Experimental Studies on Ettringite-Induced Heaving in Soils

Anand J. Puppala, P.E.[1]; Napat Intharasombat[2]; and Rajan K. Vempati[3]

Abstract: Sulfate-induced heaving in soils is primarily attributed to ettringite formation from the reactions between calcium of a lime or cement stabilizer, reactive alumina in soils, and sulfates in soils. Ettringite formation and subsequent heaving in soils are complex topics that are not well understood. This research is an attempt to advance the state of the knowledge on these topics. Ettringite was successfully synthesized in the laboratory by simulating conditions close to those in chemically treated sulfate-bearing soils. Soils spiked and compacted with the synthesized ettringite did not undergo heaving in one-dimensional free swell tests. However, heaving was observed when ettringite was formed inside the lime-treated soil specimen by including ionic reactions. Mineralogical studies including x-ray diffraction and scanning electron microscope studies were used to evaluate the presence of ettringite. Experimental investigations showed that the ettringite or sulfate-induced heaving was higher in clays than in sands under similar chemistry and environmental conditions. This is attributed to the void sizes of soil types and crystalline ettringite formation in the voids. Also, the effects of soil type, lime and sulfate amounts on this type of heaving, and hypothesized threshold levels of chemical ions to form ettringite mineral in treated soils, are addressed.

Introduction and Background

Natural expansive soils have been found in several countries across the world, and in the majority of the states in the United States (Chen 1988). Expansive soils undergo large volume changes when subjected to moisture changes. Both swell and shrinkage volume changes depend on several factors including type and amount of clay minerals, moisture content, dry density, soil structure, confining pressure, and climate. These volume changes may eventually cause severe damage to structures built above them (Chen 1988; Nelson and Miller 1992).

Soil stabilization is the modification of soils to enhance original soil properties to meet specific engineering requirements. Several stabilization methods are widely used in the field conditions to control soil heaving (Nelson and Miller 1992; Puppala et al. 2003). Methods include use of calcium-based stabilizers, noncalcium-based stabilizers, and geosynthetic reinforcements. Among these methods, calcium-based stabilizers, such as lime

[1]Associate Professor, Dept. of Civil and Environmental Engineering, Box 19308, Univ. of Texas at Arlington, Arlington, TX 76019-0308 (corresponding author). E-mail: anand@uta.edu

[2]Doctoral Research Assistant, Dept. of Civil and Environmental Engineering, Box 19308, Univ. of Texas at Arlington, Arlington, TX 76019-0308.

[3]Adjunct Professor, Dept. of Chemistry, Southern Methodist Univ., ChK Group Inc., 2045 Belgium Drive, Plano, TX. E-mail: chkgroup@worldnet.att.net

and cement, are widely used for expansive subsoil treatments due to their effectiveness in improving expansive soil properties and controlling volume changes (Chen 1988; Hausmann 1990).

Calcium-based stabilizers, including lime and cement, have been used to increase strength and to decrease plasticity index and swell and shrinkage strain potentials of expansive soils (Hausmann 1990). Several studies have shown that calcium-based stabilizer treatments of natural expansive soils rich with sulfates may lead to a new heave distress problem instead of mitigating it (Mitchell 1986; Hunter 1988; Mitchell and Dermatas 1992; Petry 1994; Kota et al. 1996; Puppala et al. 1999; Rollings et al. 1999). This phenomenon is referred to as sulfate-induced heave in the literature (Mitchell 1986; Dermatas 1995).

Sulfate-induced heave is primarily attributed to the presence of sulfates in natural expansive soils and usually occurs when lime or cement treatments are used for stabilizing these soils. Sulfates in soils react with lime or cement stabilizers in a basic environment (pH between 11 and 13) and this reaction will lead to the formation of an expansive sulfate mineral, known as ettringite, in a hydrated system. This mineral will continue to form as long as there are sufficient amounts of reactants present in the soil. This mineral will contribute to the sulfate-induced heave in lime- or cement-treated soils either by hydration or by continuous growth of itself or by both. Since this heave is caused by the addition of calcium-based stabilizers, it is regarded as a manmade or post-treatment expansive soil problem. Since the heave is primarily due to the formation of ettringite, it is also termed as "ettringite-induced heave" in the literature (Mitchell and Dermatas 1992). Others referred to it as lime-induced heave (Hunter 1988; Perrin 1992).

In the past, practitioners attributed sulfate heave distress to swell movements of natural underlying soils. However, studies conducted by Hunter (1988), Perrin (1992), Mitchell and Dermatas (1992), Petry (1994), Kota et al. (1996), Puppala et al. (1999), and other researchers attributed this heave to treated subgrades.

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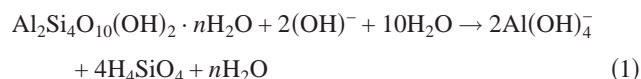
Table 1. Case Studies on Sulfate Heave Problems

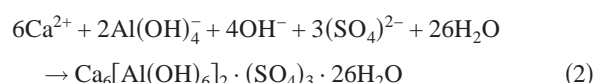| Reference (Year)	Location	Soil type	Lime (L)/cement (C) percent level	Sulfate content (mg/kg)	Heave appearance after construction
Mitchell (1986)	Las Vegas, Nev.	Silty clay	4% (L)	Up to 15,000	2 years
Hunter (1988)	Stewart Avenue, Las Vegas, Nev.	Silty clay	4.5% (L)	43,500	6 months
Perrin (1992)	Lloyd Park, Joe Pool Lake, Dallas, Tex.	OC Clays	5% (L)	2,000–9,000	Immediately
Perrin (1992)	Auxiliary Runway, Laughlin AFB, Spofford, Tex.	Clays	6–9% (L)	14,000–25,000	2 months
Perrin (1992)	Cedar Hill State Park, Joe Pool Lake, Dallas, Tex.	Highly plastic residual clays	6% (L)	21,200	2 months
McCallister and Tidwell (1994)	Denver International Airport, Denver, Colo.	Expansive clays	NA (L)	2,775	NA
Kota et al. (1996)	SH-118, Alpine and SH-161, Dallas, Tex.	Clayey subgrades	4% (C) 6–7% (L)	>12,000	6 to 18 months
Burkart el al. (1999)	Localities in Dallas–Fort Worth Region, Tex.	Clays	6%–9% (L)	233–18,000	Varies
Puppala (1999)	Dallas–Fort Worth International Airport, Irving, Tex.	Clay	5% (L)	320–13,000	3 months
Gaspard, personal communication, 2000	Near Shreveport, La.	Aggregates	NA	NA	NA
Rollingsee and Rollings (2003)	Holloman Air Force Base, N.M.	Crushed concrete	NA	NA	Several years

Note: NA=Not available.

Several case studies are reported in the literature and a list of them is presented in Table 1.

From case studies covered in Table 1, it is interesting to note that the sulfate content as a percent of dry weight of soil needed to induce heaving varied from 320 mg/kg (or ppm) to as high as 43,500 mg/kg. The time of sulfate heave appearance after chemical stabilization ranged from a few days to 18 months. Also, soils that experienced this heaving ranged from sands to silts to clays. These large variations in sulfate content and time to heave occurrence could be attributed to the fact that soil physical, chemical and mineralogical properties, as well as environmental conditions including temperature and humidity conditions in the case studies were different from site to site. Additionally, it is likely that some other parameters may be critical for soil heaving. However, one common conclusion is that the sulfate-induced heave was caused by the formation and hydration of ettringite mineral in lime- and cement-treated sulfate-rich natural soils.

Hunter (1988) presented a chemical relationship model of time-treated montmorillonite sulfate-rich clays to explain the formation of ettringite. These reactions could be applicable for other clayey soils. In this model, at pH>10, hydroxide ions (OH^-) released from the lime hydration reaction combine with aluminum disassociated from montmorillonite $[Al_2Si_4O_{10}(OH)_2]$ to form $Al(OH)_4^-$ (Lindsay 1979). The $Al(OH)_4^-$ in turn reacts with sulfates to form ettringite. The chemical reactions between these minerals are shown in the following (Hunter 1988):

$$Al_2Si_4O_{10}(OH)_2 \cdot nH_2O + 2(OH)^- + 10H_2O \rightarrow 2Al(OH)_4^-$$
$$+ 4H_4SiO_4 + nH_2O \tag{1}$$

(Dissolution of clay mineral, at pH>10.5)

$$6Ca^{2+} + 2Al(OH)_4^- + 4OH^- + 3(SO_4)^{2-} + 26H_2O$$
$$\rightarrow Ca_6[Al(OH)_6]_2 \cdot (SO_4)_3 \cdot 26H_2O \tag{2}$$

(Formation of ettringite)

The chemical reaction model indicates that dissolution of any clay minerals (alumina and amorphous silica) will occur due to the high pH conditions caused by the addition of lime stabilizer. Soluble sulfates $(SO_4)^{2-}$ present in soils react with anionic alumina species released from the dissolution of the clay mineral and calcium ions from stabilizer to form crystalline ettringite mineral. Thus, SO_4^{2-}, anionic Al, and Ca ions are referred to as candidate ions for ettringite formation in a hydrated system. Availability of water is important for this formation as indicated in Eq. (2).

Ettringite can form under different soil compositional and environmental conditions including the availability of high moisture content in soil and at moderate to high temperature conditions (25 to 40°C). Under favorable conditions, sufficient amounts of calcium, $Al(OH)_4^-$ species, and sulfate ions will be either released or present in soil to react with each other to form ettringite. However, the critical and physical conditions essential for the formation of ettringite is unclear. Table 2 presents a summary of previous research studies that addressed sulfate heave in soils under

Table 2. Previous "Experimental" Research Studies to Simulate Sulfate-Heaving Problems

Research	Soil type	Sulfates	Stabilizer
Mitchell and Dermatas (1992)	30% of kaolinite and montmorillonite mixed with 70% sand	Sodium sulfate and gypsum	Hydrated lime $Ca(OH)_2$
Petry (1994)	Natural clays	Gypsum	Lime
McCallister and Tidwell (1994)	Low plasticity clay	Gypsum	Lime
Wild et al. (1998)	kaolinite and Kimmeridge clay	Gypsum	Hydrated lime and slags
Viyanant (2000)	kaolinite,	Gypsum	Quick lime CaO
Chomtid (2000)	Illite and natural soils	Gypsum	Quick lime CaO

laboratory conditions. Some of these studies yielded ambiguous results, due to variations in soil and stabilizer conditions as well as the presence of chemical species in soils. Nevertheless, these studies provided valuable insights into soil and lime stabilizer aspects, which are considered in the present research.

Future research recommendations mentioned by previous studies are the need to understand fundamental chemistry of ettringite formation and heaving mechanisms in soils, and to develop quantitative relationships for predicting heaving in soils. The fundamental chemistry and the ettringite formation is the primary focus of the present research and results from this research evaluation are presented in this paper.

Research Objectives

The primary objective of this research was to understand ettringite formation and heaving mechanisms in lime-stabilized soils. Ettringite formation and heaving time periods in field conditions vary from days to months due to differences in soil composition and environmental temperature conditions. Hence, in order to accomplish this objective, an attempt was made to form ettringite mineral in lime-treated soils under laboratory conditions within a few minutes to hours, and then study the heaving potentials of the ettringite mineral in a compacted soil sample. These findings were used to identify the potential ettringite-induced heaving mechanisms such as crystal growth and ettringite hydration in lime-treated soils.

Other objectives of the research were to address the effects of soil type, lime and sulfate levels, reactive alumina, and compaction conditions on the sulfate-induced heaving mechanisms. Experimental results obtained from the evaluations of these objectives are covered in this paper.

Test Program Outline

Swell strain tests were conducted on two types of compacted kaolinite clay specimens; one compacted with synthesized ettringite as shown in Fig. 1(a) and the other by inducing ettringite formation in a lime-treated compacted soil specimen in the presence of a solution containing candidate ions as shown in Fig. 1(b). The latter approach was successful in the quick evaluation of sulfate heaving mechanisms and was adopted for the remainder of the testing.
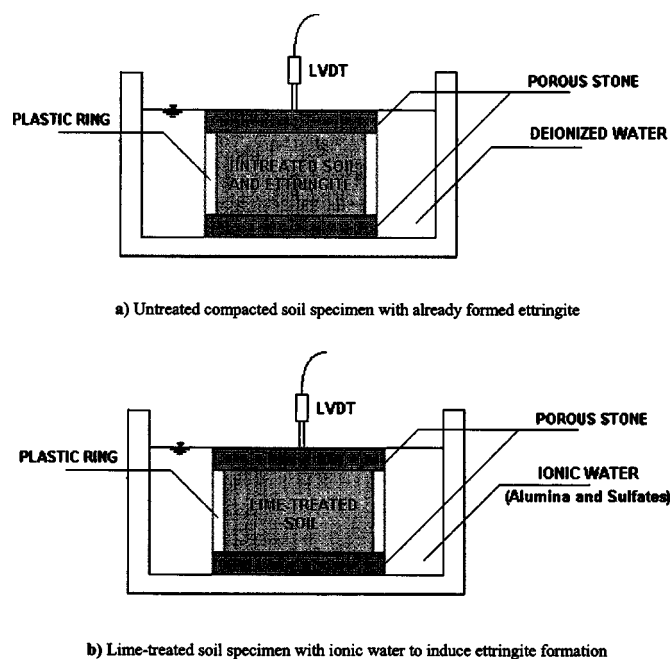

a) Untreated compacted soil specimen with already formed ettringite

b) Lime-treated soil specimen with ionic water to induce ettringite formation

Fig. 1. Types of soil specimens subjected to swell strain tests: (a) untreated compacted soil specimen with already formed ettringite and (b) lime-treated soil specimen with ionic water to induce ettringite formation

Testing Procedures

Both engineering and mineralogical tests were conducted on soil specimens to identify ettringite formation in soils and to measure the associated volume changes. Free vertical swell tests were conducted on treated soils to monitor soil volume changes due to lime, sulfate, and soil reactions. X-ray diffraction (XRD), scanning electron microscopy (SEM), and energy dispersive analysis of x-ray (EDAX) studies were conducted on soil specimens to identify the presence of ettringite mineral. The following sections describe soil specimen preparation and test procedures followed in this research.

One-Dimensional Free Swell Test

The one-dimensional free swell test measures the amount of soil heave in the vertical direction of a confined specimen. The amounts of materials required to obtain 0, 1,000, 2,500, 5,000, and 10,000 ppm of sulfates, and 0, 4, and 8% of lime in the dry soil samples were first calculated. Both amounts were based on the dry weight of the soil samples. The dry soil was then manually mixed with lime to prepare lime-stabilized soil specimens. The amount of water required for the compaction of each soil specimen was calculated and added to the soil mixture. The soil mixture was then compacted to a rigid plastic mold (6-cm in diameter and 2.4-cm in height). Immediately after compaction, porous stones were placed on both sides of the soil specimen, and the soil specimen was transferred to a free swell test apparatus. No seating load except for the weight of the porous stone was placed on the soil specimen, which was soaked with an aqueous solution of 4.46 mmol of sodium sulfate (Na_2SO_4) and 4.46 mmol of sodium aluminum oxide $(Al_2O_3Na_2O)$. This solution provided both soluble sulfates and reactive alumina for rapid formation of ettringite in the soils. The sources of aluminum in

Table 3. Previous Studies on Ettringite Synthesis Steps in Laboratory Conditions

Research	Chemicals used in reactions
Wang (2002)	Quick lime (CaO), gypsum ($CaSO_4 . 2H_2O$), and aluminum sulfate $[Al_2(SO_4)_3]$
Struble and Brown (1984)	Aluminum sulfate $[Al_2(SO_4)_3]$ and calcium oxide (CaO)
Odler and Abdul-Maula (1984)	Aluminum sulfate $[Al_2(SO_4)_3]$ and calcium hydroxide $[Ca(OH)_2]$
Jonathan et al. (1999)	Calcium hydroxide $[Ca(OH)_2]$, aluminum hydroxide $[Al(OH)_3]$, and sulfates

natural soils are amorphous hydroxyl aluminum, aluminosilicate phases of soil and from hydroxyl phases from smectite and montmorillonite layers, and organic complexes. All of these aluminum sources are collectively referred to as reactive alumina in this paper.

The amount of soil heave was measured using a micrometer dial gauge or linear voltage displacement transformer against actual time elapsed. The swell displacement readings of the soil specimen were continued until there were no significant changes in displacements for three days. In cases of small displacement changes, tests were continued for 21 days until the swell rates had reached plateau conditions. The final swell displacements along with the original heights of soil specimens were used to calculate swell strains in the vertical direction.

Mineralogical Studies

Two mineralogical studies were conducted in this research to evaluate the presence of ettringite mineral in soil samples using XRD and SEM studies with capabilities to conduct EDAX studies. XRD can provide qualitative and semiquantitatively identified ettringite; SEM provides ettringite morphology, and EDAX provides qualitative elemental data in the scanned sample. The following sections present test procedures.

Soil specimens were first mixed with water and ground into a smooth paste. The paste was deposited on a glass slide and dried on a hot plate (35°C). The soil was then subjected to Cu $K\alpha$ radiation with the scan speed of 0.07 degrees per minute. The data were recorded and analyzed to determine the presence of heaving mineral, ettringite.

SEM studies were conducted to understand the morphology of the ettringite. The samples were gold coated and then scanned, and several digital images at different magnifications were recorded. The ettringite minerals typically appear in needle shapes at higher magnifications. EDAX was used to analyze chemical compositions of the specimen. In this technique, electrons are bombarded in the area of desired elemental composition; the elements present will emit characteristics x rays, which are then recorded on a detector.

Ettringite Syntheses

Ettringite was synthesized in the laboratory by mixing aqueous solutions containing the candidate ions (calcium, sulfates, and aluminum), followed by the filtering the resulting precipitates. After initial tests confirmed that ettringite could be synthesized in this manner, similar procedures were used to form ettringite within lime-treated compacted soil samples by exposing the samples to the ionic mixtures. Details of the steps followed to synthesize ettringite using aqueous mixtures in the laboratory are described in this section.

Ettringite synthesis under laboratory conditions is well documented but the fundamental mechanisms of ettringite formation in soil matrices are poorly understood. Several syntheses methods have been attempted in laboratory settings by mixing appropriate molar solutions of different chemicals (Odler and Abdul-Maula 1984; Struble and Brown 1984; Vempati et al. 1996; Jonathan et al. 1999). A summary of these methods is presented in Table 3. All of these methods were successful since the ettringite formation was verified by mineralogical studies including XRD, SEM, and EDAX studies. Studies presented in Table 3 were carefully reviewed and based on the information reported in these studies, quick lime was selected for use in this study.

Preliminary Ettringite Synthesis

This preliminary synthesis was attempted at 25°C using distilled water. In one container, 4.46 mmol of sodium sulfate (Na_2SO_4) and 4.46 mmol of sodium alumina oxide ($Al_2O_3Na_2O$) were mixed and dissolved in 120 cc of distilled water. In a separate container, 8.92 mmol quick lime (CaO) was dissolved in 80 cc of distilled water by constant stirring for several seconds. Next, mixtures in both containers were combined and stirred for a 10-min time period. This mixture was then filtered through a membrane filter of pore size of 0.1 μm and the precipitate was dried.

Both XRD and SEM studies conducted on the synthesized material are presented in Figs. 2(a and b). The XRD analyses data of the synthesized mineral showed five dominant ettringite peaks, which were close to standard d spacings (9.67, 5.60, 3.88, 2.76, and 2.56 Å) (JCPDS 1998). Other peaks noted in this figure match those of $CaCO_3$. The SEM micrograph shown in Fig. 2(b) indicates that the presence of needlelike structures in the synthesized mineral, which reconfirms that the ettringite was successfully synthesized under laboratory conditions in a short period of time. The $CaCO_3$, which are of cuboid/blocky morphology, are not detected in the present SEM studies, possibly due to trace amounts of this mineral or amorphous/poor crystallinity of the mineral formed resulting in a different shape and smaller particle size, which may have obscured its detection.

Final Ettringite Synthesis

There are two possible ettringite formation mechanisms in chemically treated soils as described in the literature (Dermatas 1995): (1) Formation through solution reactions and (2) formation through topochemical assisted reactions. In solution reactions, the chemical compounds quick lime, alumina, and sulfates are first dissolved into a basic solution at pH > 10, leading to the formation of ettringite (Hunter 1988). The second mechanism hypothesizes that the ettringite can be formed directly on the surface of calcium aluminate grains by reactions between sulfate ions (SO_4^{2-}) in the solution and calcium aluminate in the solid phase (Shizong 1995).
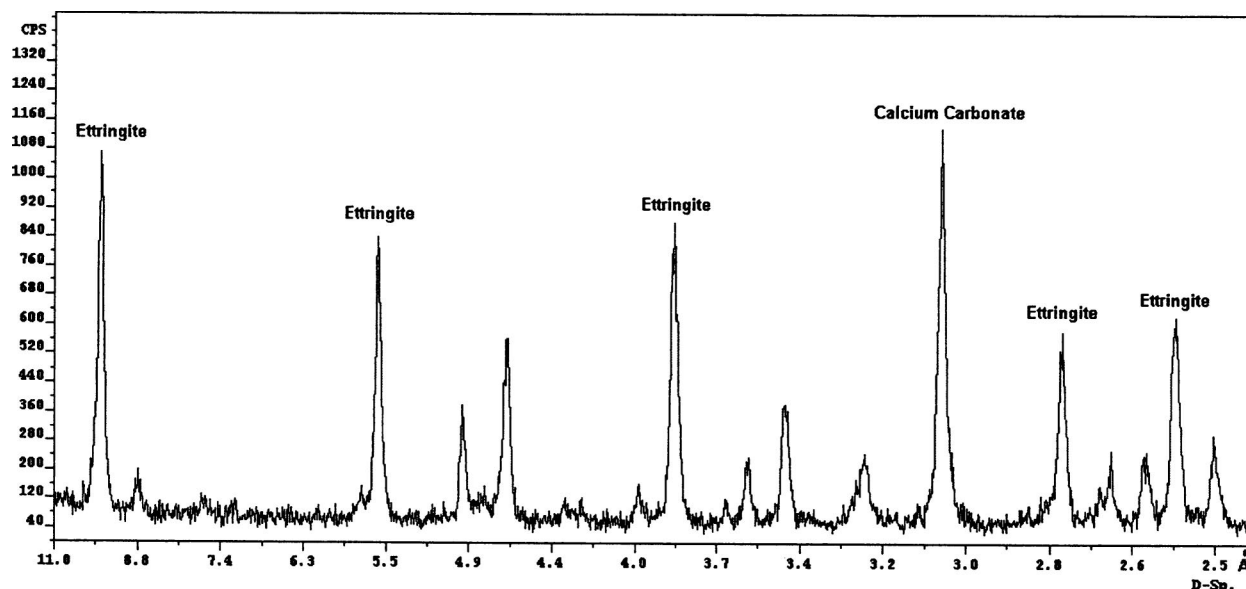
a) XRD pattern
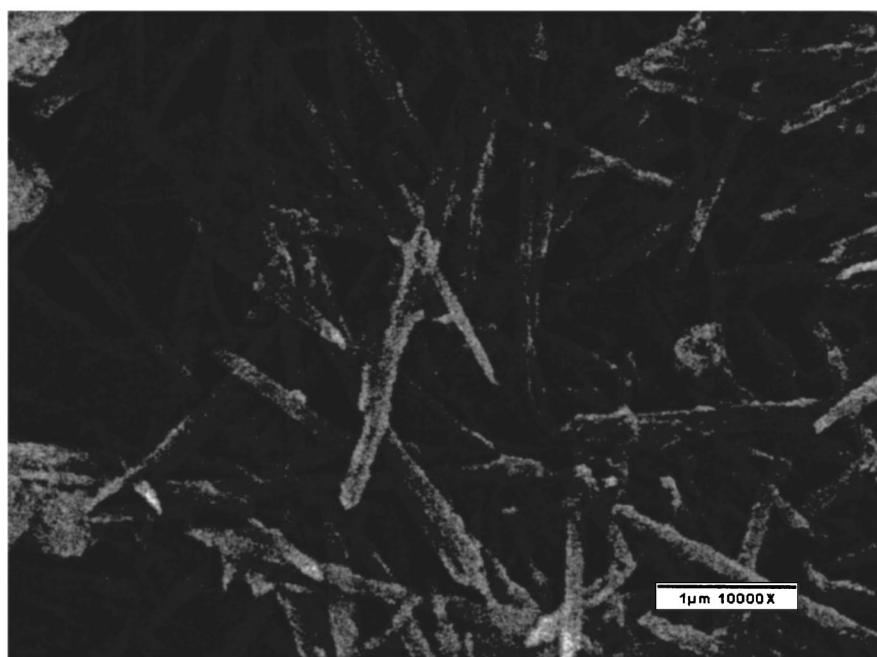
b) SEM image

Fig. 2. Mineralogical studies on precipitate formed in the ettringite synthesis: (a) x-ray diffraction pattern and (b) scanning electron microscope image

Ettringite Formation from Filtered Solution and Filtered Lime Residue

Ettringite formation was attempted by stirring quick lime in distilled water and then filtering the mixture to separate undissolved or excess quick lime (CaO). Stirring and temperature conditions were similar to those noted in the preliminary synthesis studies section. A membrane filter (0.1 μm) was used, which would retain undissolved lime particles in the dimension of 0.2 μm. Then, both filtered solution and the filtered lime residue on the filter paper were mixed with sodium sulfate (Na_2SO_4) and sodium aluminate oxide ($Al_2O_3Na_2O$) solutions and were stirred for 10 min. Both mixtures were filtered to get the precipitate. The XRD analyses were conducted on both precipitates and these results were presented in Figs. 3(a and b).

From both figures, it can be mentioned that ettringite mineral is formed regardless of whether dissolved lime or filtered lime residue is used [Figs. 3(a and b)], but with varying crystallinity. To summarize these results, the filtered lime residue predominantly formed calcium carbonate ($CaCO_3$) with minor amount of poorly crystallized ettringite, whereas, the filtered CaO extract produced crystalline ettringite with $CaCO_3$. This suggests that excess of undissolved CaO reacts with atmospheric CO_2 at ambient conditions to form $CaCO_3$, which then acts as seeding or templating material favoring more calcite formation rather than ettringite formation. In synthesis, seeding is commonly used to speed the formation of desirable mineral, and improve mineral crystallinity (Vempati, U.S. Patent, 2002). Overall, it can be summarized that ettringite can be synthesized and formed through
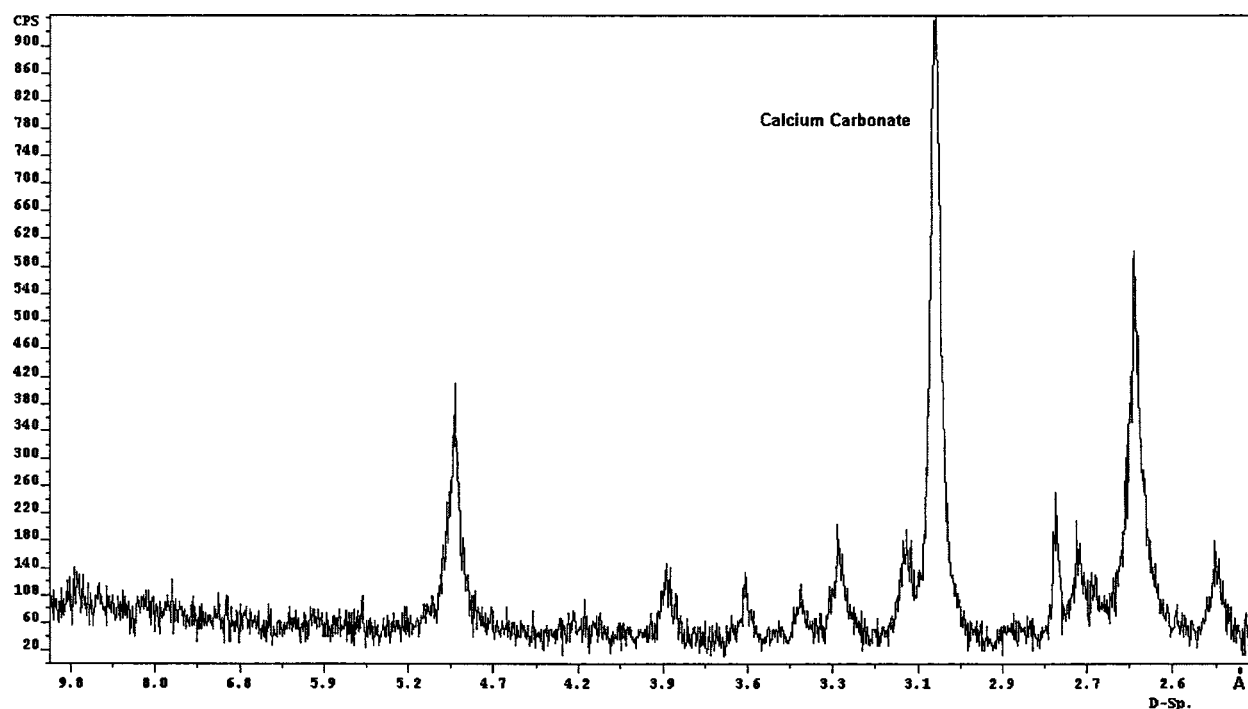
a) Precipitate resulting from solid lime residue
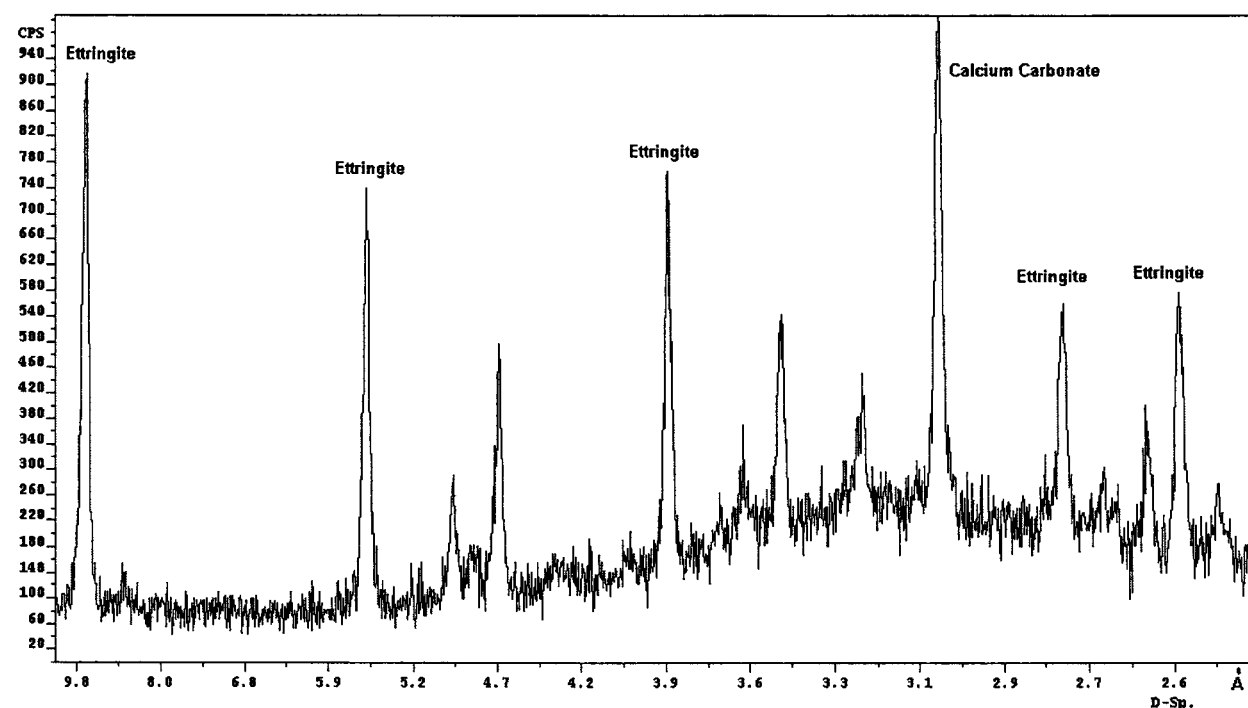
b) Precipitate from lime solution

Fig. 3. X-ray diffraction patterns: (a) precipitate resulting from solid lime residue and (b) precipitate from filtered lime solution

solution reactions simulating treated soil conditions. This suggests that when the soil pores are supersaturated with respect to ettringite bearing candidate ions, then its formation is likely to occur.

Heave Studies on Compacted Kaolinite

The kaolinite clay used in this research study is not reference clay and SEM images indicated the presence of frayed kaolinite edges

due to weathering. Basic properties of the kaolinite used in these experiments, including optimum moisture content and dry unit weights are presented in Table 4. The swelling behavior of ettringite was studied on three identical kaolinite specimens compacted with 8% of synthesized ettringite (from final synthesis) and three other specimens without ettringite. The 8% of ettringite material added to the soil specimens was based on dry weight of soil. Standard Proctor compaction method was followed in the preparation of soil specimens. All soil specimens were compacted

Table 4. Physical Properties of Soils Used in the Experimental Studies

Property	Kaolinite	Mixed soil (30% kaolinite and 70% sand)	Sand
Passing No. 200 (%)	100	30	0.70
Liquid limit (LL, %)	46.7	16	16
Plastic index (PI, %)	13.0	2	Nonplastic
Optimum moisture Content (%) at control, 4% lime and 8% lime treatments	24.1 (control) 25.8 (4% lime) 28.2 (8% lime)	17.5 13.5 13.2	15.4 11.9 10.0
Optimum dry unit weight (kN/m^3) at control, 4% lime and 8% lime treatments	15.0 (control) 14.6 (4% lime) 13.7 (8% lime)	16.8 17.7 18.4	15.6 16.6 16.9
Specific gravity, G_s	2.65	2.78	2.71
Void ratio, e	0.73 (control) 0.78 (4% lime) 0.90 (8% lime)	0.62 0.54 0.48	0.70 0.60 0.57
AASHTO Classification	A-7-5	A-2-4	A-3
USCS	Lean clay	Silty sand	Poorly graded sand
Classification	(CL)	(SM)	(SP)

to optimum moisture content and maximum dry unit weight conditions, and were then subjected to one-dimensional free swell tests. Fig. 4 presents both average and individual test results of soil specimens with and without admixed ettringite. As seen in the figure, the free swell test results on identical soil specimens were close to each other with a low coefficient of variation (less than 7%). Hence, the average swell strain results are used hereafter in the figures depicting swell strain behaviors.

Ettringite is perceived to swell by the adsorption of water as suggested by several studies including Dermatas (1995). On the contrary, from Fig. 4, soil specimens compacted with synthesized
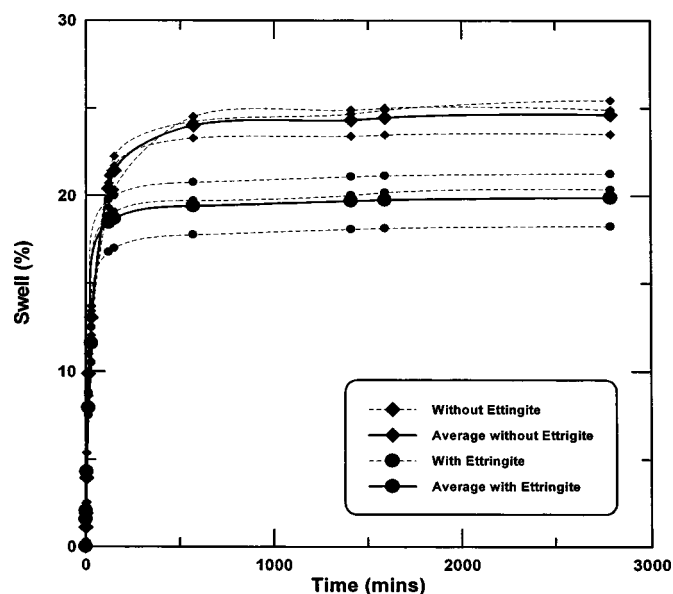
Fig. 4. Swell test results of kaolinite clay with 8% of synthesized ettringite

ettringite minerals inside experienced less swelling than soil specimens prepared without ettringite minerals. This phenomenon could be attributed to two reasons, which are described in the following paragraph.

The first possibility is that ettringite, being a fibrous or needle-shaped material of a high aspect ratio, is known to reinforce soil particles by inducing interlocking attraction. This attraction is known to strengthen materials including soils (Kujala et al. 1983; Schoute 1999). This strengthening phenomenon was observed in the present kaolinite clay specimens compacted with ettringite minerals. Compacted soil samples without ettringite disintegrated rapidly after swelling whereas those with ettringite were stiff and did not show any signs of disintegration after swelling. The adsorption of water by ettringite minerals did not cause any further swelling since the dispersive forces generated during the adsorption of water are less than the interlocking and strengthening forces between ettringite and soil particles. The second possible explanation, which requires experimental evaluation, is that water might not have migrated into interlayers of "compacted" ettringite minerals to induce moisture hydration-related swelling; hence, low swell strains were recorded.

Swell Tests on Kaolinite with Induced Ettringite

The next step was to prepare soil specimens and create the formation of the ettringite mineral in the soil by inducing chemical reactions inside the soil skeleton or pores. The amounts of sodium sulfate added to each soil specimen were determined based on the residual sulfate contents in the control soil, a procedure similar to the one used by Viyanant (2000) and Chomtid (2000).

Vertical swell tests were performed on kaolinite clay specimens using three different conditions: (1) Specimen prepared without lime (Condition 1 or simply control soil); (2) specimen prepared with lime additive at 8% in the presence of sodium sulfates in solid form (Condition 2); and (3) specimen with 8% lime in the presence of sodium sulfates and sodium aluminate in solution form (Condition 3). The sodium sulfate was first dissolved in distilled water to prepare sulfate-rich water, which was then used to mix and prepare soil samples to reach targeted compaction moisture contents. Additionally, dissolved sodium aluminum oxide was added to the sulfate-rich solution to provide reactive alumina for the quick formation of ettringite. It should be noted that other forms of reactive alumina from clay minerals are also available for chemical reactions. All these steps were performed prior to conducting swell tests.

Three swell tests were performed on triplicate identical specimens for each condition and the average results of each condition are presented in Fig. 5. It is interesting to note that the control kaolinite (Condition 1) exhibited an average swell strain of 24%. High swelling was attributed to optimum compaction moisture content and low seating pressure applied in the swell tests. The 8% lime-treated kaolinite (Condition 2) exhibited a swell strain of 19%, which indicates that the lime treatment was somewhat effective on this soil even in the presence of sodium sulfates. On the other hand, the lime-, aluminate-, and sulfate-treated kaolinite clay (Condition 3) experienced highest swelling of 29%, which is more than the control soil indicating the formation of ettringite mineral and subsequent heaving in this soil.

To confirm the ettringite formation, XRD studies were conducted on all of three above conditioned soil samples and these results are presented in Figs. 6(a–c). These figures indicate that no ettringite peaks were noticed in control soils (Condition 1) in Fig. 6(a). The ettringite peaks of very low intensities were observed
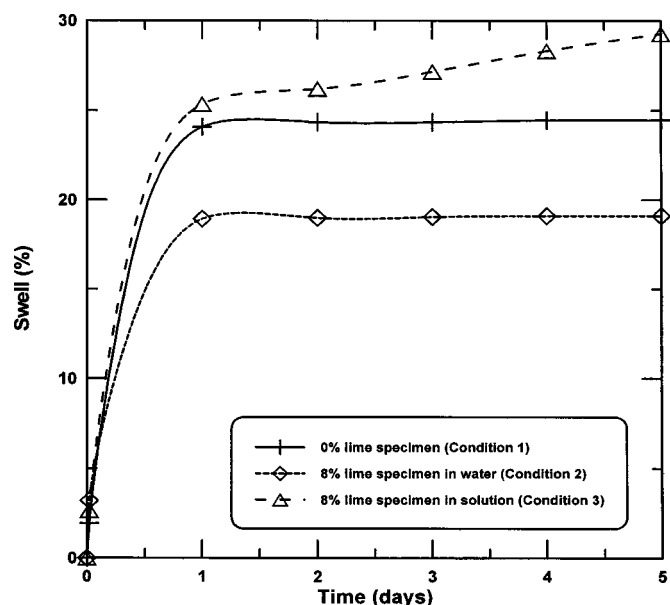
Fig. 5. Swell strain test results of kaolinite clay subjected to three different treatment conditions

for soils prepared with lime and sulfates (Condition 2) in Fig. 6(b). However, more pronounced ettringite peaks were observed for soils with lime, sulfates, and sodium aluminate in solution forms (Condition 3) as presented in Fig. 6(c). This substantiates that in situ formation of ettringite due to the presence of Al $(OH)_4^-$ and sulfate species in a lime-treated soil specimen and hydration of this mineral resulted in soil heaving as noted in Fig. 5. Therefore, for pronounced formation of ettringite in soil in addition to sulfate and calcium ions, a source of alumina is essential in addition to high pH and hydrated environment.

Elemental composition of soil specimen of Condition 3 was analyzed using EDAX. The EDAX test results on needle-shaped particles present in lime-treated kaolinite specimen are shown in Fig. 7. This elemental analysis showed the presence of gold (Au), calcium (Ca), sulfur (S), and aluminum (Al) in the soil specimen. The specimen was gold coated for SEM studies in order to prevent charging. The presence of other chemicals, Ca, S, and Al, in soils reconfirm that these are the main components for the formation of ettringite mineral. Based on XRD, SEM, and EDAX studies, it can be concluded that the ettringite was formed in kaolinite clay under Condition 3.

Other Experimental Studies

Soil Compositional and Environmental Variables

Previous research studies (Chomtid 2000; Viyanant 2000) concluded that the magnitude of swell is related to soil type, compaction dry unit weight, moisture content, and lime and sulfate levels in soils. An experimental program was hence designed in this research to determine the effects of a few important soil variables on ettringite-induced swelling by inducing quick formation of ettringite inside the soil specimens. The variables studied here are described in the following.

A total of three soil types were studied in this research. These were: (1) Kaolinite clay, (2) mixed soil (30% kaolinite clay and 70% sand), and (3) pure sand. Physical properties of all three soils

including optimum moisture contents are presented in Table 4. Among chemical treatments, quick lime treatment was researched here at three different dosage levels, 0, 4, and 8% by the dry weight of soil. The present tests were conducted at optimum moisture content, which corresponds to maximum dry unit weight condition. Five different sulfate levels were used for kaolinite clay, including 0, 1,000, 2,500, 5,000, and 10,000 mg of sulfates/kg of soil or mg/kg or ppm. In the case of mixed clay soil and pure sand, only high and low sulfate levels were considered. These sulfate levels were 0, 5,000 and 10,000 mg/kg for the mixed soil and 0 and 10,000 mg/kg for the sand.

After the soil samples were mixed with the selected lime and sulfates, they were compacted close to targeted optimum moisture content and dry unit weight conditions. These samples were then subjected to free swell and mineralogical tests at room temperature conditions.

Discussion of Test Results

Swell tests were conducted on all three different soil types to study the effects of soil type and soil properties on sulfate-induced heave. Swell test results of kaolinite clay and mixed and sandy soils were presented in Table 5 and were used in the following discussion and analysis.
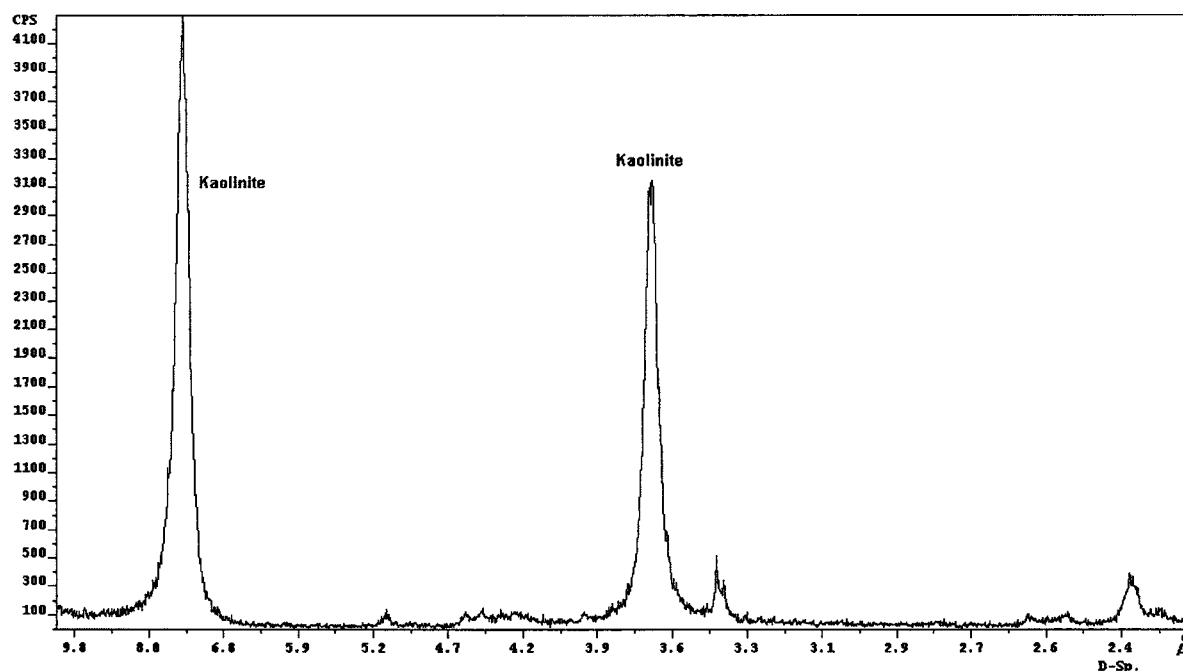
Under natural conditions, kaolinite clay exhibited more swelling than mixed and sandy soil specimens. The amount of soluble sulfates in untreated compacted soil specimens did not show any appreciable effects on the swelling behavior of the same specimens. Sandy soil test results showed low magnitudes of sulfate-induced swell strains, which were around 1%. Reasons for low swell strains are explained in the next section.

Results in Table 5 confirm that both mixed and clayey soils are more susceptible to sulfate-induced heaving than the sandy soil. However, the occurrence of sulfate heaving phenomenon in non-expansive sands should not be disregarded. The formation of ettringite is based on presence of the candidate ions including aluminum ions and optimum environmental conditions.

By fixing and isolating soil types and the amounts of lime stabilizer used, the effects of sulfates on heaving could be well understood. Fig. 8(a) presents swell test results of lime-treated kaolinite clay at different sulfate levels. From Fig. 8(a), it can be mentioned that the amount of soluble sulfates had minor influence on the swell magnitudes of untreated kaolinite clay. This is because of lack of formation of ettringite minerals in the soils since these soils are not treated with calcium based lime stabilizer. Fig. 8(b) of mixed soil shows similar trends.

In the case of lime-treated soil specimens, swell magnitudes initially decreased when compared to the swell strains of control soils at zero sulfate level and then increased as the dosages of sulfates increased. At low soluble sulfate contents ($<2,500$ mg/kg), the 4% lime-treated kaolinite clay and mixed soil specimens had more swelling than the 8% lime-treated soil samples. Additionally, the swell magnitudes of both 4 and 8% lime-treated soils are higher than those of control soils. This explains that the detrimental reactions were caused by the presence of sulfates and reactive alumina in lime-treated soils. Reactive alumina in treated soils was provided by alumina solution mixed with the soaked water and alumina disassociated from clay particles at high pH conditions (between 10 and 13) resulting from lime treatments. However, the rate of dissolution of alumina from clays will depend on clay mineralogy, crystallinity, and particle size.

At higher soluble sulfate levels ($>2,500$ mg/kg), the 8% lime-

a) Specimen submerged in water with no sulfates (Condition 1)
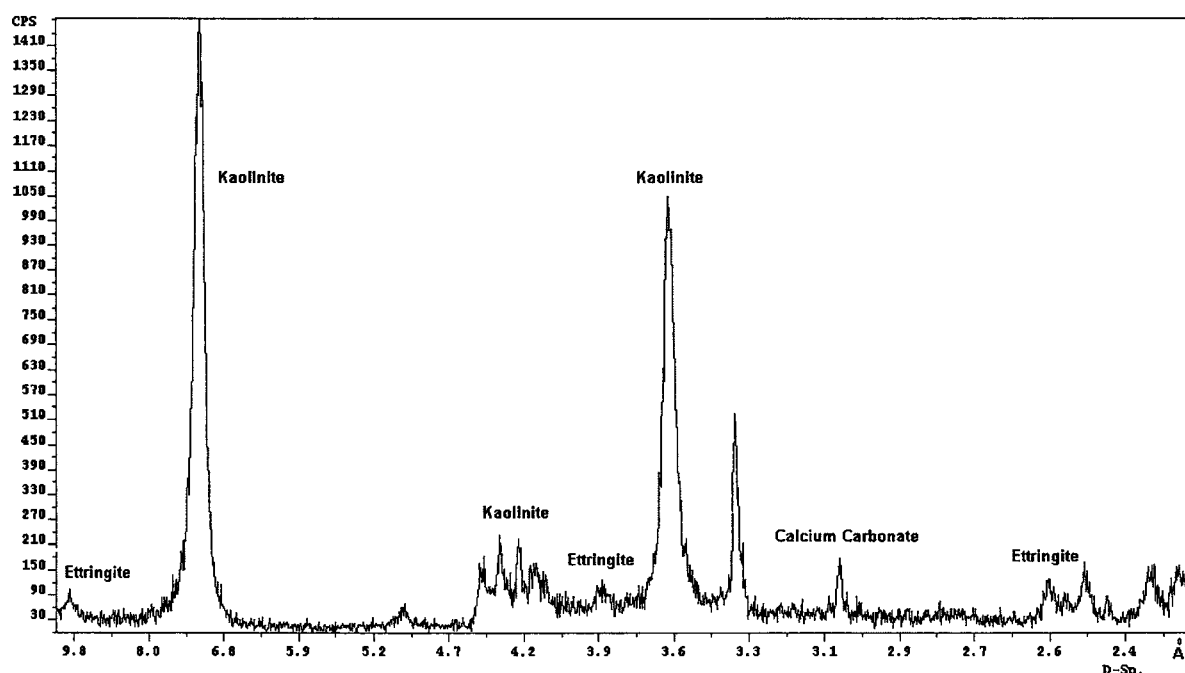
b) Sulfates in solid form and submerged under water (Condition 2)

Fig. 6. X-ray diffraction results on 8% lime-treated clay: (a) water (Condition 1), (b) sulfates in solid form (Condition 2) and (c) sulfates in solution form (Condition 3)

treated soil samples exhibited more swelling than the 4% lime-treated soil samples. This is because of the large amounts of calcium and reactive alumina ions $[Al(OH)_4^-$ predominant species] released at higher dosages of lime, which lead to increased amounts of ettringite formation and heaving in these soils. Overall, the trends observed in Fig. 8 can be summarized in the following observations:

1. At zero sulfate levels, lime stabilization mechanisms play a dominant role in the lime-treated soils, and, hence these soils

experience lower swelling than the control soil. Ettringite was not formed in these soils due to lack of sulfates in the soils. Lime is known as an effective stabilizer to reduce swell potentials of kaolinite clay since it suppresses diffused double water layers of clays and their plasticity and related swell potentials.

2. At sulfate levels ranging between 0 and 2,500 mg/kg, lime stabilization reactions and sulfate heave generation reactions occur at the same time due to the presence of lime, sulfates,
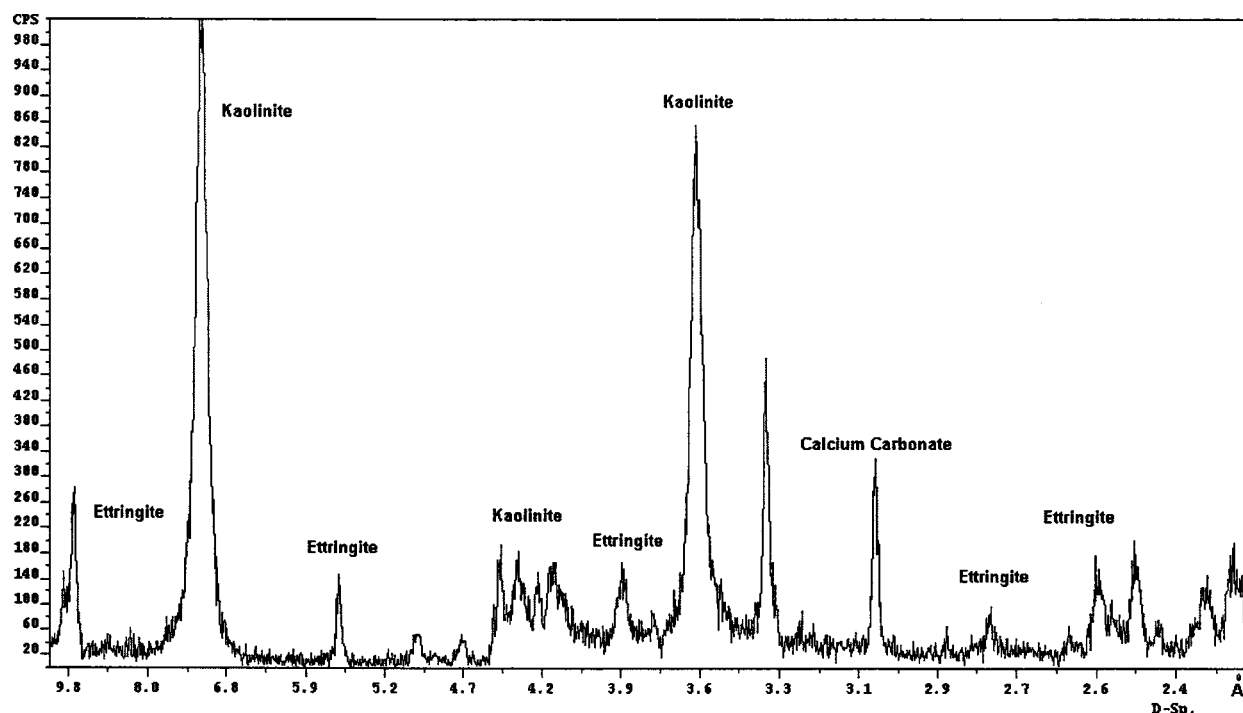
c) Sulfates in solution form (Condition 3)

Fig. 6. (*Continued*).

and high pH conditions. In the present soils at these low sulfate ranges, the sulfate heave reactions might have dominated the lime stabilization reactions. Hence, slightly higher swells were noted for 4% lime-treated soils. However, the swell magnitudes were smaller at higher lime dosage (i.e., 8%), explaining the significance of high amounts of lime stabilization reactions at low sulfate levels. Also, a high lime dosage induces the formation of $CaCO_3$ because of its reaction with atmospheric CO_2.

3. At sulfate levels ranging above 2,500 mg/kg, sulfate heave reactions dominated the lime stabilization reactions. High sulfate-induced swells were recorded at these sulfate levels.
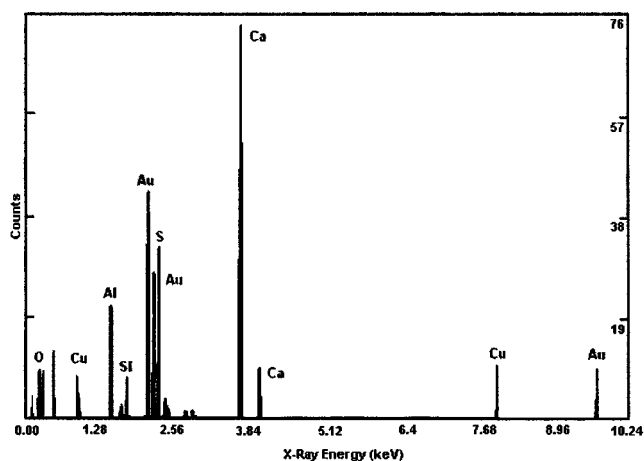

Fig. 7. Energy dispersive analysis x ray of 8% lime-treated specimen at 10,000 mg/kg of sulfates

The presence of lime and reactive alumina at these sulfate levels appeared to contribute more to ettringite formation than stabilization reactions and hence large swells were recorded at increased lime dosages. This observation, along with the previous observation, raises an important conclusion, i.e., critical or threshold amounts of calcium, reactive alumina and sulfates must be present in chemically treated soils to form ettringite. Any variations in these amounts might not result in the formation of ettringite as noted in the case of 8% lime-treated soils with low amounts of sulfates. The on-going research is currently in the process of establishing these threshold levels.

4. The swell strains in kaolinite clay are higher than those of mixed soil and this variation is attributed to amounts of reactive alumina present in the treated soils. Large quantities of reactive alumina in solution led to higher sulfate-induced swelling in the kaolinite soils. This ettringite formation resulted in high swell strains in the kaolinite specimens due to small size of voids in kaolinite. Therefore, the size of the voids is important, rather than the total volume of voids. Currently efforts are being made to develop a simple laboratory testing technique for quantification of reactive alumina. The writers believe this technique will lead to the development of a predictive model for ettringite formation in various soil compositional and environmental conditions.

Fig. 9 presents the same kaolinite and mixed soil test data by plotting swell strains versus lime dosage levels at different sulfate levels. Fig. 9(a) indicates that, at 0 mg/kg of soluble sulfates, lime was effective in reducing swell magnitudes of kaolinite clay. At soluble sulfate content less than 2,500 mg/kg, swell magnitudes at 4% lime treatment were the highest among the three dosages. At these moderate sulfate levels, the 8% lime treatment reduced swell strains of kaolinite. At high soluble sulfates

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Table 5. Swell Strain (%) Properties of Present Soils

Lime	Soil type	Soluble sulfate (mg/kg)				
		0	1,000	2,500	5,000	10,000
0% lime	Kaolinite clay	24.9	23.4	24.1	22.9	24.2
	Mixed soil	3.5	NT	NT	3.2	4.3
	Sand	0.00	NT	NT	NT	0.00
4% lime	Kaolinite clay	22.9	25.1	28.7	29.7	32.3
	Mixed soil	2.7	NT	NT	5.55	7.3
	Sand	0.0	NT	NT	NT	0.7
8% lime	Kaolinite clay	19.9	23.7	27.4	31.1	39.2
	Mixed soil	2.3	NT	NT	4.9	9.4
	Sand	0.0	NT	NT	NT	1.2

Note: NT=Not tested.

(>2,500 mg/kg), sulfate heaving was evident in all treated kaolinite clays and is directly proportional to the amount of lime treatment used to stabilize kaolinite. Fig. 9(b) shows that mixed soil results display similar trends with lower swell strain magnitudes. Overall, these results indicate that the threshold problematic sulfate levels for inducing heaving in subgrade soils depend on soil type, including clay mineralogy, lime dosages, reactive alumina, pH conditions, sulfate amounts as well as the amount and size of voids present in the compacted soils.

Causes of Ettringite-Induced Heaving

The writers attribute the ettringite crystal formation and growth as one of the main contributors to the overall ettringite-induced heaving in the present treated soils. Ettringite crystal growth in soils was first hypothesized by Dermatas (1995). The ettringite crystals are generally formed in the soil voids during initial reactions. This formation is expected to occur within an hour after soaking soil specimens in water. These crystals will then start to accumulate with the continued reactions between candidate ions at high pH conditions. When the existing pore void space of soil could not accommodate any more ettringite crystals, the soil will start expanding. Another reason for the crystal growth hypothesis here is due to the fact that even nonplastic-treated sandy soils displayed low heaving, which was attributed to the large size of voids and low amount of reactive alumina in sandy soils. The hypothesis needs further experimental verification by conducting ettringite formation studies in real-time scanning force microscopy or atomic force microscopy under controlled environmental conditions using environment cell device (Vempati and Cocke 1994). Additional swell movements could be attributed to the hydration reactions of ettringite minerals formed inside the soil matrix. However, the present investigations showed that very low magnitudes of heaving were recorded due to hydration of already formed ettringite mineral in compacted soils (see Fig. 4).

Future Research Needs

One of the important factors for ettringite formation is the presence of reactive alumina in soils. Very little is known about the presence of reactive alumina presence in soils, except for the disassociation of alumina from clay minerals. Lindsay (1979) noted that the amounts of this alumina disassociation from well crystallized clay minerals at pH>8 are insignificant. Hence, the writers opine that the alumina in the present treated soils may have been contributed from other sources including those in the

form of oxides, hydroxides, and amorphous aluminosilicates including poorly crystalline colloidal size clay minerals. Current research is exploring the presence and causes of reactive alumina in soils. Other important research aspects are focused on the evaluation studies on the pH conditions and organic matter, especially of humic acid in soils and their impact on the ettringite formation. Future research should also address the effects of compaction dry unit weight or different void ratios of the same soil on the ettringite formation and related heaving.

Summary and Conclusions

It should be mentioned here that the observations noted in this paper are based on the majority of the trends noted in the test results. These conclusions are valid for soils similar to the ones tested in this research. The following lists a few of the important conclusions arrived from this research:

1. Ettringite can be quickly synthesized in the laboratory by mixing quick lime (CaO), sodium sulfate, and sodium alumina oxide in water and stirring it for 10 min at room temperature conditions.

2. Mixing synthesized ettringite with compacted soils did not result in the simulation of ettringite induced heave in the same soils, possibly due to strengthening reactions of the ettringite to the soils and low moisture adsorption capacities of the compacted and hydrated ettringite minerals.

3. The formation of ettringite minerals inside the compacted soil voids was successful and this formation resulted in sulfate heaving when subjected to engineering vertical swell tests. Time frame to induce this ettringite formation was again relatively short (less than an hour) and the heaving of the mineral was recorded within two or three days.

4. In the presence of certain thresholds of chemical reactants and water, ettringite crystals continued to form and accumulate in the voids of soils in all directions. Ettringite-induced heaving in the present soils was attributed to crystal growth since the pore sizes in the test soils could not accommodate any more ettringite formation and hence exhibited swelling. This observation was based on the test results and heaving patterns noted in the present three test soils including sands and clays. The lower the void size in soil samples, the higher the sulfate heaving in soils. Hence, sandy soils exhibited low amounts of heaving in this research, possibly due to large sizes of the voids present in the sands. Also, limited amount
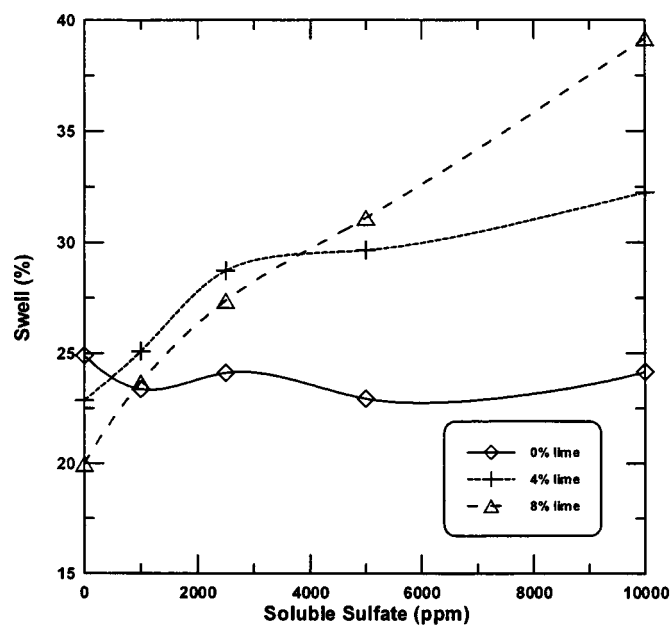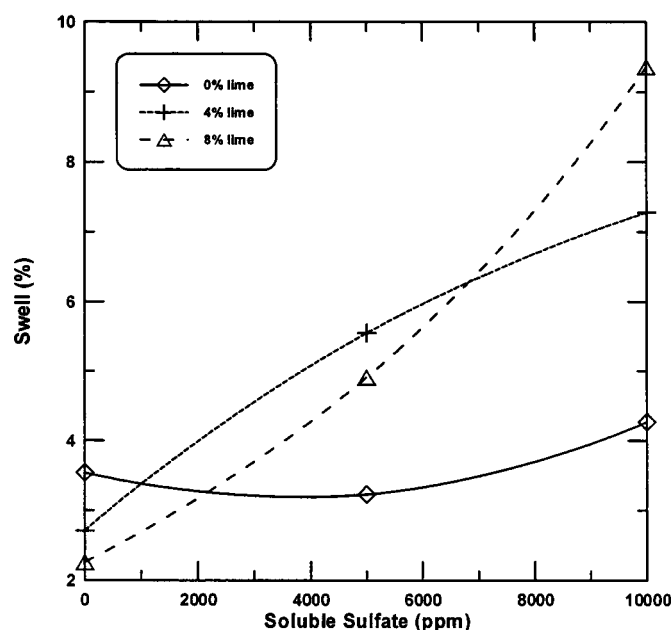
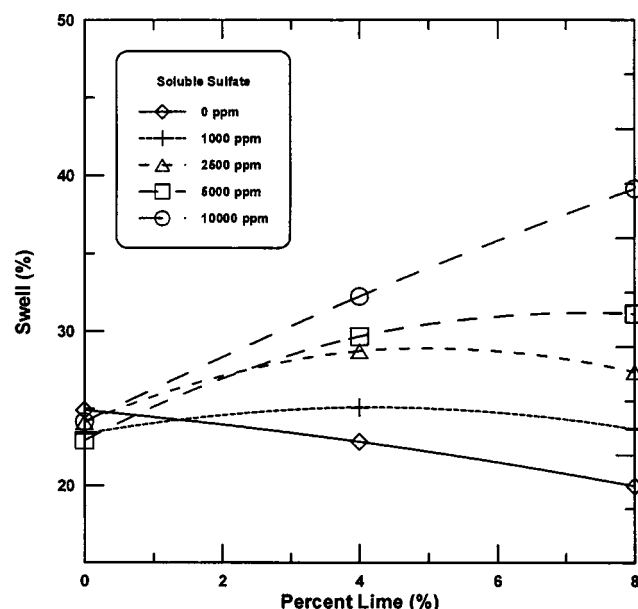**Fig. 8.** Effects of soluble sulfates on free vertical swell test results of (a) kaolinite clay and (b) mixed soil
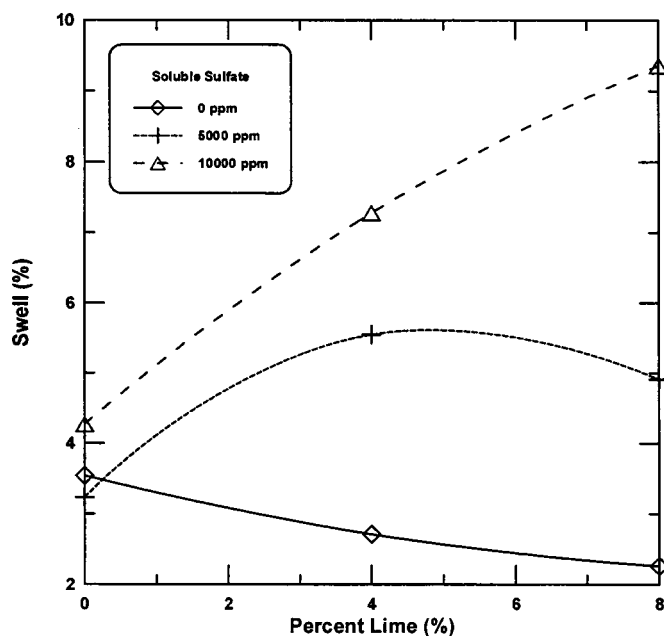

Fig. 9. Effects of lime treatment on free vertical swell test results of (a) kaolinite clay and (b) mixed soil

of clay or alumina fraction in sand did not facilitate continuous formation of ettringite mineral. Hydration of ettringite mineral did not cause any heaving in the kaolinite clay, suggesting low to moderate contribution of this heave mechanism to the overall ettringite induced heaving.

5. At low sulfate levels (<2,500 ppm), an increase in lime stabilization resulted in low magnitudes of sulfate-induced heaving, possibly due to lime treatment reactions dominating ettringite formation reactions. At high sulfate levels (>2,500 ppm), an increase in lime stabilization resulted in higher heaving due to increased amounts of ettringite formation at these sulfate conditions.

6. Mineralogical XRD and SEM studies are powerful tools to

identify ettringite presence in the treated soils. Based on the present mineralogical studies, it can be mentioned that SEM studies should be used in conjunction with XRD studies, in particular when swell strains are measured in treated soils, but ettringite XRD peaks are obscured. Otherwise, poor interpretations will be made regarding ettringite formations.

Acknowledgments

References

Burkart, B., Goss, C. G., and Kern, P. J. (1999). "The role of gypsum in production of sulfate-induced deformation of lime-stabilized soils." *Environ. Eng. Geosci.*, 5(2), 173–187.

Chen, F. H. (1988). *Foundations on expansive soils*, 2nd ed., Elsevier, New York.

Chomtid, S. (2000). "Laboratory evaluation of sulfate heaving mechanisms in artificial illite soil and natural soil." MS thesis, Univ. of Texas at Arlington, Arlington, Tex.

Dermatas, D. (1995). "Ettringite-induced swelling in soils: State-of-the-art." *Appl. Mech. Rev.*, 48(10), 659–673.

Hausmann, M. R. (1990). *Engineering principles of ground modification*, McGraw–Hill, New York.

Hunter, D. (1988). "Lime-induced heave in sulfate-bearing clay soils." *J. Geotech. Eng.*, 114(2), 150–167.

Jonathan, L. W., Griffin, P. V. C., Andrew, W., and Roger, D. (1999). "Design and synthesis of macrocyclic ligands for specific interaction with crystalline ettringite and demonstration of a viable mechanism for the setting of cement." *J. Chem. Soc., Perkin Trans. 2*, 10, 1973–1981.

Joint Committee on Powder Diffraction Standards (JCPDS). (1998). International Centre for Diffraction Data, Publication DBM-1-23, Philadelphia.

Kota, P., Hazlett, D., and Perrin, L. (1996). "Sulfate-bearing soils: Problems with calcium based stabilizers." *Transportation Research Record. 1546*, National Research Council, Washington, D.C. 62–69.

Kujala, K., and Nieminen, P. (1983). "On the reactions of clays stabilized with gypsum lime." *Proc., 8th European Conf. on Soil Mechanics and Foundation Engineering*, 2, 929–932.

Lindsay, W. L. (2001). *Chemical equilibria in soils*, Wiley, New York.

McCallister, L. D., and Tidwell, L. (1994). "Double treatment to minimize sulfate-lime-induced heave in expansive clays." *Final report*, United States Military Academy, West Point, N.Y.

Mitchell, J. K. (1986). "Practical problems from surprising soil behavior." *J. Geotech. Eng.*, 112(3), 255–289.

Mitchell, J. K., and Dermatas, D. (1992). "Clay soil heave caused by lime-sulfate reactions." *ASTM STP 1135*, Innovations and Uses for Lime, Philadelphia.

Nelson, J. D., and Miller, D. J. (1992). *Expansive soils: Problems and practice in foundation and pavement engineering*, Wiley, N.Y.

Odler, I., and Abdul, M. (1984). "Possibilities of quantitative determination of the Aft—(ettringite) and AFm—(monosulphate) phases in hydrated cement pastes." *Cem. Concr. Res.*, 14(1), 133–141.

Perrin, L. L. (1992). "Expansion of lime-treated clays containing sulfates." *Proc., 7th Int. Conf. on Expansive Soils*.

Petry, M. T. (1994). "Studies of factors causing and influencing localized heave of lime-treated clay soils (sulfate-induced heave)." *Final Report*, Univ. of Texas at Arlington, Arlington, Tex.

Puppala, A. J., Hanchanloet, S., Jadeja, M., and Burkart, B. (1999). "Evaluation of sulfate-induced heave by mineralogical and swell tests." *Proc., 9th Pan-American Conf. on Soil Mechanics and Geotechnical Engineering*.

Puppala, A. J., Wattanasanticharoen, E., Intharasombat, N., and Hoyos, L. R. (2003). "Studies to understand soil compositional and environmental variables on sulfate heave problems." *Proc., Soil Rock America, 12th Pan American Conf. on Soil Mechanics and Geotechnical Engineering*.

Rollings, M. P., and Rollings, R. S. (2003). "Sulfate attack on bound bases." *Proc., Transportation Research Board Annual Meeting*, (CD-ROM) Washington, D.C.

Rollings, R. S., Burkes, J. P., and Rollings, M. P. (1999). "Sulfate attack on cement-stabilized sand." *J. Geotech. Geoenviron. Eng.*, 125(5), 364–372.

Schoute, E. (1999). "Chemical stabilization of soft clays." MS thesis, Center of Engineering Geology, No. 188, Delft Univ.

Shizong, L., Yanrog, W., and Chen, L. (1995). "Investigation on the formation of ettringite in the presence of BaO." *Cem. Concr. Res.*, 25(7), 1417–1422.

Struble, L. J., and Brown, P. W. (1984). "An evaluation of ettringite and related compounds for use in solar energy storage." Progress Report, *NBSIR 84-2942*, National Bureau of Standards, Gaithersburg, Md.

Vempati, R. K., and Cocke, D. L. (1994). "Application of atomic force microscopy to soil minerals." *Proc., 15th Int. Soil Science Congress*.

Vempati, R. K., Chintala, A. K., Mollah, M. Y. A., and Cocke, D. L. (1996). "Stabilization and solidification of toxic wastes using coke and coal combustion by-products." *Waste Manage.*, 15, 433–440.

Viyanant, C. (2000). "Laboratory evaluation of sulfate heaving mechanisms in artificial kaolinite soil." M.S. thesis, Univ. of Texas at Arlington, Arlington, Tex.

Wang, L. (2002). "Cementitious stabilization of soils in the presence of sulfate." PhD thesis, Louisiana State Univ., La.

Wild, S., Kinuthia, J. M., Jones, G. I., and Higgins, D. D. (1998). "Suppression of swelling associated with ettringite formation in lime stabilized sulfate bearing soils by partial substitution of lime with ground granulated blast furnace slag." *Eng. Geol. (Amsterdam)*, 51, 257–277.

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