for 24 hours. Next, the dry material was carefully mixed, for about 15 minutes, with different percentages of NP (0, 2, 4, 6, 8 and 10%), according to the specifications of Standard NF EN 14227-1 (2005). The details of the different mixtures, which were prepared and tested, are summarized in Table 1. The mixing was achieved by means of a blade mixer, using distilled water when necessary, until a homogeneous material was obtained.

Several tests, such as the Atterberg limits tests, Proctor compaction tests, shear tests, as well as oedometer tests, were carried out on the different mixtures prepared in order to better appreciate the effect of treating these two soils with NP. The methods adopted in the tests, which were carried out on the different mixtures prepared, are briefly described while referring to the procedures and interpretation criteria that are specified in the AFNOR and ASTM Standards.

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Designation	% of clay soils	% of NP
Bou-NP0	100% Bourmadia soil	0%
Bou-NP2	98% Bourmadia soil	2%
Bou-NP4	96% Bourmadia soil	4%
Bou-NP6	94% Bourmadia soil	6%
Bou-NP8	92% Bourmadia soil	8%
Bou-NP10	90% Bourmadia soil	10%
Mzi-NP0	100% M'zila soil	0%
Mzi-NP 2	98% M'zila soil	2%
Mzi-NP4	96% M'zila soil	4%
Mzi-NP6	94% M'zila soil	6%
Mzi-NP8	92% M'zila soil	8%
Mzi-NP10	90% M'zila soil	10%

2.2.1 Atterberg limits or consistency limits

The Atterberg limits are essential geotechnical parameters that are intended for the estimation of the swelling potential of expansive soils. The Atterberg limits tests were carried out in accordance with the NF P 94-051 Standard for the initial untreated soils as well as for soils treated with different percentages of natural pozzolan (2, 4, 6, 8 and 10%). It should be noted that these tests were performed on the fraction of soil particles with a size less than 425 μ m. The test, as its name suggests, allows characterizing the transition between three different clayey soil states. The test results are then used to determine plasticity index between the liquid state (W_L) and the plastic state (W_P) . Then, the plasticity index (I_P) can be deduced from the two previous quantities (Equation 1):

Ip: plasticity index Ip =
$$W_L - W_p$$
 ...(1)

2.2.2 The standard Proctor test

The Proctor compaction characteristics of a material are determined from the normal Proctor test. The principle of the test consists in humidifying a material at several water contents and compacting it, for each water content, according to a conventional process and with a specific energy, as described in Standard NF P 94-093. The dry density of the material was determined for each of the water contents considered here, and the curve representing the variations of this density as a function of water content was plotted. This curve, which is generally called the Proctor curve, shows a maximum density value for the dry material; it corresponds to a particular water content value. These two quantities are then called the optimal Proctor compaction characteristics. The samples under study were then compacted in CBR cylindrical molds, with dimensions of 116.5 mm in height and 101.5 mm in diameter. These tests allowed determining the optimum compaction values (γd_{max} and ω_{opn}).

2.2.3 Shear strength test

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For the realization of the rectilinear shear box test, the same problem as for the oedometer test arose during the preparation of the sample in the raw state and after the mixing of the clayey soils with the different rates of NP. Indeed, it was not possible to insert the ring of the shear box in soil in the natural state (reworked) and, consequently, it was necessary to find for each test specimen, the same compaction degree in order to obtain the same initial state of the samples. To do this, the Proctor test results were then used. Hence, it was necessary to first perform a normal Proctor energy compaction in a Proctor mold of the twelve mixtures studied, with water contents equivalent to the Proctor optima ω_{opt} (section 2.2.2.). The sample was then extracted by means of an extractor and kept as is. Then, the shear box ring was inserted into the compacted soil sample. Afterwards, the samples were placed in the shear box.

This test aims primarily to determine the stability parameters of road embankment slopes of all types of soils, i.e. natural or reconstituted soils. To do this, the rectilinear box shear test was carried out on both soils (natural and treated soils), at different NP contents, from 0 to 10%. In addition, the so-called consolidated-undrained (CU) test was performed on unsaturated samples, after their compaction, under the Proctor optimum conditions. According to the NF P 94-071 Standard, the apparatus used in the test consists of two half-circular boxes of 6cm in diameter, in which the soil sample is placed. A piston located above the sample exerts a constant vertical force on it. The stability parameters provided are the cohesion (C) and the friction angle (φ) .

2.2.4 Swelling test

The oedometer test, which is initially intended for the study of compressibility, also allows measuring the swelling potential of soil. A series of oedometer tests were then conducted with the aim of performing direct swelling potential measurements. For this, the sample was placed in a cylindrical cell, between two porous stones. Next, as a result of flooding, the sample could swell vertically under the pressure of the piston for severaldays, until stabilization (Serratrice and Soyez.1996). Then, the relative variation of the volume of the sample (G), expressed as a percentage (%), could be found using Equation 2:

$$G\% = \frac{Hf - Hi}{Hi} \times 100$$
 ...(2)

- Hi: Initial height;
- Hf: final height after stabilization;

Furthermore, the constant-volume swelling method was adopted for the purpose of measuring the swelling pressure, according to the ASTM D4546-21 Standard. The tendency of the sample to swell was neutralized by the application of an increase in gload as soon as the comparator displacement reaches 1/100 mm. The value of the load, when the sample stabilizes, then gives the swelling pressure.

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3. RESULTS AND DISCUSSIONS

3.1 Identification test results

In general, in a geotechnical study, the rule is that the soils under study must first be identified, regardless of the objectives sought. The identification of materials consists of determining a set of their physical, chemical and mineralogical characteristics. This procedure makes it possible to guide subsequent geotechnical analyses and eventually to classify the materials encountered.

3.1.1 Chemical analysis results

The results of the chemical analyses of the most important elements, expressed in mass percentages of oxides, of the two clays from Bourmadia and M'zila, as well as those of natural pozzolan from Béni-saf, are summarized in Table 2. It is clearly noted that the three materials contain different types of oxides, in varying proportions. Indeed, a high silica content (SiO₂, 45.77%) was observed in the clay from Bourmadia, along with alumina (Al₂O₃, 9.98%), and ferrite (Fe₂O₃, 5.57%). In addition, a significant presence of calcium oxide (CaO, 6.52%) needs also to be mentioned. It is worth highlighting that the M'zila clay is mainly composed of silica oxide SiO₂ (55.95%), aluminum oxides (Al₂O₃, 12.82%), iron (Fe₂O₃, 5.69%) and calcium (CaO, 7.37%), with some small amounts of MgO, Na₂O, K₂O and SO₃. The ratio (silica/alumina) is around 4.38 for the Bourmadia clay and 4.58 for the M'zila clay. On the other hand, and as shown in Table 2, the natural pozzolan sample also contains significant levels of silica (SiO₂, 47.21%), and alumina (Al₂O₃, 18.85%). In addition, the presence of significant amounts of calcite (CaO, 10.84%) and ferrite (Fe₂O₃, 8.98%) is also noted.

Table 2: Chemical compositions of soils and natural pozzolan

	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	K ₂ O	Na ₂ O	Cl ⁻
Bourmadia soil (%)	45.77	9.98	5.57	16.52	2.51	0.07	2.07	0.45	0.014
M'zila soil (%)	55.95	12.82	5.69	7.37	2.81	0.43	2.32	1.32	0.02
Natural Pozzolan (%)	47.21	18.85	9.98	10.84	3.01	0.01	1.89	3.082	0.042

3.1.2 Mineralogical composition by X-ray diffraction

The main purpose of the mineralogical characterization, which was carried out by the X-ray diffraction technique, was to analyze the crystallization state of the materials, in order to better assess the nature of the different mineral phases present in the two clays investigated in this study. The identification of the soils examined in this study was carried out using the Bruker D8 Advance diffractometer analysis. This technique is highly advantage ous because it allows to directly knowing the crystalline structure of the clay minerals. The diagrams of the DRX analyses of the two materials are given in Table 3 and Figure 3. The recordings were made for the

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angle 2θ varying between 8° and 65° . The diagrams were obtained with a cumulative integration over a period of 1hour.

Close examination of the X-ray diffractograms of the two tested soils shows that these materials are composed of a significant amount of quartz, i.e. approximately 31.7% and 31.2% for Bourmadia clay and M'zila clay, respectively. The appearance of peaks related to the presence of quartz confirms the results obtained, with high levels of silica oxide. These findings were confirmed by chemical analysis similarly, the mineralogical analysis results of clays, which are presented in Table 3, clearly show that M'zila clay contains a higher level of illite (clay mineral, 21.12%), with the presence of other minerals such as calcite (15.15%) and microcline (10.27%). With regard to the Bourmadia clay, the second most dominant component is calcite (26.44%), followed by illite (14.68%), with the presence of other minerals, in small quantities, like kaolinite (clay mineral, 6.60%), dolomite (5.62%), and microcline (5.23%). These findings are in good agreement with the chemical composition values that were found with X-ray fluorescence spectrometry. It is useful to specify that the X-ray diffractometer analysis was not done for the NP brought from Beni-saf.

Table 3: Mineralogical compositions of the soils tested

Soils	Minerals(%)									
Sons	<u>Clay minerals</u>				Non-clay minerals					
	Illite	Kaolinite	Chlorite	Quarts	Calcite	Dolomite	Pyrophylite	Albite	Microcline	Topaz
Bourmadia	14,68	6,6	0,79	31,7	26,44	5,62	5,53	3,08	5,23	0,3
M'zila	21,12	4,96	2,02	31,2	15,15	1,9	8,96	3,65	10,27	0,76

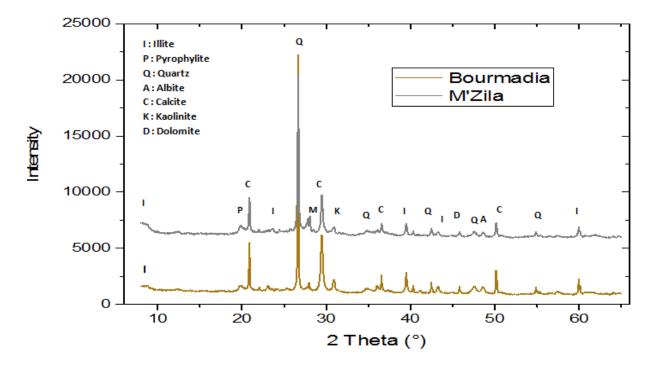


Fig. 3: XRD of the soils tested

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3.1.3 Physical and geotechnical analysis results

The identification test results are gathered in Table 4. Measurements of the density of solid grains (γ s), as determined by Standard NF P 94-054), indicate that the γ s value of Bourmadia soil is higher than that of M'zila. The difference is certainly due to the presence of a higher quantity of organic matter in the soil of M'zila.

Furthermore, the granulometric characteristics were obtained by wet sieving (Standard NF P 94-056) and by sedimentometry (Standard NF P 94-057). In addition, the size and distribution of the particles of both clayey soils are represented by the particle size curves shown in Figure 4. They show that the fine fractions, i.e. those less than 80 µm in size, are predominant, as they represent approximately 81.9% and 92.6% by mass, for the Bourmadia and M'zila soils, respectively. In this fine fraction, the clayey particles (less than 2 µm) are dominant; they represent 40.3% for Bourmadia soil, and 54.8% for M'zila soil. Additionally, the results of the Atterberg limit test allow noticing that the values of the liquidity and plasticity limits, as well as that of the plasticity index, of Bourmadia soil, are much lower than those of M'zila soil. The observed disparity may be explained by the difference between the contents and types of clay minerals, as well as the quantity of organic matter present in each soil, in accordance with Standard ISO 10694. According to the USCS/LCPC classification, from the Casagrande diagram, both Bourmadia and M'zila soils can be classified among very high plastic clayey soils (Ap).

Table 4: Parameters for identifying the soils studied

The parameters of the soils studied	Standard	Unit	Bourmadia soil	M'zila soil
Density weight of solid grains,γs	NF P 94-054	g/cm ³	2.45	2.23
Organic matter	ISO 10694	%	2.21	2.93
% particles < 2 μm	NF P 94-056 &	0/	40.3	54.8
% particles < 80 μm	NF P 94-057	%	81,9	92,6
Liquidity limit, W_L		%	65.50	126.36
Plasticity limit,W _P	NF P 94-051	%	28.06	51.42
Plasticity index,I _P		%	37.44	74.94
Activityof clay, Ac	$A_c = I_p/(P_2-10)$	/	0.93	1.36
Optimal water content,ω _{opt}	NE D 04 002	%	21.70	32.60
Maximum dry density,γd _{max}	NF P 94-093	g /cm ³	1.6	1.27
Methylene blue value,VB	NF P 94-068	g/100g	8.83	18.67
Total specific surface area,Sst	Sst = 20.83*VBS	m^2/g	184.82	390.80
Cation exchange capacity, CEC	NF X 31–130	$m_{eq}\!/100g$	55	65
USCS Classification	/	/	СН	СН

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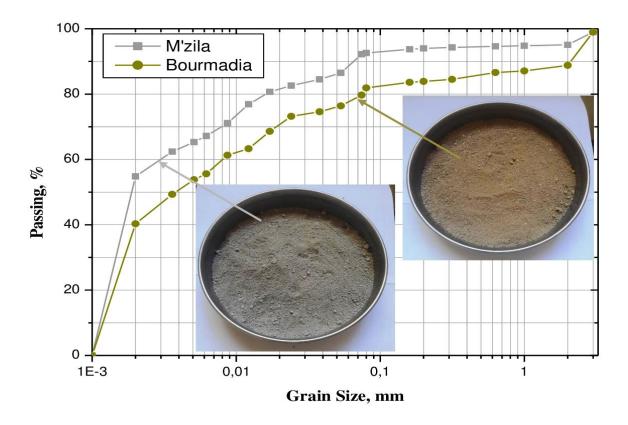


Fig. 4: Particle size analysis of the two soils

Furthermore, in 1953, Skempton defined the activity of clay (Ac) as the ratio of the plasticity index (Ip) to the percentage of elements smaller than 2μm. Then, based on the results obtained for Ac, it was concluded that the clayey soils of Bourmadia and M'zila have respectively normal and very active activities. Moreover, the values of the total specific surface area (Sst), which were deduced from the methylene blue test (NF P 94-068) using the formula given in (Lan. 1977), were found equal to 184.82 m²/g for Bourmadia clay and 390.80 m²/g for M'zila clay. These figures correspond to the low values of montmorillonite in the case of M'zila clay (300 m²/g < Sst < 800 m²/g), and to those of illite-type clays in the case of Bourmadia clay. Likewise, the other parameter that was evaluated in accordance with the AFNOR X 31-130 Standard is the cation exchange capacity (CEC). This parameter provides a good appreciation of the concentrations of compensating cations that can be exchanged with other cations that are generally encountered in clayey materials. It was then found that the CEC of Bourmadia clay was equal to 55meq/g while that of M'zila clay was 65meq/g. These values indicate that the cation exchange capacity of Bourmadia soil is lower than that of M'zila soil. However, they are both relatively low compared to those of pure montmorillonites which may exceed the value of 100meq/g. These results confirm the absence of this mineral in the mineralogical composition of the two clays.

3.2 Indirect estimation of swelling potential

Many empirical approaches have been proposed in the literature for the a priori assessment of the swelling potential of a soil, based on physical parameters such as the particle size, and also on some geotechnical parameters. Some of the methods employed to determine the swelling potential of clayey soils, which deserve to be

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mentioned and taken up in this work, are those that have been widely utilized by several authors and whose results generally converge on the qualitative prediction of swelling. Some of these methods are those that were proposed by Costet and Sanglerat (1981), Snethen (1980), Building Research Establishment in Taylor & Smith (1980), Williams et Donaldson (1980), Dakshanamurthy and Raman (1973) and Seed et al. (1962). The synthesis of the resulting outcomes, following the application of these methods, is presented in Table 5.

Table 5: Swelling potential of soils examined using the different methods proposed in the literature.

Method for estimating swelling potential	Ranki	ng Settings	Soils		
viction for estimating swelling potential	Kaliki	ng octungs	Bourmadia	M'zila	
Ghen (1988)	W_{L}	%<74 μm	High	very high	
Costet & Sanglerat (1981)	I_p	%<35 μm	Critical	very high	
Snethen (1980)		I_p	High	very high	
Building Research Establishment, (1980)	I_p	%< 2 μm	High	very high	
Williams & Donaldson (1980)	\mathbf{I}_{p}	%< 2 μm	very high	very high	
Dakshanamurthy&Raman(1973)	\mathbf{I}_{p}	\mathbf{W}_{L}	High	very high	
Seed etal. (1962)	Acor 1	%< 2 μm	High	very high	

¹The corrected activity: Acor = Ip/(P2-10)

It is observed that, in general, there is good agreement between the six methods of classifying the swelling potential of the two clays. Indeed, all six methods agree that Bourmadia clay has a high class swelling potential, while M'zila clay has a very high class swelling potential, with the exception of the Williams & Donaldson method which classifies Bourmadia clay in the category of clays with very high swelling potential. This last method tends to overestimate the swelling potential of Bourmadia clay. This observation is not surprising since these classifications are not based on the same experimental parameters.

All these results lead us to conclude that these are both swelling clays. Consequently, direct measurements of the swelling potential of any soil prove essential, from the preliminary reconnaissance campaign, when it is suspected that it is a swelling soil.

3.3 The effect of NP on the Atterberg limits

For the purpose of better assessing the effect of stabilization with pozzolanic additions on the consistency limits of Bourmadia and M'zila clays, it was deemed necessary to carry out some measurements of the liquidity and plasticity limits of the two soils that were treated with different percentages of NP from Beni-saf. It is useful to recall that the consistency limits are essential parameters in estimating the swelling potential of clayey soils. The results of the consistency limit tests are reported in Figures 5, 6 and 7.

At first glance, it is noted that the consistency limits of M'zila clay may clearly be distinguished from those of Bourmadia clay. Indeed, the results obtained allow observing that the liquidity and plasticity limits of M'zila

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clay are much higher than those of Bourmadia clay. The difference noted can be explained by the disparity between the contents and types of organic matter contained in the two soils.

3.3.1 Liquid limit

The liquid limit (W_L) is defined as the water amount that is contained in soil, which generally corresponds to the closure of groove lips over 1 cm, after 25 impacts with the Casagrande apparatus. Figure 5 shows the variations of the liquid limit of the two clayey materials, as a function of the content of NP used as a substitute. It is first noted that all the liquid limits of the (Mzi-NP) mixtures are largely higher than the liquid limits of the (Bou-NP) mixtures. The observed difference can be explained by the disparity between the contents and types of organic matter in each of the clayey materials. In this regard, $(Zentar\ et\ al.\ 2009)$ showed that the amount of organic matter in the soil affects parameters such as the liquid limit and plasticity of clayey materials.

Furthermore, the treatment with NP of the two clays under study, as clearly shown in Figure 5, reduces the liquid limit values of the (Mzi-NP) mixtures as well as those of the (Bou-NP) mixtures. It was indeed found that replacing 10% of Bourmadia and M'zila clays by the pozzolan of Béni-saf leads to a decrease in the liquid limit value of the (Bou-PN10) and (Mzi-PN10) mixtures by 21.2% and 34%, respectively. It can therefore be concluded that the decrease in the liquid limit value is due to the low sensitivity of the added natural pozzolan particles to water, which is not the case for clayey soils which present a very high sensitivity to water. In this regard, a large number of experimental studies, conducted on expansive soils, have indicated that the swelling rate of a soil decreases proportionally with the decrease in its liquid limit (Al-Rawas 2006, Allam 2011). Similar tendencies were also reported by (Muntohar and Hantoro. 2002) who used rice husk ash (RHA) which possesses some pozzolanic potential.

Figure 5 depicts the relationships between the liquid limit (W_L) values and the natural pozzolan content of the two clays studied. The evolution of the W_L values suggests a polynomial form, in accordance with Equations 3 and 4, for Bourmadia clay and M'zila clay, respectively. The coefficients of determination (R^2), which are respectively equal to 98.56 and 94.56%, indicate that a very good correlation exists between the W_L values and pozzolan content.

$$W_L = 0.08 \text{ NP}^2 - 2.1614 \text{ NP} + 65.107$$
, with $R^2 = 0.9856$...(3)

$$W_L = 0.6989 \text{ NP}^2 - 10.676 \text{ NP} + 122.97$$
, with $R^2 = 0.9456$...(4)

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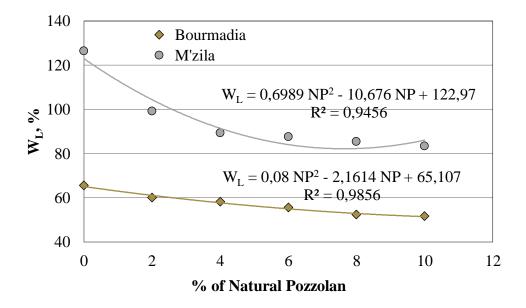


Fig.5: Evolution of the liquidity limit mixtures depending on the pozzolan content.

3.3.2 The plastic limit

The plastic limit (Wp) is the conventional water content of a soil roll that begins to crack as soon as its diameter reaches 3 mm. It is calculated from the arithmetic mean of the water contents obtained from 3 tests. The results of the Wp measurements for each of the Bou-NP and Mzi-NP mixtures are grouped and represented in Figure 6. In contrast to the liquid limit results, it is observed that increasing the NP content leads to an increase in the Wp values.

In this context, it is noticed that the (Bou-NP) mixtures showed a Wp increase of about 9.2% and 58%, for the (Bou + 2% NP) and (Bou + 10% NP) mixtures, respectively, compared to that of Bourmadia raw clay. Similarly, the (Mzi-PN) mixtures exhibited an increase of about 21% and 40.3% for the (Mzi + 2% PN) and (Mzi + 10% PN) mixtures, respectively, compared to that of M'zila clay. This development has caused a shift in the plasticity domain towards higher water contents. It should be noted that the shift in the plasticity limit is linked to the very high activity of natural pozzolan from Beni-Saf.

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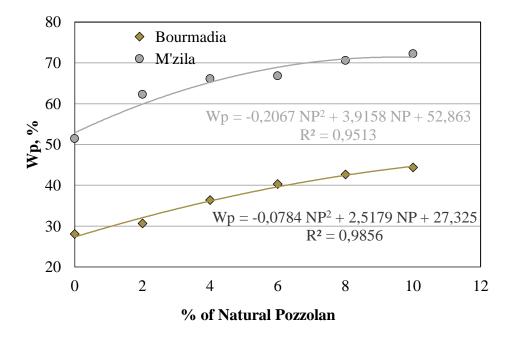


Fig.6: Evolution of the plasticity limits mixtures depending on the pozzolan content.

As soon as natural pozzolan is incorporated into a moist clayey soil, it starts acting on the electrical charges of the fine particles of the two clayey materials, and thus modifies the inter-particle electric fields, which leads to their flocculation and therefore results, geo-technically, in a significant increase in the plasticity limit of soil.

Similar results were obtained by (Sreekrishnavilasam et al. 2007) who found out that treating soil with cement kiln dust (CKD) contributes to increasing its plasticity limit. On the other hand, (Allam 2011) studied the stabilization of swelling clays by fly ash. The laboratory test results on these clayey soils indicated that fly ash is highly effective in improving the texture and plasticity of the treated soils.

Likewise, Figure 6 depicts the variation of the plastic limit measurements of Bourmadia and M'zila clays as a function of the natural pozzolan substitution rate. The curves obtained show that a polynomial correlation exists between the Wp values and NP substitution rate. It is in fact observed that the Wp values increase as the NP substitution percentage rises, according to Equations 5 and 6 given below:

$$Wp = -0.0784 \text{ NP}^2 + 2.5179 \text{ NP} + 27.325, \text{ with } R^2 = 0.9856$$
 ...(5)

$$Wp = -0.2067 \text{ NP}^2 + 3.9158 \text{ NP} + 52.863, \text{ with } R^2 = 0.9513$$
 ...(6)

It should also be noted that the correlation coefficient (R²) values, which were found to be equal to 98.56% for Bourmadia clay and 95.13% for M'zila clay, suggest that there is a very good correlation between the Wp values and the percentage of natural pozzolan added as a substitution to the two clayey soils.

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3.3.3 The plasticity index

The plasticity index (Ip) is defined as the interval between the liquid and plastic states. Plasticity is a property that characterizes very fine or clayey elements of soil. Figure 7 depicts the Ip values for both clays that were treated with different percentages of NP.

An instantaneous decrease in the plasticity index was then noticed following the addition of NP, especially in the case of M'zila clay. It was shown that an addition of only 2% of NP was sufficient to increase the workability of the clayey soil and reduce the plasticity index of the mixtures (Bou-NP2) and (Mzi-NP2), by 21.2% and 50.6%, respectively, compared to that of the raw clay.

Hence, the treatment of the two clayey soils with NP allowed reducing the plasticity of soils with an immediate plastic limit increase and a liquid limit decrease, which helped to obtain workable structures easy to compact.

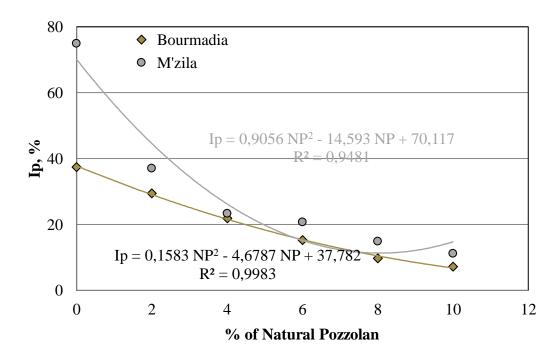


Fig.7: Evolution of the plasticity index mixtures depending on the pozzolan content

Furthermore, Figure 7 clearly illustrates the relationships between the reduction in the plasticity index values of Bourmadia and M'zila clays and the increase in the NP rate. These relationships are expressed by Equation 7 and 8. It is noteworthy that the correlations existing between the Ip values and the incorporated NP percentages are polynomial expressions. In addition, the R² coefficients, which are greater than 94%, indicate that a very good correlation exists between the thermal conductivity and the density of the composites.

$$Ip = 0.9056 \text{ NP}^2 - 14.593 \text{ NP} + 70.117, \text{ with } R^2 = 94.81\%$$
 ... (7)

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Ip =
$$0.1583 \text{ NP}^2 - 4.6787 \text{ NP} + 37.782$$
, with $R^2 = 99.83\%$... (8)

3.3.4 Effect of pozzolan on the classification of clays

Thorough examination of the Casagrande diagram allows observing the evolution of the plasticity index as a function of the liquid limit, for the different mixtures of the two untreated soils and those treated with NP at different percentages.

Figure 8 illustrates the change in classification undergone by both Bourmadia and M'zila clays following their treatment with NP. It is then observed that the two clays, which are classified as very plastic clays see, after treatment with NP, their consistency evolve towards very plastic silts for the (Bou-NP) mixtures, and very plastic silts for the (Mzi-NP) mixtures. This evolution of the consistency, along with soil stabilization with NP, causes flocculation and agglomeration of the particles of clayey soils. This phenomenon results in a change in the morphology of soil as the small grains come together to form other grains of larger size.

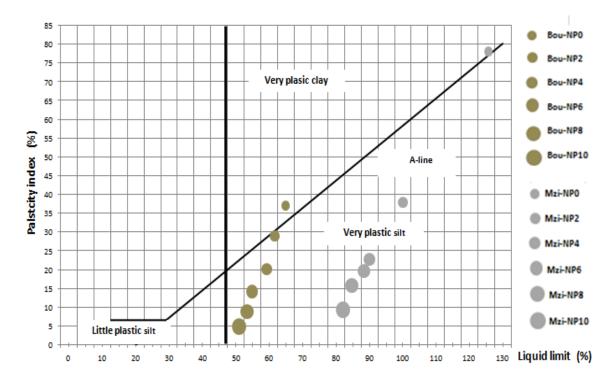


Fig.8: Result (W_L, Ip) of treated Bourmadia and M'zila soils placed in the Casagrande plasticity diagram.

3.4 Effect of natural pozzolan on compaction parameters

The Normal Proctor Test (NPT) was carried out, according to the NF P 94-093 Standard, with the aim of showing the effect of NP on compaction parameters, such as the maximum dry density and optimum water content. The evolution of these parameters, for the two clayey soils of Bourmadia and M'zila as a function of NP content, is depicted in Figures 9 and 10 respectively.

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First, the graphs in Figure 9 clearly show that the maximum dry density γ_{dmax} of all (Bou-NP) mixtures are much larger than those of (Mzi-NP) mixtures. The low value of γ_{dmax} of the mixtures (Mzi-NP), compared to those of the mixtures (Bou-NP), is certainly attributed to the low volumetric weight of the solid grains of M'zila clay ($\gamma_{s} = 2.23 \text{ g/cm}^{3}$) compared to that of Bourmadia ($\gamma_{s} = 2.45 \text{ g/cm}^{3}$).

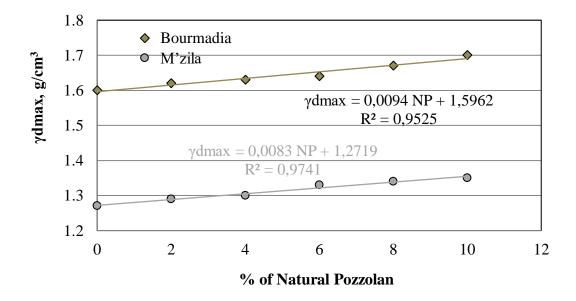


Fig.9: Evolution of maximum dry density of mixtures as a function of pozzolan content

In addition, Figure 9 allow noticing that the increase in the maximum dry density of the two soils treated with NP is proportional to the increase in the percentage of natural pozzolan added, for the same compaction energy. The maximum dry density (γ_{dmax}) of the mixtures (Bou-NP10) and (Mzi-NP10) increased by 6.25% and 6.3%, respectively, with respect to those of the raw clays (without pozzolan) from Bourmadia and M'zila. It is also important to understand that the increase in the maximum dry density is a good indicator of stabilization of the two soils under study.

Furthermore, the addition of NP to the fine grains of both clays leads to flocculation and agglomeration of particles due to cation exchange. This phenomenon results in an apparent change in the texture since the small grains come together to form other grains of larger size. Therefore, the flocculation of clay particles causes an increase in the effective grain size, which leads to density augmentation of the mixtures. It can therefore be concluded that flocculation plays a fundamental role in soil stabilization.

On the other hand, a similar behavior was observed by (Aref al-Swaidani et al. 2016) during the Proctor tests they conducted on soils treated with natural pozzolan in Saudi Arabia. In the same context, in 2019, Ziani reported that the increase in the maximum dry density of soil (Sand-Kaolin) is mainly due to the filling of soil pores by NP particles which have a relatively high specific density compared to the specific density of untreated soil.

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According to Figure 9, the relationships between the maximum dry density (γ_{dmax}) of the mixtures (Bou-NP) and (Mzi-NP) and the percentage of substitution of clays with NP show a linear trend (Equation 9 and 10). In addition, the regression coefficients (R^2), which are equal to 95.25% and 97.41%, for the mixtures (Bou-NP) and (Mzi-NP), respectively, indicate that a very good correlation exists between the maximum dry density (γ_{dmax}) and the amount of NP added.

$$\gamma d_{\text{max}} = 0.0094 \text{ NP} + 1.5962, \text{ with } R^2 = 95.25\%$$
 ... (9)

$$\gamma d_{\text{max}} = 0.0083 \text{ NP} + 1.2719, \text{ with } R^2 = 97.41\%$$
 ... (10)

In return, Figure 10 depicts the evolution of the optimum water content (ω_{opt} , %), which was measured on the mixtures of both clayey soils, as a function of the NP substitution rate. As opposed to the results obtained for the maximum dry density (γ_{dmax}), it turns out that the values of the ω_{opt} of the (Bou-NP) mixtures are lower than those of the (Mzi-NP) mixtures.

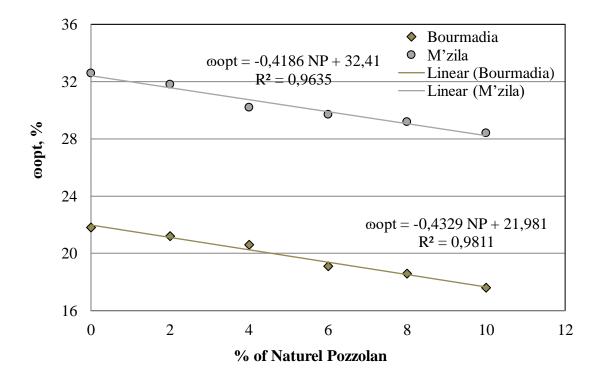


Fig. 10: Evolution of the optimum water content mixtures depending on pozzolan content

It was also noticed that the optimum water content of the two soils treated with NP decreased when the mass fraction of NP increased. The optimum water demand of the mixtures (Bou-NP10) and (Mzi-NP10) decreased by 19.3 % and 12.9 %, respectively, as compared to those of the raw clays (without pozzolan) from Bourmadia and M'zila. This decrease is certainly due to the very pronounced water absorption and retention mechanisms of the clayey soils, which is not the case for the NP from the town of Beni-saf.

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The correlations between the ω_{opt} values of the different (Bou-NP) and (Mzi-NP) mixtures and the soil substitution rate by NP are illustrated in Figure 10. It is observed that the ω_{opt} values decreased linearly as the amount of NP in the mixture went up. These correlations are expressed by Equations 11 and 12:

$$\omega \text{ opt} = -0.4186 \text{ NP} + 32.41, \text{ with } R^2 = 96.35 \%$$
 ... (12)

3.5 Effect of pozzolan treatment on shear strength

The mechanical behavior of soil can be analyzed, in a satisfactory way, using the shear strength test. In this study, the rectilinear shear test with the Casagrande box was carried out with the aim of determining the mechanical characteristics of two types of untreated clayey soils and others treated with 2, 4, 6, 8 and 10% of NP, under consolidated and undrained conditions (CU), with a shear rate of 1.5 mm/min.

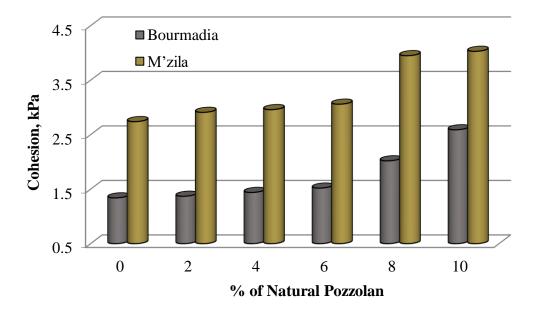


Fig. 11: Effect of adding NP on the cohesion of the two clay soils

Figures 11 and 12 illustrate, respectively, the histograms representing the evolution of shear parameters, such as cohesion (C) and the internal friction angle (ϕ), of the two clayey soils, as a function of the increase in the amount of NP added.

Figure 11 clearly shows that using NP, in partial substitution of each of the clayey soils, had a positive impact on the improvement of the shear strength of the mixtures (Bou-NP) and (Mzi-NP). It is observed that when the NP content in these mixtures increases, the cohesion coefficient also increases.

For example, the cohesion coefficients of the (Bou-NP10) and (Mzi-NP10) mixtures increased by 93.3% and 47.1%, respectively, with the inclusion of 10% NP, with respect to the coefficients of the mixtures (Bou-

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NP0) and (Mzi-NP0) (without NP). Based on these findings, it can therefore be concluded that the soil including NP opposes the sliding of the grains that compose it, and consequently resists better to the shear stress.

On the other hand, the results regarding the second shear parameter, i.e. the internal friction angle (ϕ) , of Bourmadia and M'zila clays mixed with NP, at different percentages, are shown in Figure 12. It is indeed seen that the graphical representation of the recorded data gives a bell shape. It suggests that, initially, when the percentage of NP in the mixture is increased, its friction angle (ϕ) also increases. Then, the curve reaches its maximum for a NP substitution rate of 4%. Beyond this value, as the NP substitution rate rises, the friction angle decreases slightly again but always remains higher than that of the untreated reference mixtures (Bou-NP0) and (Mzi-NP0).

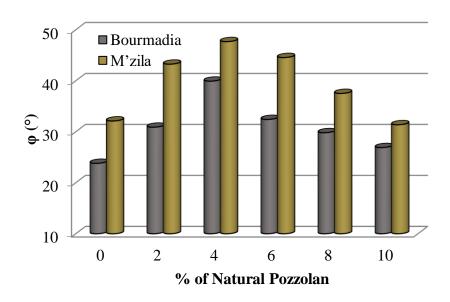


Fig. 12: Effect of adding natural pozzolan on the friction angle of the two clay soils

The above results can be explained by the fact that the treatment with NP changes the mineralogy of soil and, hence, produces new secondary minerals that positively influence the shear parameters. Therefore, using NP as a soil stabilization material seems to be an interesting solution. The approach adopted here offers important environmental advantages along with good technical performances. In this regard, (Zoubir 2009) found out that the cohesion increase is attributed to the cementation of clay particles, while that of the friction angle is assigned to the flocculation of these same particles.

These findings were later confirmed by (Sezer et al. 2006) who showed that the shear parameters of Izmir clay (very poor quality soil) could be improved following its treatment with 20% fly ash. However, (Zoubir 2009) revealed that the treatment of a clayey soil with a combination (lime + NP) resulted in a significant shear stress increase.

3.6 Measurement of swelling and swelling pressure by means of the odometer

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Once the swelling character has been authenticated in the preliminary reconnaissance phase, the swelling amplitude and pressure can then be assessed using a direct method, in order to better understand the effect of treating the tested soils by different NP percentages. Afterwards, a number of oedometer tests were carried out on a series of treated and untreated mixtures, according to Standard ASTM D4546-21.

3.6.1 Swelling potential according to the direct method (swelling amplitude)

For the purpose of studying the effect of NP on the free swelling of the soils under study, in accordance with the direct method, the samples were saturated with distilled water in an oedometer cell, and the swelling was then measured as a function of time until stabilization. The curves representing the evolution of the free swelling potential as a function of the logarithm of time, for the two mixtures (Bou-NP) and (Mzi-NP), which were prepared with different percentages of NP, are shown in Figures 13 and 14, respectively. It is then observed that, at the beginning of the tests, the swelling of all mixtures was low, but then, it started to increase and became quite significant over time.

The first slope, i.e. primary swelling, corresponds to a rapid hydration of soil particles by intrusion of hydrated cations. Next, an intense swelling of the osmosis-reinforced clay layer (secondary swelling) is observed. This second slope corresponds to the saturation of micro pores and inter-layer spaces. Then, the swelling rate decreases for a few days, until the state of stability of swelling is reached in the long term. Furthermore, it is noted that the trend in the variation of swelling potential, over time, remains unchanged for the two soil mixtures tested because, according to the graphs, when the percentage of NP increases, the swelling potential decreases. In addition, it seems that substantial swelling potential reductions can be achieved when the percentage of NP increases, for both types of soils. This is particularly true for the most swelling clay, i.e. M'zila clay, for which the swelling potential reductions can reach 31.4% and 40% for the (Mzi-NP8) and (Mzi-NP10) mixtures, respectively, after the 25th day.

Moreover, Figure 13 shows that the swelling rate of raw Bourmadia clay (untreated clay) is quite significant as it exceeds 13%, which implies a high swelling potential. However, it is noted that this percentage decreases proportionally with the NP percentage added, and may go down to less than 5% for samples treated with 10% NP, after only 17 days. Hence, the results obtained therefore lead to the conclusion that NP from Béni-saf is capable of inhibiting the swelling of both clays.

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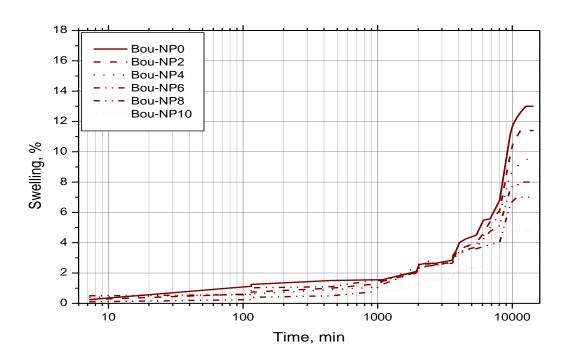


Fig. 13: Evolution of the swelling potential of mixtures Bou-NP versus time

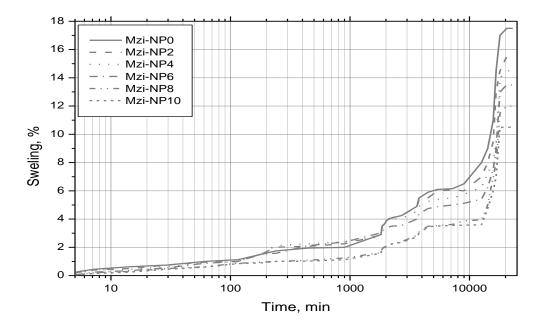


Fig. 14: Evolution of the swelling potential of mixtures Mzi-NP versus time

It should also be noted that these results are similar to those obtained by (Kalkan and Akbulut 2004) who showed that silica fume has a positive effect on treated clays. This is particularly manifested by a swelling

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potential decrease following the chemical reaction that occurred between clay and silica fume particles, which resulted in the production of calcium silicate hydrates (C-S-H).

3.6.2 Swelling pressure

The swelling pressure is defined as the pressure generated during the hydration of a sample under confined conditions. It may also be defined as the pressure that must be exerted to maintain a sample at its initial volume, before water adsorption (Vincent et al. 2006).

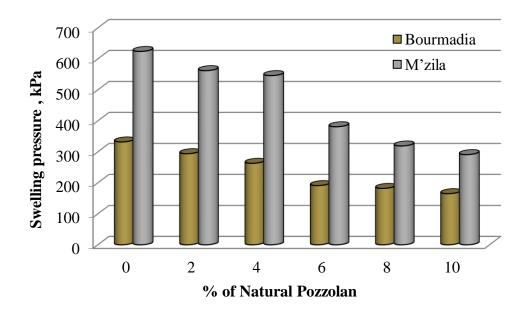


Fig. 15: Variation in swelling pressure of mixtures Bou-NP and Mzi-NP depending on the percentage of pozzolan used.

Figure 15 shows the swelling pressures of untreated soils and soils stabilized with NP. The swelling pressure values of untreated soils and those of soils treated with different pozzolan contents show variations similar to those of their swelling potential.

Furthermore, the results illustrated in Figure 15 indicate that the addition of NP to clayey soils contributes to improving their stability with respect to swelling as the amount of added pozzolan increases. It can also be observed that the swelling pressure significantly decreased when the PN replacement rate is greater than 6%, which confirms the effectiveness of NP in the stabilization of clay soils against the swelling phenomenon. It is also noticed that the swelling pressures decreased by about 50.3 % and 53.4% for the (Bou-NP10) and (Mzi-NP10) mixtures, respectively, as compared to the respective untreated corresponding soils. In addition, the effect of stabilization is slightly higher in M'zila soil which contains a clay proportion (54.8%) higher than that of Bourmadia soil (40.3%).

4. CONCLUSION

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The primary purpose of this paper was to improve the geotechnical characteristics of two expansive soils from two different regions of the western region of Algeria, namely Bourmadia soil from the Wilaya of Relizane and M'zila soil from the Wilaya of Mostaganem. For this, these two soils were treated with natural pozzolan that was brought from the town of Béni-saf in the Wilaya of Ain-Temouchent. Thus, both soils were treated with five different natural pozzolan percentages (2, 4, 6, 8 and 10%). The following conclusions were then drawn based on the results reported in this paper:

- First, the mineralogical identification allowed concluding that the two clays may belong to the mineralogical family of illite or kaolinite, with significant quantities of chlorite.
- The granulometry tests found a relatively high fines rate for both soils, with a small quantity of organic matter (less than 3%).
- The identification of the swelling character of the two soils under study was carried out using several empirical models available in the literature. These models used physical granulometric and geotechnical parameters easily measurable during preliminary tests. It is noteworthy that the empirical swelling models generally give comparable results. These models showed that the Bourmadia soil has a high swelling potential, while that of M'zila has a very high swelling potential, which can cause significant damage to buildings.
- The class of both soils, after treatment with NP, changed from CH to MH, according to the USCS classification system.
- The Atterberg limits of both clayey soils could considerably be modified by the addition of NP. Indeed, the liquid limits decreased considerably, while the plastic limits increased. This behavior resulted in a reduction in the plasticity indices when the percentage of NP increased.
- The maximum dry density of soils stabilized with NP increased as the NP content increased due to the flocculation and agglomeration of NP particles around the clay grains. On the other hand, the optimum water content decreased as the NP content went up. This was certainly due to the high water absorption and retention of the clayey phases contained in the soils under study.
- Both treated soils became more resistant to shear stresses. In addition, the addition of NP increased the friction angle (ϕ) and the cohesion (C) of the (soil + pozzolan) mixtures. The optimum values of cohesion were obtained for a NP content equal to 4%. This behavior can be explained by the considerable modification of soil structure following the addition of NP.
- The most important finding to report in this study regarding the stabilization of both soils with NP is that the swelling potential and swelling pressure reductions are proportional to the percentages of addition of this finely ground volcanic rock.
- Using natural pozzolan may be viewed as an interesting economic and ecological solution due to its low cost.
 This approach contributes to reducing the amount of greenhouse gases emitted in the atmosphere in comparison with other stabilization techniques, such as cement stabilization and lime stabilization.
- Finally, this work is of great interest to researchers in this field due to the evolution of the current socioeconomic context. Moreover, if the geotechnical report establishes that the bearing capacity of a land intended

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to accommodate a construction project is low, then the builder must remove the soil completely and replace it with another of better quality.

Author's contributions: Conceptualization: BENAISSA Mohamed and SENHADJI Yassine; methodology: BENAISSA Mohamed and SENHADJI Yassine; validation: SENHADJI Yassine; formal analysis: BENAISSA Mohamed and SENHADJI Yassine; investigation: BENAISSA Mohamed and SENHADJI Yassine; resources: BENAISSA Mohamed; writing and original draft preparation: BENAISSA Mohamed and SENHADJI Yassine; writing and review and editing: SENHADJI Yassine; supervision: SENHADJI Yassine.

All authors have read and approved the final version of the manuscript

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