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Effects of Na_2SO_4 on the geotechnical properties of clayey soils stabilised with mineral additives

Hamid Gadouri^{1,2*}, Khelifa Harichane² and Mohamed Ghrici²

An experimental investigation was undertaken to study the effects of sodium sulphate (Na_2SO_4) on the behaviour of two clayey soils stabilised with lime (L), natural pozzolana (NP) and their combination (L–NP). The geotechnical properties investigated are the Atterberg limits and unconfined compressive strength (UCS) on samples cured for 1–120 days curing period. The results show that the L and L–NP reduce the plasticity index (PI) and increase the UCS of two clayey soils. However, the PI of these soils increases when the Na_2SO_4 is present. In addition, higher UCS values are recorded with 2% Na_2SO_4 . But at later stage, the samples containing 4 and 6% Na_2SO_4 were completely deteriorated due to the formation of ettringite mineral. In general, the effect of Na_2SO_4 on the soil stabilisation process depends on the type of additive and its content, the soil nature, the Na_2SO_4 content and the curing period.

Keywords: Clayey soil, Natural pozzolana, Lime, Sodium sulphate, Atterberg limits, Unconfined compressive strength

Introduction

The chemical soil stabilisation using cement, lime and other additives is not new and remain so far the cheapest technique used in soils treatment (Ola 1977; George *et al.* 1992; Hossain *et al.* 2007; Harichane *et al.* 2012; Yi *et al.* 2015). For a short curing period, the stabilisation with lime is known by its immediate effect on the plasticity of treated soils (e.g. Rahman 1986; Okagbue and Yakubu 2000). But for a longer curing period, the mechanical properties of soil can be improved by developing pozzolanic reactions to form cementitious products such as calcium silicates hydrates (C–S–H) (for soils with a small amount of alumina) (Wild *et al.* 1993) and calcium aluminates hydrates (C–A–H) (for soils with a high amount of alumina) (Croft 1964) responsible on the increase in soil strength.

The chemical soil stabilisation using additives has found a better place in civil engineering field. The stabilisation of bad soils without going to appeal the good quality of materials leads to the significant reduction in transportation costs and time of project completion. Despite these advantages, the chemical soil stabilisation has a real problem due to the presence of sulphates in natural soils (Mitchell 1986). Indeed, the sulphates are present with a soluble form in the groundwater (SO_4^{2-} ions), or with a solid form in sedimentary grounds (gypsum, $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$; epsomite, $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$; arcanite, K_2SO_4 ; and thenardite, $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) (Wild *et al.* 1999). However, the pyrite (FeS_2) can give birth to a hydrated calcium

sulphate (gypsum) in the presence of certain conditions and after series chemical reactions (Floyd *et al.* 2003).

The different effects caused by different types of sulphates (sulphates present in the soil or added in the mixture) on the physico-mechanical properties of soils improved with various type of additives have been investigated by several researchers (e.g. Hunter 1988; Kinuthia *et al.* 1999; Sivapullaiah *et al.* 2000, 2006; Celik and Nalbantoglu 2013). The addition of L alone or in combination with other additives produces a beneficial effect on the engineering properties of stabilised soil. Indeed, the cation exchange capacity of soil is the origin of the negatively charged surfaces of their particles. These negative charges develop repulsive forces between clay particles. At early stage, the addition of L (CaO) in the soil with the presence of water produces the calcium hydroxide hydrate [$\text{Ca}(\text{OH})_2$]. The ionisation of L produces the calcium (Ca^{2+}) and hydroxyl (OH^-) when they are attracted to the surface of clay particles which reduces the repulsive forces and increases the adhesion between them to form flocks (Locat *et al.* 1990). This change made by the addition of L reduces the plasticity index (PI) of stabilised soil (e.g. Guney *et al.* 2007) and decreases their maximum dry density and increases their optimum moisture content (e.g. Rahman 1986). However, at later stage the increase in calcium concentration from the addition of L or cement increases the pH of soil (George *et al.* 1992). The high value of pH causes the dissolution of alumina (Al_2O_3) and silica (SiO_2) of soil, thereafter returning in a pozzolanic reaction with L added to form new cementing agents such as C–S–H, C–A–H and calcium aluminosilicates hydrates (C–A–S–H) (Mitchell 1986). These cementing agents increase the unconfined compressive

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strength (UCS) (e.g. Hossain *et al.* 2007) and shear strength of stabilised soil (e.g. Sezer *et al.* 2006) but decrease their potential of swelling (e.g. Afès and Didier 2000) and compressibility index (e.g. Nalbantoglu and Tuncer 2001).

The presence of certain types of sulphates in the stabilised soil affects greatly the stabilisation process by changing the cation exchange process and pozzolanic reactions (Hunter 1988), or even excludes certain types of soils to be treated and provokes the formation of expansive phases (ettringite) responsible to the damages made to the structures built (Le Borgne 2010). These damages depend to the mineralogical composition of stabilised soil (e.g. Sivapullaiah *et al.* 2000, 2006), and the additive content added and the test conditions such as the temperature of conservation (Le Borgne 2010). In addition, the effect of sulphates depends also to the type of the cation (Ca²⁺, Mg²⁺, Na²⁺ and K⁺) associated with sulphate ions (SO₄²⁻) (Kinuthia *et al.* 1999).

Several researchers were reported that the use of volcanic materials alone or in combination with L produces beneficial effects on soils improvement (e.g. Hossain *et al.* 2007; McCarthy *et al.* 2012). The natural pozzolana (NP) is found in abundance in areas of Beni-Saf located in the west of Algeria (Ghrici *et al.* 2007). This material was used in combination with L to improve the engineering properties of two clayey soils such as: Atterberg limits, compaction, durability, shear strength and UCS (Harichane *et al.* 2010, 2011a, 2011b, 2011c, 2012). However, there is no investigation of the sulphates influence on the engineering properties of these clayey soils. This work is devoted mainly to study the effect of Na₂SO₄ on the Atterberg limits and UCS of both grey soil (GS) and red soil (RS) stabilised with the addition of L, NP and L–NP.

Materials used and identification

Soils

In this study, two clayey soils were selected, the first is a GS obtained from an embankment project site located in the west of Algeria. The liquid limit (LL) and plastic limit (PL) values of this soil are 82.8 and 32.3%, respectively. The second is a RS obtained from a highway project site located in the same region. The LL and PL values of this soil are 46.5 and 22.7%, respectively. These soils were excavated, placed in plastic bags and transported to the laboratory for preparation and testing (Fig. 1(a)). The physico-mechanical and chemico-mineralogical properties of these soils are presented in Tables 1 and 2, respectively.

Mineral additives

In this study, the NP used is collected from Beni-Saf located in the west of Algeria. It is ground to the specific surface area of 420 m²/kg (Fig. 1(b)). However, the L used is a hydrated lime (Ca(OH)₂). It is commercially available lime typically used for construction purposes (Fig. 1(c)). The physico-chemical properties of these additives are presented in Table 3.

Chemical compounds

The Na₂SO₄ was used in this study (Fig. 1(d)). The physico-chemical properties of this element are shown in Table 4.

Test procedures and specimen preparation

Laboratory tests on plasticity and UCS were conducted on both selected clayey soils. Several combinations of NP and L were used for their stabilisation. These combinations were mixed with or without Na₂SO₄. A total of 72 combinations based on GS and RS is shown in Table 5.

Atterberg limits

Atterberg limits were performed according to ASTM D4318 (2000). The variations in LL, PL and PI of two untreated soils samples before and after admixtures added were studied.

UCS test

The UCS tests were performed according to ASTM D2166 (2000) and were conducted on both untreated and treated soil samples. The specimens were prepared with or without Na₂SO₄ by compaction at the maximum dry unit weight and optimum moisture content obtained from compaction tests.

Samples preparation

Soil–L, soil–NP and soil–L–NP mixtures

For both Atterberg limits and UCS tests, the air-dried soils were initially mixed with the predetermined quantity of NP (0, 10 and 20%), L (0, 4 and 8%) and L–NP in a dry state. On the one hand, the distilled water was added to the soil mixture for the Atterberg limits test. To let the water invade and permeate through the soil mixture, the samples are preserved in the airtight container for about 1, 15 and 30 days of curing period prior to testing. After curing, the paste obtained was remixed again with each stabiliser thoroughly for at least 15 min before performing the first test. The PL tests were performed on material prepared for the LL test. The PL was determined as the average of the two water contents. Both LL and PL tests were conducted at room temperature. The PI value is the difference between the LL and PL.

On the other hand, the calculated water was added to the soil mixture for the UCS test. The samples are preserved in the airtight container for about 1 h of curing prior to the preparation of specimens by static compaction using static press. Indeed, the obtained specimens were prepared by compaction at the maximum dry unit weight and optimum moisture content deduced of compaction tests. The specimens were stored in plastic boxes to prevent possible loss of moisture which they were kept in the laboratory at the temperature of 25°C and the relative humidity of 50%. Furthermore, after 7–120 days of curing, the specimens are tested. The tests of all samples were repeated on three identical specimens and the peak stress accepted was an average of three tests carried out on each sample type.

Soil–L–sulphate, soil–NP– sulphate and soil–L–NP– sulphate mixtures

For both Atterberg limits and UCS tests, the samples were mixed in the same way as presented above except that different contents of Na₂SO₄ powder (0–6% by weight of dry soil) were



1 Materials used and their preparation a RS sieved to 1-mm sieve, b NP rock ground in the laboratory in order to obtain a fine powder with specific surface area of $420 \text{ m}^2/\text{kg}$, c lime typically used for construction purposes and d Na_2SO_4

also added into the soil–L, soil–NP and soil–L–NP mixtures in a dry state. In addition, when the water (distilled water was used for Atterberg limits test) was added to the mixtures, the Atterberg limits and UCS tests were performed in the same way as presented above and tested after the same curing periods.

Results and discussion

Atterberg limits

Variation of the PI in the absence of Na_2SO_4

Figures 2(a) and 3(a) present the changes in the PI values of both GS and RS samples treated with L, NP and their combination

without Na_2SO_4 . In the case of the NP, the addition of this additive to both GS and RS samples produced a slight decrease in their PI. For example, with 20%NP as an additive the PI of the GS decreases from 50.5% to only 43.1 and 42.4% after curing for 1 and 30 days, respectively (Fig. 2(a)). As shown in Fig. 3(a), with of 20%NP the PI of the RS decreases from 23.8% to only 18.9 and 18.4% after curing for 1 and 30 days, respectively. For the same class soil, Yadu and Tripathi (2013) observed that the PI decreases from 17 to 13% for the addition of 12% of granulated blast furnace slag. Similar trends were observed by several researchers (Rahman 1986; Parsons and Kneebone 2005; Eberemu 2013; Sivrikaya et al. 2014). On the other hand,

Table 1 Physico-mechanical properties of both clayey soils (after Harichane et al. 2011a)

Physico-mechanical properties	GS	RS
Depth (m)	4.0	5.0
Natural water content (%)	32.90	13.8
Specific Gravity (-)	2.71	2.84
Passing 80-µm sieve (%)	85.0	97.5
Liquid limit (LL, %)	82.8	46.5
Plastic limit (PL, %)	32.2	22.7
Plasticity index (PI, %)	50.6	23.8
Classification system (USCS), (-)	CH	CL
Optimum moisture content (W _{OPM} , %)	28.30	15.3
Maximum dry density (γ _{dmax} , kN/m ³)	13.80	16.9
Unconfined compressive strength (UCS, kPa)	100	510
Loss on ignition (%)	17.03	7.13

Table 2 Chemico-mineralogical properties of both clayey soils

Chemical/mineralogical name	Chemical formula	GS (%)	RS (%)
Calcium oxide	CaO	14.43	2.23
Magnesium oxide	MgO	1.99	2.14
Iron oxide	Fe ₂ O ₃	5.56	7.22
Alumina	Al ₂ O ₃	14.15	19.01
Silica	SiO ₂	43.67	57.02
Sulfite	SO ₃	0.04	0.19
Sodium oxide	Na ₂ O	0.34	0.93
Potassium oxide	K ₂ O	1.96	3.17
Titan dioxide	TiO ₂	0.65	0.83
Phosphorus	P ₂ O ₅	0.18	0.14
pH	-	9.18	9.05
Calcite	CaCO ₃	26.0	4.0
Albite	NaAlSi ₃ O ₈	-	8.0
Illite	2K ₂ O·Al ₂ O ₃ ·- 24SiO ₂ ·2H ₂ O	16.0	24.0
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	12.0	16.0
Montmorillonite	Al ₂ ((Si ₄ Al)O ₁₀) (OH) ₂ ·H ₂ O	20.0	-
Chlorite	Mg ₂ Al ₄ O ₁₈ Si ₃	-	9.0
Ferruginous minerals	-	6.0	7.0
Organic matter	-	0.33	-

Degirmenci *et al.* (2007) observed that the PI increases with increasing fly ash content due to its small size particles developing a high surface area to compare with that of the NP.

However, with L as an additive, the workability of both GS and RS samples is improved due to the significant decrease in the PI values. The PI of both GS and RS samples decreases with increasing L content and curing period but, the decrease is more pronounced in the GS than in the RS. For example, as shown in Fig. 2(a), the addition of 8%L is sufficient to reduce the PI of the GS from 50.5% to 15.6 and 13.9% after curing for 1 and 30 days, respectively. However, the PI of the RS stabilised with the same content of L decreases from 23.7% to only 19.6 and 16.9% after curing for 1 and 30 days, respectively (Fig. 3(a)). For the same class soil, Afès and Didier (2000) reported that with 6%L the PI reduces from 23.7% to 10.3 and 8.4% after curing for 7 and 30 days, respectively. Similar observations were reported by several researchers (Ola 1977; Attoh-Okine 1995; Okagbue and Yakubu 2000; Bagherpour and Choobbasti 2003; Ansary *et al.* 2006).

Table 3 Physico-chemical properties of lime and natural pozzolana (after Harichane et al. 2011a)

Physical/chemical name	L (%)	NP (%)
Physical form	Dry white powder	Dry brown powder
Specific Gravity	2.0	-
Over 90 µm (%)	<10.0	-
Over 630 µm (%)	0	-
Insoluble material (%)	<1.0	-
Bulk density (g/L)	600-900	-
Loss on ignition	-	5.34
CaO	>83.3	9.90
MgO	<0.5	2.42
Fe ₂ O ₃	<2.0	9.69
Al ₂ O ₃	<1.5	17.5
SiO ₂	<2.5	46.4
SO ₃	<0.5	0.83
Na ₂ O	0.4-0.5	3.30
K ₂ O	-	1.51
CO ₂	<5.0	-
TiO ₂	-	2.10
P ₂ O ₃	-	0.80
CaCO ₃	<10.0	-

Table 4 Physico-chemical properties of Na₂SO₄

Physico-chemical properties	Sodium sulphate
Physical form	White
Chemical formula	Na ₂ SO ₄
Molar weight (g/mol)	142.04
Auauy (dried), (%)	99.5
pH (50 g/L, 25 °C)	5 to 8
Insoluble matter (%)	0.005
Chloride (Cl, %)	0.001
Iron (Fe, %)	0.0005
Calcium (Ca, %)	0.01
Phosphorus (PO ₄ , %)	0.001

The better decreases in PI values are achieved with the combination of both (L-NP). It is obvious to see that the combination L-NP has a significant effect on the PI of the GS than that of the RS. In addition, there is a considerable decrease in PI values with increasing L-NP content and curing period. For example, as shown in Fig. 2(a), the PI of the GS treated with 20%NP and 8%L decreases from 50.5% to 12.5 and 10.9% after 1 and 30 days of curing period, respectively. However, for the RS treated with the same contents of additives, the PI decreases from 23.8% to 14.8 and 11.1% after curing for 1 and 30 days, respectively (Fig. 3(a)). Ansary *et al.* (2006) reported that for a similar class soil, the PI decreases from 19 to 2.3% for the addition of 6% fly ash and 3%L. In all cases, the high reduction in PI values is observed for samples stabilised with the combination of both. This can be attributed to the complementary roles played by the L and NP where the beneficial effects of one can compensate for the disadvantages that could present another.

Variation of the PI in the presence of Na₂SO₄

The effects of Na₂SO₄ on the PI of both GS and RS samples stabilised with NP, L and L-NP are shown in Figs. 2(b-d) and

Table 5 Combinations of both clayey soils studied

Designation	Sample mixture (%)			
	Soil	NP	L	Sodium sulphate
P0L0N0	100	0	0	0
P0L4N0	96	0	4	0
P0L8N0	92	0	8	0
P10L0N0	90	10	0	0
P20L0N0	80	20	0	0
P10L4N0	86	10	4	0
P20L4N0	76	20	4	0
P10L8N0	82	10	8	0
P20L8N0	72	20	8	0
P0L0N2	98	0	0	2
P0L4N2	94	0	4	2
P0L8N2	90	0	8	2
P10L0N2	88	10	0	2
P20L0N2	78	20	0	2
P10L4N2	84	10	4	2
P20L4N2	74	20	4	2
P10L8N2	80	10	8	2
P20L8N2	70	20	8	2
P0L0N4	96	0	0	4
P0L4N4	92	0	4	4
P0L8N4	88	0	8	4
P10L0N4	86	10	0	4
P20L0N4	76	20	0	4
P10L4N4	82	10	4	4
P20L4N4	72	20	4	4
P10L8N4	78	10	8	4
P20L8N4	68	20	8	4
P0L0N6	94	0	0	6
P0L4N6	90	0	4	6
P0L8N6	86	0	8	6
P10L0N6	84	10	0	6
P20L0N6	74	20	0	6
P10L4N6	80	10	4	6
P20L4N6	70	20	4	6
P10L8N6	76	10	8	6
P20L8N6	66	20	8	6

3(b-d). In the case of treatment with NP as an additive, the presence of different contents of Na₂SO₄ produces a slight decrease in the PI values of both GS and RS samples. However, the PI of the GS samples treated with L increases with increasing Na₂SO₄ content and curing period. For the same class soil stabilised with 5%L, Celik and Nalbantoglu (2013) reported that the PI decreases from 32 to 15% in the presence of 2000 ppm of Na₂SO₄ but, increases up to 34 and 38% in the presence of 5000 and 10,000 ppm of Na₂SO₄, respectively. However, the PI of the RS samples treated with L decreases with increasing Na₂SO₄ content but increases with curing period. The modification in the PI values is the result of cation exchange processes which affect the viscosity of the clay–water mix. In addition, the interaction between two particles of clay soil is considerably affected by the cation exchange process because the increase in cation concentration results an increase in the distance between these clay particles. This promotes the increase in the clay particles size and affects the pores distribution due to the particles arrangement which leads to the change in the consistency limits of soils (Kinuthia et al. 1999). The large values of PI observed for L-treated both GS and RS samples containing Na₂SO₄ are probably due to the large surface area of their clay particles which develops a large storage capacity

of water governed the exchange of monovalent cations such as Na⁺ from Na₂SO₄ added.

Furthermore, the use of L–NP as an additive in the presence of any content of Na₂SO₄ affects deeply the PI of both GS and RS samples. As shown in Fig. 2(b–d), the PI of GS decreases with increasing L–NP content but increases with increasing Na₂SO₄ content and curing period. In contrast, the PI of RS decreases with increasing L–NP content and Na₂SO₄ content but increases with curing period (Fig. 3(b–d)). For example, in the case of the GS and after 30 days of curing period, the combination of 10%NP and 4%L increases considerably the PI from 16.3% up to 48, 51.8 and 58.2% in the presence of 2, 4 and 6% Na₂SO₄, respectively. However, for the same soil and the same curing period, the combination of 20%NP and 8%L increases the PI from 10.9% up to 26.7, 37.9 and 44.6% in the presence of 2, 4 and 6% Na₂SO₄, respectively (Fig. 2(b–d)). In addition, in the case of the RS and after 30 days of curing, the combination of 10%NP and 4%L increases the PI from 17.6% up to 32.4, 26.2 and 22.4% in the presence of 2, 4 and 6% Na₂SO₄, respectively. However, for the same soil and the same curing period, the combination of 20%NP and 8%L increases the PI from 11.1% up to 32.9, 28.4 and 22.9% in the presence of 2, 4 and 6% Na₂SO₄, respectively (Fig. 3(b–d)).

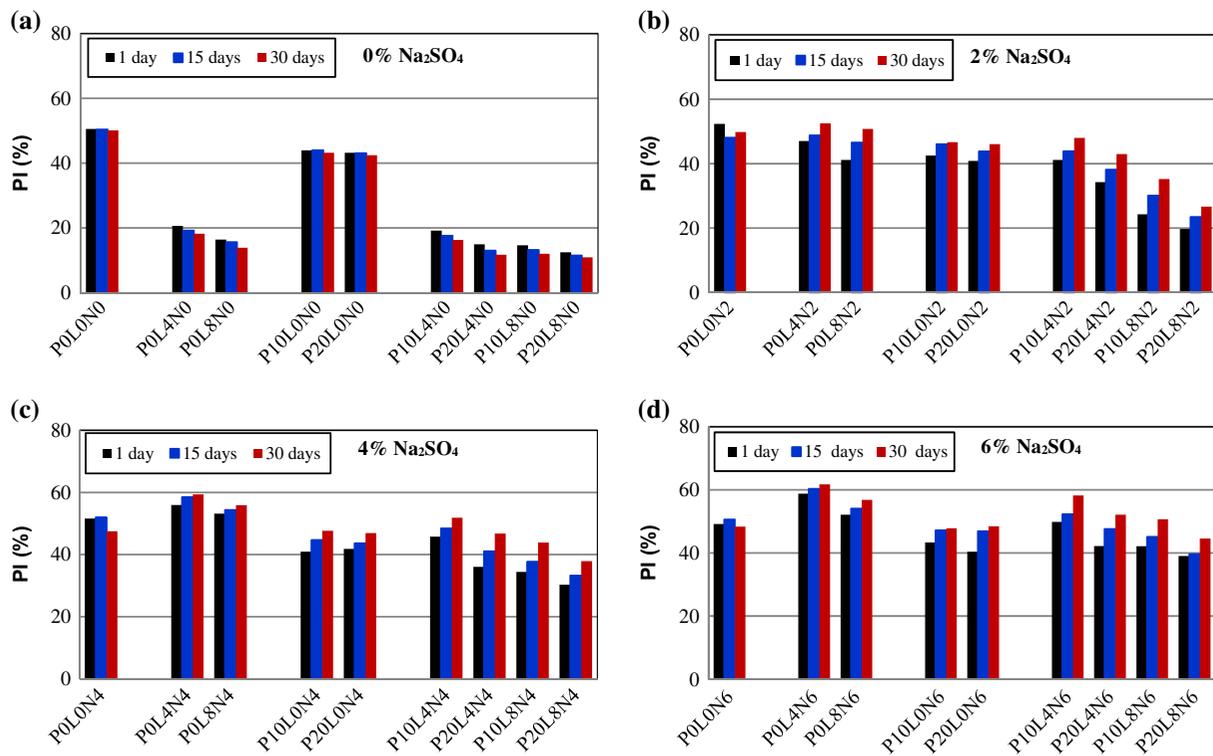
It should be noted that the presence of Na₂SO₄ produces a little effect on the PI of RS but causes an undesirable effect on that of the GS. Generally, the improvement of the consistence of both GS and RS samples depends on the type of additive and its content, the Na₂SO₄ content, the mineralogical composition of soil and the curing period. It is indispensable to take into account the effects of the presence of Na₂SO₄ on both the soil classification and stabilisation process for all soils used in civil engineering projects.

Unconfined compressive strength

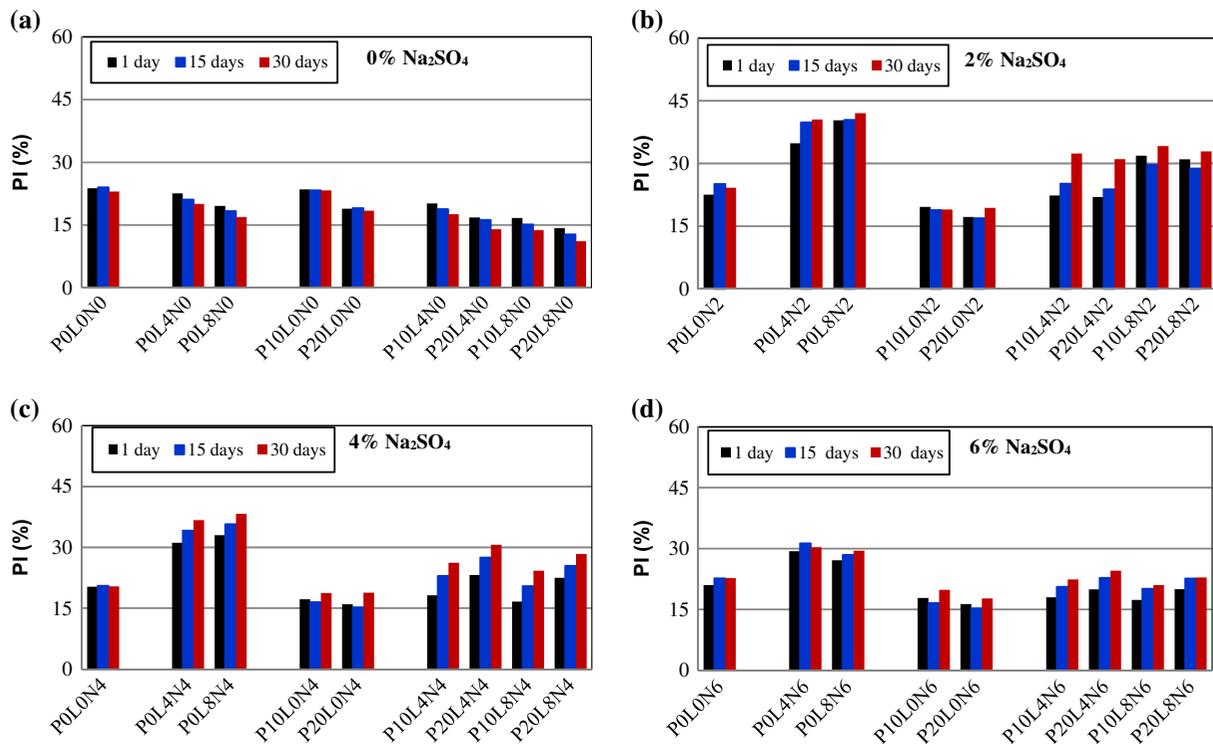
Variation of the UCS in the absence of Na₂SO₄

Figures 4(a), 5(a) and 6(a) illustrate the results of the effect of L, NP and L–NP without Na₂SO₄ on the UCS of both GS and RS. The addition of L alone to both GS and RS samples binds their particles and produces a significant increase in the UCS which increases with increasing L content and curing period. A similar behaviour was observed by McCarthy et al. (2012). In addition, Asgari et al. (2015) reported that the UCS of the soil obtained from north-west of Arak city increases with curing period and L content up to 3% but decreases after this content. The increase in strength is due to the formation of cementing compounds binding the soil particles which is the result of the L reaction with the clay particles (Harichane et al. 2012). However, there is a negligible increase in UCS values of both GS and RS samples when the NP is used alone due to its low reactivity with clay particles. Therefore, it is not possible to use the NP alone for the stabilisation of these soils. The differences in the UCS between L and NP are more pronounced with the RS than with the GS. This behaviour is probably due to the mineralogical composition and high PI value of the GS to compare with the RS.

It can be seen that the better results of UCS are achieved when the L and NP are combined. However, the UCS of both GS and RS samples increases considerably with curing period and L–NP content. For example, as shown in Fig. 4(a), the



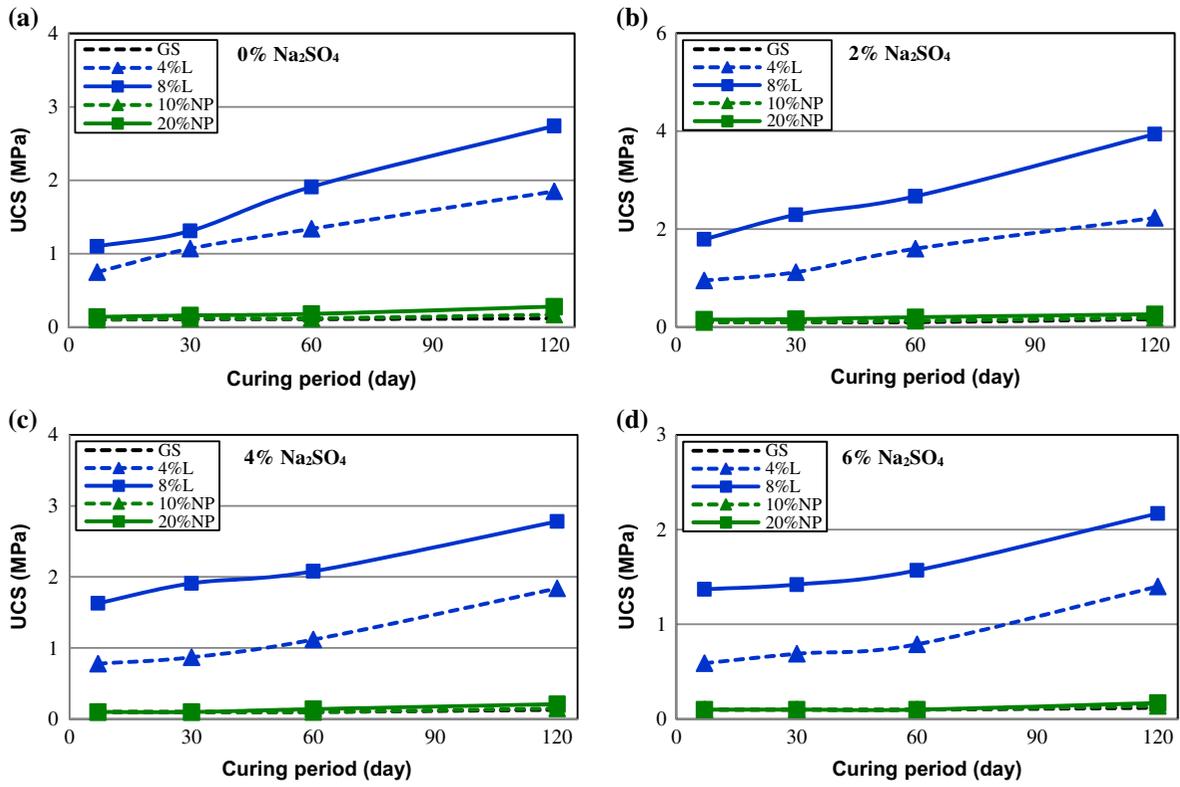
2 Effect of different contents of Na_2SO_4 on the PI of GS samples treated with L, NP and their combination



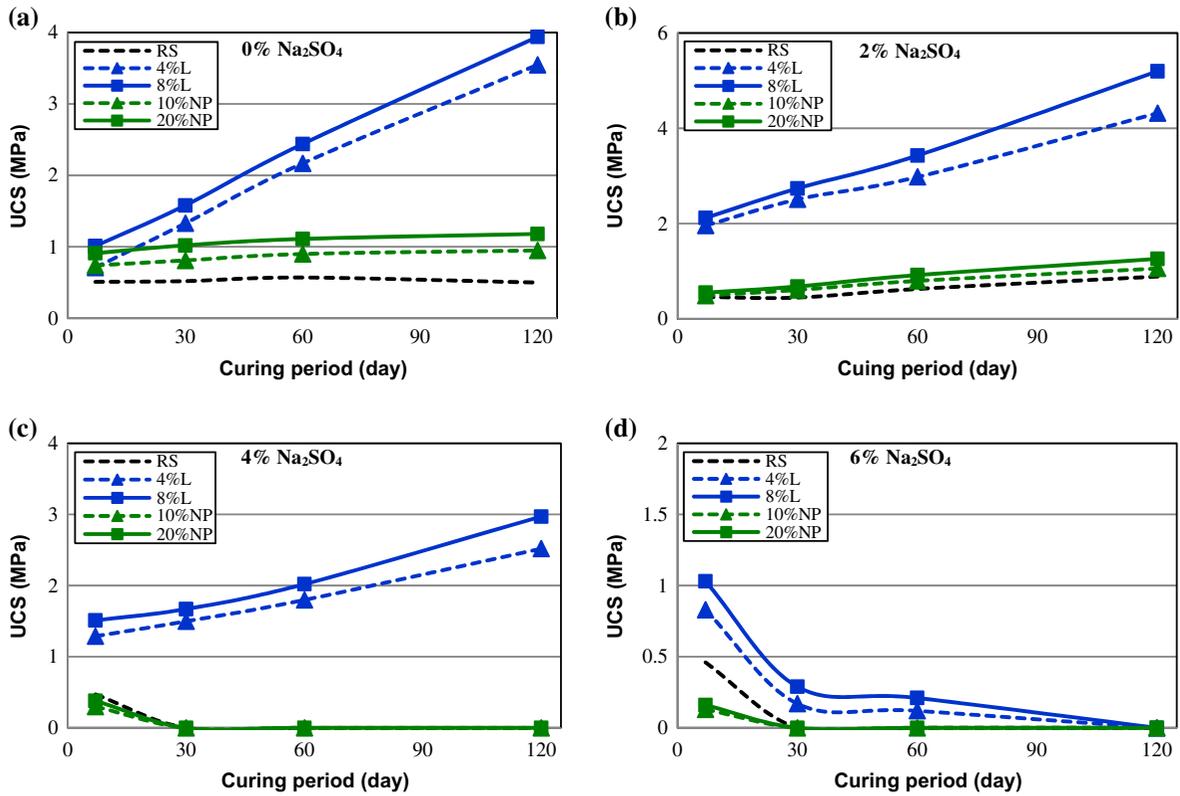
3 Effect of different contents of Na_2SO_4 on the PI of RS samples treated with L, NP and their combination

UCS of the GS treated with the combination of 20%NP and 4%L increases from 0.1 MPa up to 1.3 and 3 MPa after curing for 7 and 120 days, respectively. For a similar class soil treated

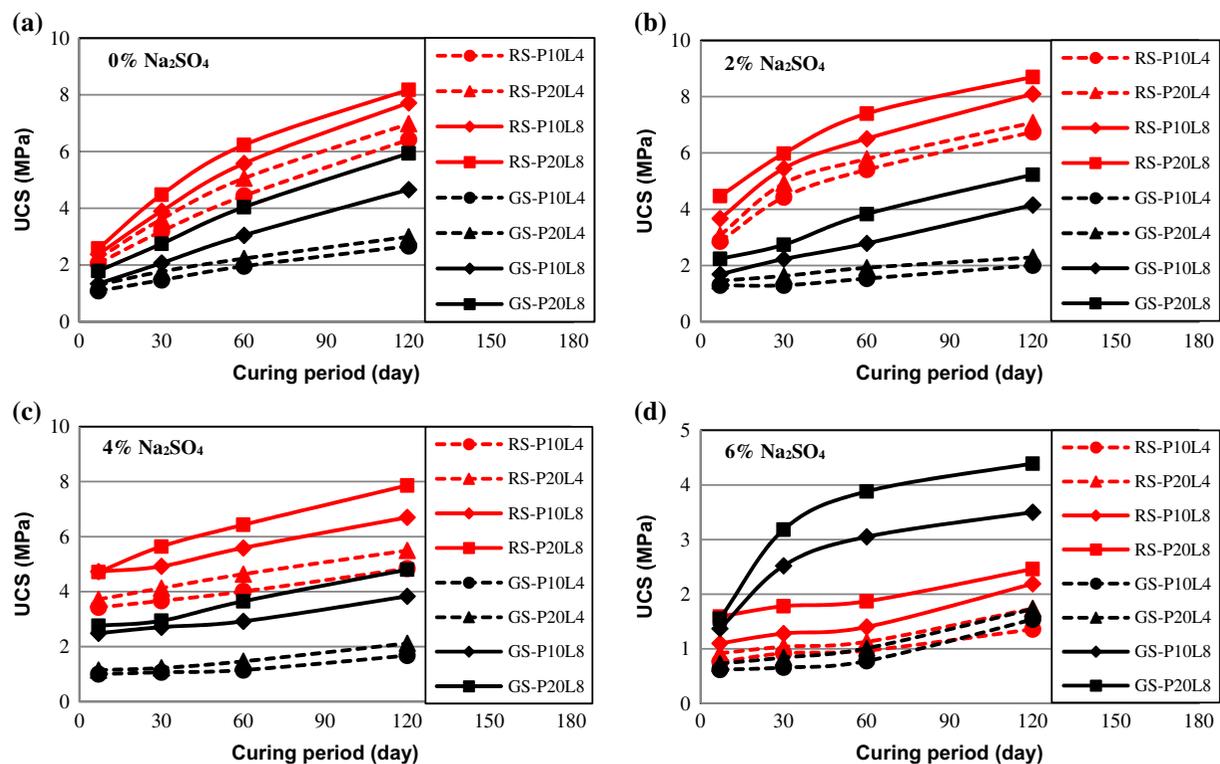
with a combination of 18% fly ash and 3%L, McCarthy *et al.* (2012) observed that the UCS increases from 0.4 MPa up to 1 and 1.8 MPa after curing for 7 and 90 days, respectively. A



4 Effect of different contents of Na₂SO₄ on the UCS of GS samples treated with L and NP alone



5 Effect of different contents of Na₂SO₄ on the UCS of RS samples treated with L and NP alone



6 Effect of different contents of Na₂SO₄ on the UCS of both GS and RS samples treated with the combination of L and NP

similar behaviour for a similar soil was observed by Kumar *et al.* (2007).

In the case of the RS treated with the combination of 20%NP and 4%L, the UCS increases from 0.5 MPa up to 2.2 and 7 MPa after curing for 7 and 120 days, respectively (Fig. 5(a)). For a similar class soil treated with the combination of 18% fly ash and 3%L, McCarthy *et al.* (2012) reported that the UCS increases from 0.4 MPa up to 1.1 and 2.1 MPa after curing for 7 and 90 days, respectively. This difference shows that the combination of L with NP develops high UCS values than the combination of L with fly ash especially for a longer curing period.

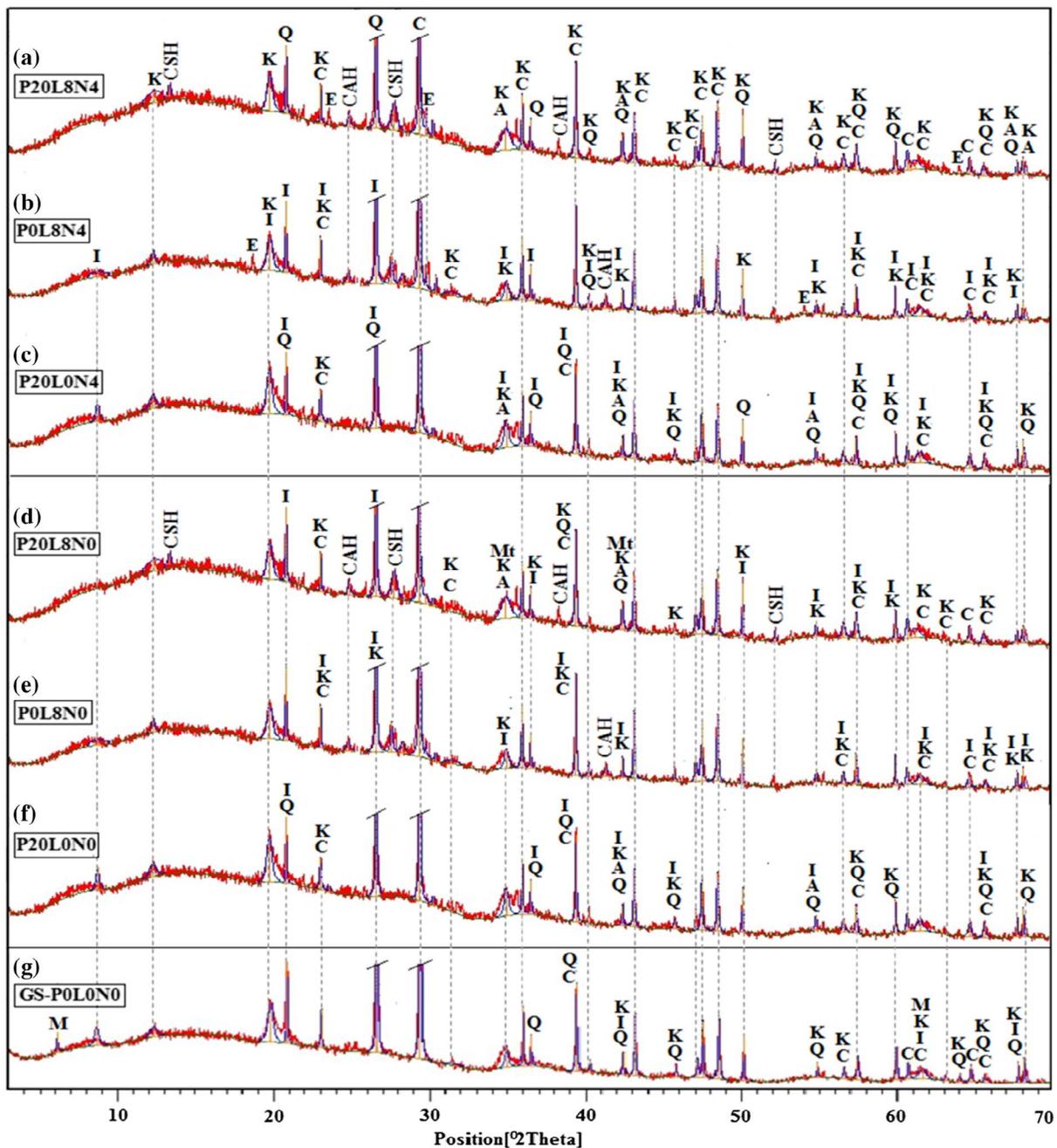
However, it is quite clear to observe that the combination of L and NP has a much better effect on the UCS of the RS than the GS. This effect becomes very important at later stage. For example, after curing for 120 days the UCS of both GS and RS samples stabilised with the combination of 20%NP and 8%L represents, respectively, an increase of 47 and 16 times to compare with untreated soil samples (Fig. 6(a)). Similar observations were reported by Hossain *et al.* (2007) where they found that the combination of 10% volcanic ash and 4%L for both S1 and S2 soils represented an increase of 21 and 10 times, respectively, to compare with untreated soil samples.

The dissolution of alumina and silica from soil and/or NP depends strongly on the L content which produces more cementitious products (C–S–H and C–A–H) responsible to the increase in the UCS of both GS and RS samples. In all cases, high UCS values are observed for samples stabilised with the combination of L and NP compared to those stabilised with L or NP alone. The same behaviour is observed by Koliass *et al.* (2005). Generally, the better increase produced by the addition

of L alone or in combination with NP on the UCS of both GS and RS samples can be explained by the pozzolanic reactions which form new cementitious products and consequently bind the soil particles together (Harichane *et al.* 2011c). Indeed, the XRD diagrams show that the high increase in UCS values of both GS and RS samples treated with L or L–NP in the absence of sulphates is due to the formation of new cementing agents such as C–S–H and C–A–H (Figs. 7(d, e) and 8(d, e)).

Variation of the UCS in the presence of Na₂SO₄

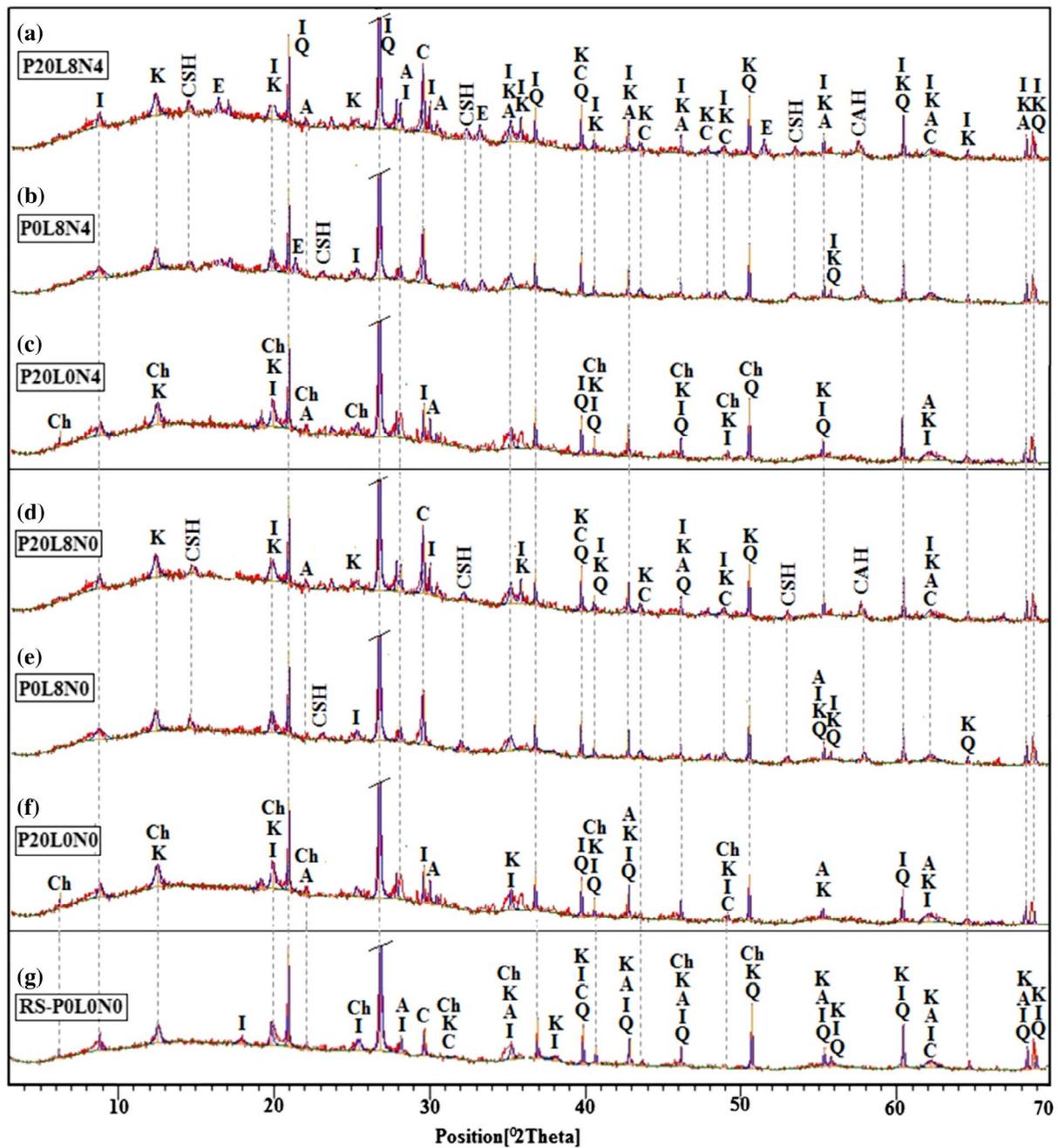
Figures 4(b–d), 5(b–d) and 6(b–d) present the results of the effect of Na₂SO₄ on the UCS of both GS and RS samples stabilised with L, NP and their combination. It is obvious to observe that for 7 days of curing the GS and RS samples stabilised with L in the presence of different contents of Na₂SO₄ present high UCS values to compare with samples without Na₂SO₄ (Figs. 4(b–d) and 5(b–d)). However, for a longer curing period, the presence of Na₂SO₄ with high content affects considerably the UCS of both GS and RS samples stabilised with the same additive (Figs. 4(b, d) and 5(b, d)). This alteration is very pronounced for the RS samples than the GS samples. The early increase in strength can be attributed to the presence of sodium hydroxide which accelerates the pozzolanic reaction rate mainly during the short time (Shi and Day 2000a). In addition, the presence of sodium hydroxide increases the pH and causes the dissolution of a large amount of alumina and silica that come into reaction with the remaining L to form cementitious products (Sridhran *et al.* 1995). This chemical reaction explains the early increase in UCS values of both GS and RS samples. However, the UCS of both stabilised GS and RS samples decreases significantly



7 X-ray diffraction analyses of untreated and treated GS samples without or with 4% Na_2SO_4 after curing for 60 days: E – Ettringite, Mt – Magnetite, A – Albite, Q – Quartz, K – Kaolinite, I – Illite, M – Montmorillonite and C – Calcite

with increasing Na_2SO_4 content (Figs. 4(d) and 5(d)). However, for both GS and RS samples treated with L in the presence of 2% of Na_2SO_4 , the UCS increases with increasing L content and curing period. But after 120 days of curing, the UCS of the GS stabilised with L or NP decreases gradually with increasing Na_2SO_4 content. The decrease in strength can be explained by the reduction in the capacity of cementing due to the adsorption of sulphate ions on the surfaces of C–S–H (Mehta 1983). Moreover, after 120 days of curing period, it can be seen in Fig. 5(c, d) that the RS samples stabilised with NP or L alone are entirely deteriorated when the Na_2SO_4 content is equal or

greater than 4%. On the one hand, the deterioration of the RS samples treated with the addition of NP is certainly not linked with the eventual formation of ettringite which is not observed in XRD diagrams (Fig. 8(c)). In contrast, the deterioration of the same soil samples (RS) is certainly linked with the formation of ettringite (Fig. 8(b)). This is due to the pressure value developed by the formation of ettringite which is very higher than that of the tensile strength value of the soil (Le Borgne 2010). Moreover, the deterioration of the GS samples is marginal to compare with the RS samples. This is leading us to suggest that this is due to the behaviour of the RS with the



8 X-ray diffraction analyses of untreated and treated RS samples without or with 4% Na₂SO₄ after curing for 60 days, E – Ettringite, A – Albite, Ch – Chlorite, Q – Quartz, K – Kaolinite, I – Illite and C – Calcite

Na₂SO₄ interaction. According to these results, it is possible to classify the Na₂SO₄ as deleterious for soil stabilisation when its content is greater than 2% by dry weight of soil. Generally, for any content of Na₂SO₄, the differences in the UCS between L and NP are more pronounced with the GS than with the RS.

For any content of Na₂SO₄ and for a shorter curing period (7–30 days), the increases in UCS values of both GS and RS samples are considerable when the L and NP are combined. However, for a shorter curing period, the UCS of both GS and RS samples decreases gradually with an increase in Na₂SO₄

content (Fig. 6(b–d)). The decrease in UCS values is more pronounced in the RS than in the GS. The same behaviour was observed by Sivapullaiah *et al.* (2006). Generally, for any content of Na₂SO₄, the RS samples are not altered when the combination L–NP is used as an additive. In all cases at short curing period, there is a great increase in UCS values of both GS and RS samples with increasing Na₂SO₄ content to compare with that of the treated samples without Na₂SO₄. This is due to enhanced lime reactions by increased availability of silica due to the increase in pH of soil solution (Davidson *et al.* 1960).

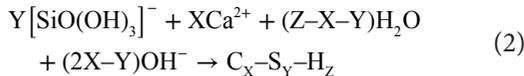
Stabilisation mechanism and strength degradation

Cause of lime–natural pozzolana soil strength

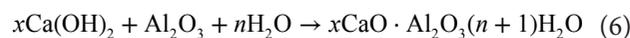
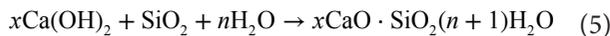
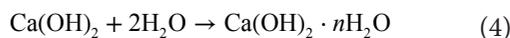
It is known that without Na₂SO₄, when the L is mixed with water (2H₂O), the hydroxide lime [Ca(OH)₂] hydrolyses first and increases the pH value of the solution very quickly as follows (Equation (1)):



According to Shi and Day (2000b), when the pH of the soil solution is greater than 12.5, the dissolution of amorphous SiO₂ increases very steeply. Generally, the pH will have a similar effect on the dissolution of NP as that on the amorphous SiO₂ because the main composition of NP is silica. However, the acceleration in the NP dissolution leads to the acceleration in the rate of pozzolanic reactions. Consequently, depolymerised monosilicates and aluminates species enter in the solution and form two main compounds [SiO(OH)₃]⁻ and [Al(OH)₄]⁻. However, C-S-H and C₄AH₁₃ compounds can be formed when calcium ions (Ca²⁺) contact these dissolved species (monosilicates and aluminates) as follows (Equations (2) and (3)):



On the other hand, Hu *et al.* (2016) reported that there are four equations of physical and chemical reactions (ion exchange, crystallisation, pozzolanic reaction and carbonation) occurred in lime–fly ash soil under the standard curing condition, shown as (Equations (4)–(7)).



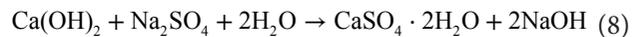
The chemical compounds C_x-S_y-H_z, C₄AH₁₃, Ca(OH)₂·nH₂O, xCaO·SiO₂(n+1)H₂O and xCaO·Al₂O₃(n+1)H₂O are the results of the chemical reactions (Equations (2)–(6)). The role of these compounds in lime–fly ash and lime–natural pozzolana soil systems is:

- to produce a layer of stable protective film that envelops the soil particles;
- to cement the soil particles together;
- to seal the voids that decreases the void index and consequently the water permeability;
- to improve the compactness and consequently the UCS of soil.

However, the CaCO₃ crystal (Equation (7)) has the capacity to cement the soil particles together, and improves the compressive strength of lime–fly ash and lime–natural pozzolana soil systems.

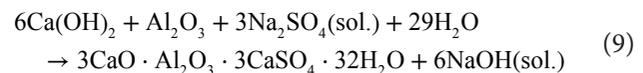
Effects of Na₂SO₄ on the strength of lime–natural pozzolana soil

When Na₂SO₄ is added, the reaction between Ca(OH)₂ and Na₂SO₄ in the presence of 2H₂O has been donated by Roy (1986) as follows (Equation (8)):

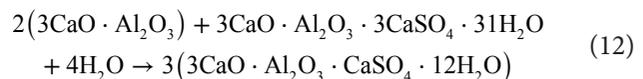
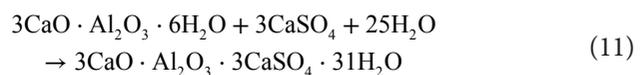
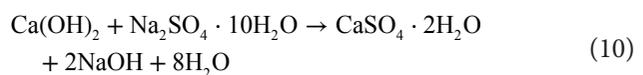


In addition, Sridhran *et al.* 1995 reported that the reaction between L, NP and Na₂SO₄ in the presence of 2H₂O produces a sodium hydroxide (NaOH) which develops a higher alkaline solution to compare with that of the Ca(OH)₂. The high pH developed by NaOH leads to the dissolution of a large amount of alumina (Al₂O₃) and silica (SiO₂) which react with the remaining L to form a high amount of cementitious products responsible on the significant increase in compressive strength values.

However, the presence of Na₂SO₄ increases the concentration of SO₄²⁻ ions and then leads to the formation of ettringite (3CaO·Al₂O₃·3CaSO₄·32H₂O), shown as (Equation (9)):



Furthermore, the chemical reactions between Na₂SO₄ and lime–fly ash soil has been donated by Hu *et al.* (2016) as follows (Equations (10)–(12)):



The new insoluble saline minerals 3CaO·Al₂O₃·3CaSO₄·32H₂O, 3CaO·Al₂O₃·3CaSO₄·31H₂O and 3(3CaO·Al₂O₃·CaSO₄·12H₂O) are the results of the chemical reactions between SO₄²⁻ ions and C_x-S_y-H_z, C₄AH₁₃, Ca(OH)₂·nH₂O, xCaO·SiO₂(n+1)H₂O and xCaO·Al₂O₃(n+1)H₂O (Equations (9), (11) and (12)). The effect of these insoluble saline minerals on lime–fly ash and lime–natural pozzolana soil systems is:

- to absorb a large quantity of water molecules which produce a high swelling;
- to fill the voids which decrease the void volume;
- to improve the compressive strength in the initial period;
- to dense the structure, reduce the compressive strength and deteriorate the specimens in the later period.

In the case of our study, it should be noted that the high early strengths can be explained by the acceleration of early pozzolanic reactions and the formation of ettringite due to the presence of Na₂SO₄.

Conclusions

The effect of the presence of Na₂SO₄ on Atterberg limits and UCS of both GS and RS stabilised with L, NP and the

combination of both has been studied. Based on the test results, the following conclusions can be drawn:

- In the absence of Na₂SO₄, the addition of L to both GS and RS samples produces a significant reduction in PI values but, when using the NP as an additive, a slight decrease is observed. However, when the L and NP are combined, a further decrease in PI values is reported.
- The presence of Na₂SO₄ increases the PI values of both GS and RS samples. Indeed, increases and decreases depend largely on the type of additive used and its content, Na₂SO₄ content and curing period. In addition, the mineralogical composition of soil plays an important role in the chemical reaction with Na₂SO₄.
- However, in the presence of 2% Na₂SO₄ and for any curing periods the UCS of both GS and RS samples stabilised with L alone or in combination with NP is higher than that of the samples without Na₂SO₄. The high early values of UCS can be attributed to the acceleration of early pozzolanic reactions caused by sodium hydroxide from Na₂SO₄.
- The deterioration of the RS samples after 120 days of curing can be explained by the formation of ettringite (observed in XRD diagrams) due to the presence of a high content of Na₂SO₄. In addition, the alteration of strength depends on the type of additive and its content, Na₂SO₄ content, curing period and the mineralogical composition of soil which plays an important role in the soil stabilisation process.
- It is indispensable to classify the Na₂SO₄ as a deleterious for soil stabilisation when its content is greater than 2% by dry weight of soil. However, at a short curing period, the Na₂SO₄ (less than 2%) can be used as an accelerator of NP dissolution and pozzolanic reactions without altering the stabilisation process.

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