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RESEARCH ARTICLE

Swelling of Natural Soil Subjected to Acidic and Alkaline Contamination

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Abstract

This paper aims at establishing the influence of acidic and alkaline pore fluids on the swell behaviour of an expansive soil. A series of laboratory one dimensional free swell tests were performed to study the behaviour of soil in acidic and alkaline environment. Three different concentrations of sodium hydroxide and sulphuric acid solutions were used as pore fluids to understand the influence of variable concentrations on the swell behaviour of soil. Results showed that, the swelling of soil that interacted with sodium hydroxide solution initially increased at lower concentration and then decreased with increase in concentration. In contrast, the swelling initially decreased at lower concentration of sulphuric acid and then increased with increase in concentration of solution. The complexity in the swell behaviour of contaminated soil was assessed by thoroughly investigating the mineralogy and microstructure alterations by carrying out X-ray diffraction analysis, Scanning electron microscopy and Energy dispersive analysis of X-ray at the end of interaction.

Keywords

Free Swell, black cotton soil, sodium hydroxide, sulphuric acid, x-ray diffraction, scanning electron microscopy

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1 Introduction

Properties of natural soils get significantly altered due to interaction with pollutants either disposed of or accidentally released on the land. The spawning of industrial projects and poor waste disposal practices necessitate a better understanding of soil behaviour under changed and extreme environmental conditions. Though the soil-pollutant interaction affects all properties of soil, swelling behaviour of soil in particular can cause serious damage to foundations and super structures built on it. Mal'tsev [1] has provided useful insight into the effects of infiltration of aggressive solutions (acids and alkalis) of various concentrations into the beds of their production structures. The production structures become unserviceable as a result of the deformation of their foundation structures and superstructures due to marked changes in soil properties. A number of researchers in the past few decades have reported several case studies on the effect of pollutants (acids and alkalis) on heaving of clayey soils [2–8]. The researchers mentioned above have identified several important factors, such as chemical composition and concentration of the pore fluid, pH of the medium, type and degree of electrolyte dissociation, chemicomineralogical composition and exchange capacity etc. which may affect swelling in soils. Further, it was also established that changes in soil volume is not only due to moisture variation, but also due to active decomposition under highly acidic and alkaline conditions leading to new formations in pore space.

2 Review of the previous works

Unexpected losses and infiltration of chemical reagents (acids, alkalis) into the ground may lead to deformations of foundations [6]. These chemical reagents get released into soil from various industries. Acids, particularly sulphuric acid gets released from many industries such as copper leaching, inorganic pigment production, petroleum refining, paper production and industrial organic chemical production. In a similar way, hydroxides, particularly sodium hydroxide is released into the soil from various industries such as paint and dye industries, paper and pulp industries, cotton mills and aluminum industries. Sokolovich [9] reported that swelling of clayey soils occurs when they interact with acid and alkali solutions. Alkali interaction of soils results in their heaving, but is less effective than acid-induced heaving, and takes place in a more complex manner and at a lower rate. Mal'tsev [1] investigated the chemical heaving of various types of sandy and sandy-clayey soils using aggressive chemical solutions (acids, alkalis) frequently employed in industry and the results showed that (i) the chemical heaving of soils depends primarily on their chemicomineralogical composition and dispersivity (ii) heaving increases with increase in alumina content and the amount of clay particles in the soils (iii) various types of soils follow the given order, sand < clayey loams < clays, in terms of tendency to increase their chemical heaving (iv) the wetting solutions can be arranged in the given sequence, water < alkali < acids, by order of the strength of their effect on the various type of soils. Chunikhin [3] has provided valuable insight into the role of different concentrations of alkali on the geotechnical properties of soils. Kabanov [5] highlighted that relative swelling depends on the type of soil, concentration of the alkali solution and duration of the interaction. Further, the swell process of clayey soils interacting with alkali solutions may be divided into three periods. In the first period, the swell may be due to accumulation of osmotic and adsorbed moisture in the soils. In the second period, which is characterized by relative stabilization of soil, it may be due to chemical reaction between the clay fraction and the alkali. In the third period, the swell may be due to rate of formation of new compounds. Mulyukov [10] from his investigation concluded that even 0.1 N alkali solution can promote activation of swelling in soils and any accidental spillage of alkali, when it enters the underground sections of buildings, may lead to failure of the underground technogenic sphere.

However, laboratory studies to understand the long-term influence of strong pollutants (acids and alkalis) on the swell behavior of natural soils were seldom carried out [11–14]. In view of the importance of studying swelling processes in soil because of acid and alkali solutions and the development of control measures, investigation of swell phenomenon needs to be carried out. Thus, the current study has been performed under laboratory conditions to assess the complex free swell behavior and mineralogical transformations of soil under the influence of sodium hydroxide and sulphuric acid solutions.

3 Materials and methods

3.1 Black cotton soil

The natural black cotton soil sample was collected by open excavation, from a depth of one meter from natural ground level at National Institute of Technology, Warangal (Latitude: 17 98′N & Longitude: 79 53′E), India. Oven dried soil passing through no.40 (425µ) sieve was used for performing tests. The physical properties of the soil are shown in Table 1.

Table 1 Physical properties of black cotton soil

Property	Value
Specific gravity	2.65
Clay (%)	60
Liquid limit (%)	54
Plastic limit (%)	25
Plasticity index (%)	34
Free swell index (ml/g)	1.3
Optimum water content (%)	20.2
Max. dry unit weight (g/cc)	1.68

3.2 Aqueous solutions

In this study, distilled water along with solutions of sodium hydroxide (NaOH) and sulphuric acid (H_2SO_4) of variable concentrations were used as pore fluid. Sodium hydroxide solution of known concentrations (1N, 4N and 8N) was prepared by dissolving required molecular weights (40g, 160g & 320g respectively) of analytical grade sodium hydroxide pellets in distilled water to make 1 liter of solution. Similarly, commercially available sulphuric acid by required volume (27.25ml, 108.69 and 217.98 ml, respectively) was diluted with distilled water to make one liter of solution of desired concentrations (1N, 4N and 8N, respectively).

3.3 One-dimensional free swell test

The one-dimensional free swell test measures the amount of swelling in the vertical direction of a confined specimen [15]. Special oedometer cells were made, using materials which were entirely non-reactive to both acid and alkali. Figure 1 shows the test setup used in this work. Soil samples sieved through 425µ sieve were used for the test. Soil was initially dried in an oven at 110^0C. Oven-dried soil once cooled was spread over a plane glass sheet and mixed with water at optimum moisture content. The mixed specimens were placed in a sealed plastic bag and then kept in the desiccator for 24 hours so as to attain uniform moisture content. The soil specimens are then compacted to their maximum dry density in a rigid Teflon cell (6cm in diameter and 2cm in height) to a height of 1.4cm. Porous stones were placed at the top and bottom of the specimens, which facilitate the movement of fluid to the soil specimen. The soil specimens were inundated with 1N, 4N and 8N concentrations of H_2SO_4 and NaOH aqueous solutions as pore fluid and allowed to swell under free loading condition. For reference, soil specimens were also tested using distilled water as a pore fluid. The swell displacement readings were measured using dial gauges until no significant changes in displacements were observed. The final swell displacements along with the original heights of the soil specimen were used to calculate percentage swell in the vertical direction. Representative soil samples were collected at the completion of the long term free swell tests and were analyzed using X-ray diffraction analysis, Scanning electron microscopy and Energy dispersive analysis of X-ray tests to identify the chemico-mineralogical and morphological changes that may have occurred.

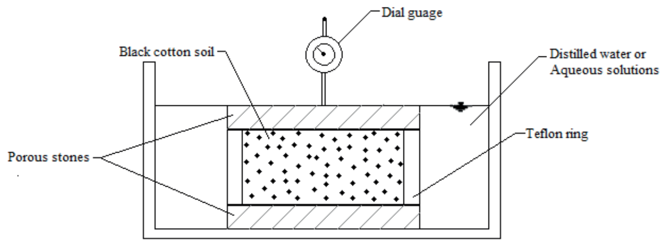

Fig. 1 Free swell test set up with black cotton soil in Teflon made ring

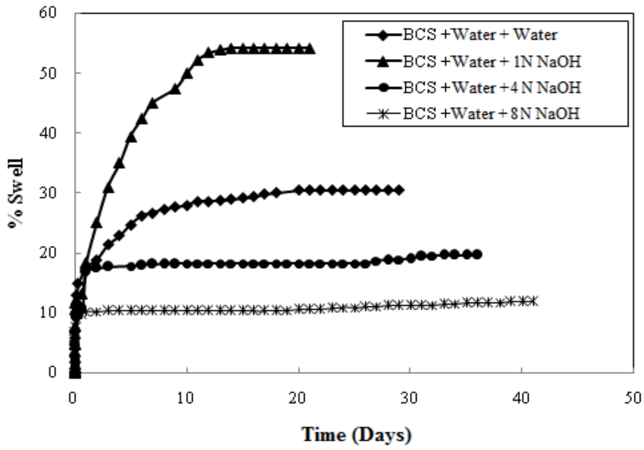

Fig. 2 Influence of pore fluids water and NaOH solutions on free swell behaviour of black cotton soil

3.4 Mineralogical studies

Mineralogical studies are conducted using PANanalytical X-pert powder diffractometer. Samples were scanned from 6º (2θ) and 70º (2θ) angle using copper K alpha radiation at a scanning rate of 2 degrees per minute. The data obtained is analyzed using X-pert high score plus software, to identify minerals by comparing the standard diffraction patterns of minerals.

3.5 Morphological studies

Morphological studies are carried out using scanning electron microscope (VEGA 3 series) with tungsten heated filament having live stereoscopic imaging using 3D beam technology. Scanning electron microscopy (SEM) with energy dispersive analysis of X-ray (EDAX) is conducted to understand the morphological and elemental composition changes occurring in the soil due to alkali and acid interaction. The soil specimens were mounted onto the tape glued to the flat surface of SEM stub and sputter coated with gold prior to scanning.

4 Experimental results

4.1 Swell test results with NaOH solution as pore fluid

Swell tests were performed on three specimens of the black cotton soil with different concentrations of NaOH solution as pore fluid (Fig.2). Results obtained from swell test are reported by plotting the percent swell strains against elapsed time intervals. From figure, variations in the swell curve can be recognized; the free swell of about 30% is observed upon inundation with distilled water, whereas it is about 54%, 19% and 12% respectively in the case of samples inundated with 1N, 4N and 8N NaOH solutions. Thus, it is noted that the specimen with lower concentration, i.e. 1N NaOH, underwent maximum amount of swell and the swell decreased with increase in concentration of alkali solution. In order to understand and explain the variations observed in free swell behavior of alkali contaminated soils, the specimens were examined for their x-ray diffraction pattern and morphological changes.

4.2 XRD results after swell test with NaOH solution

Figure 3 shows the variations in the X-ray diffraction patterns for different concentrations of NaOH solution in comparison with distilled water. X-ray diffraction pattern of soil with respect to water shows that the natural black cotton soil contains *Volkonskoite* (Peaks at 4.49, 2.56 and 1.50 [Å]) along with *Quartz* (Peaks at 3.34, 2.45, 1.81 and 1.67 [Å]) and *Microcline* (Peaks at 3.24, 2.90 and 1.98 [Å]) as their major minerals. The XRD pattern of soil inundated with 1N NaOH showed peaks pertaining to *Trona* (Peaks at 3.24, 2.90 and 1.98 [Å]),a new mineral, which is a Trisodium hydrogen dicarbonate dihydrate mineral. Soil with 4N NaOH showed *Tosudite* (Peaks at 4.48, 4.25 and 3.30 [Å]), interstratified clay mineral that consists of dioctahedral chlorite/ smectite. Further, an increase in concentration to 8N showed new mineral peaks pertaining to *Sodalite* (Peaks at 6.34, 4.48 and 3.66 [Å]), sodium aluminum silicate hydroxide, along with Trona.

4.3 SEM and EDAX results after swell test with NaOH solution

SEM images along with EDAX of the soil sample that interacted with water, 1N NaOH, 4N NaOH and 8N NaOH solutions are shown in Figure 4.The SEM image of BCS interacted with 1N NaOH solution (Fig. 4b) indicates that it has undergone some disintegration or weathering upon interaction with the alkaline solution compared to the image (Fig. 4a) obtained after interaction with water. More severe weathering is observed in case of other higher concentrations of alkali solutions. From the SEM image of BCS after interaction with 8N NaOH solution (Fig. 4d), it has been observed that particles have degraded or weathered completely. Particles with "*rosettetype*" (Sodalite mineral has the similar structure) morphology can be seen from the SEM image, which is an indication of morphological changes in soil due to soil-alkali reaction.

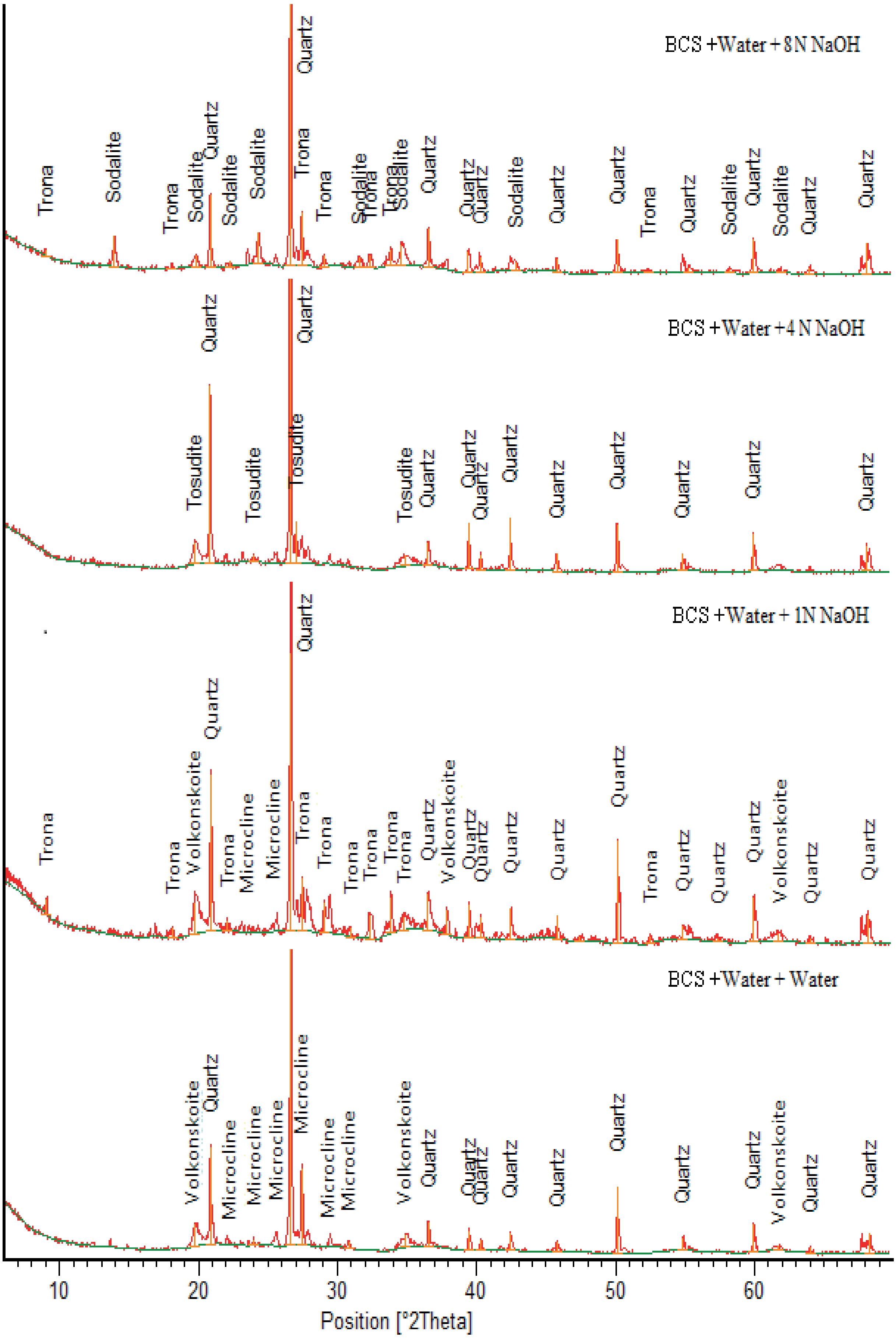

Fig. 3 X-ray diffraction patterns of black cotton soil after free swell with Water and NaOH solutions

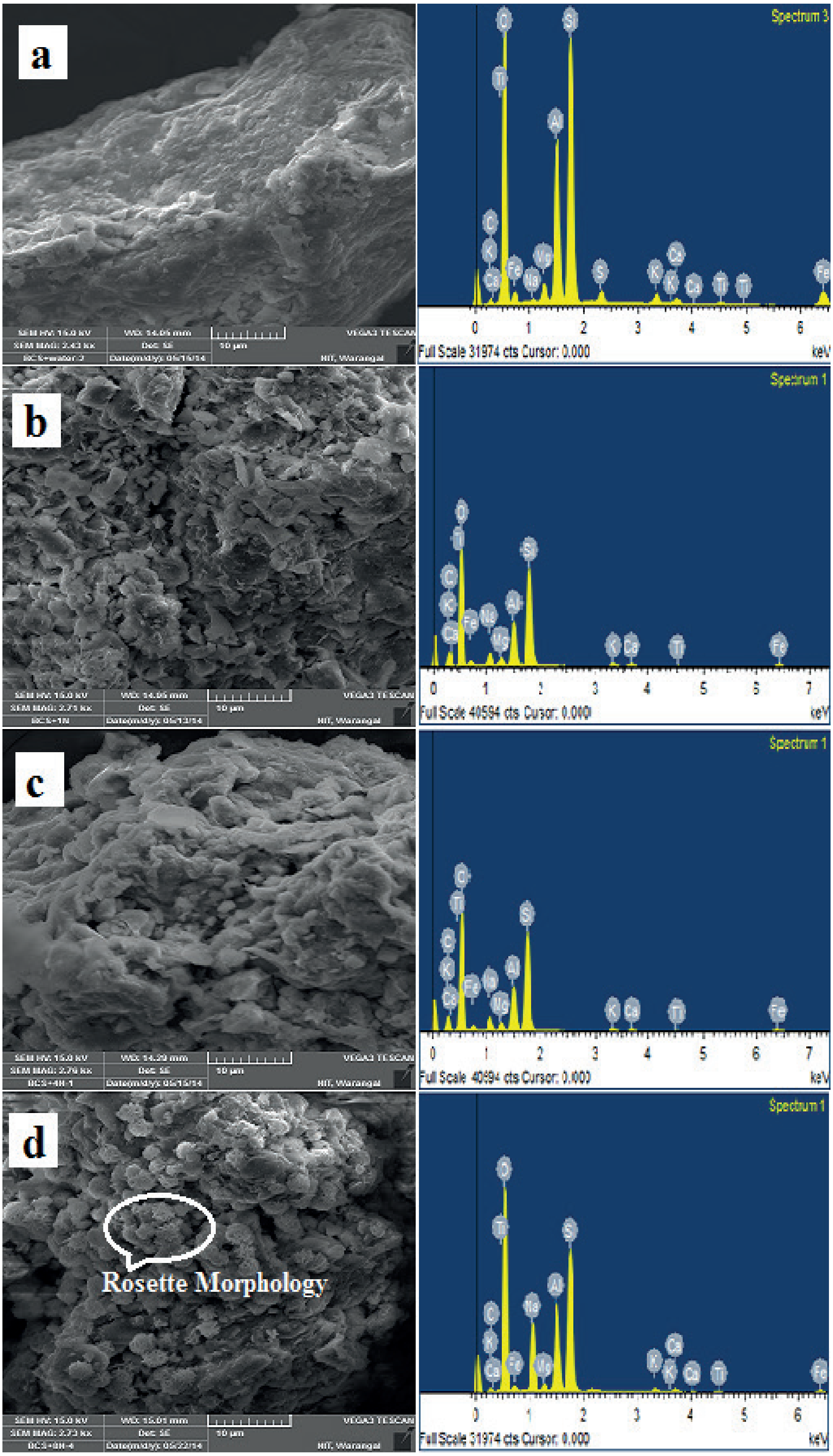

Fig. 4 Scanning electron microscopy and Energy dispersive analysis of X-ray images of black cotton soil after free swell with (a) Water (b) 1N NaOH (c) 4N NaOH and (d) 8N NaOH

4.4 Swell test results with H_2SO_4 solution as pore fluid

Swell test results of three specimens of black cotton soil with different concentrations of H_2SO_4solution as pore fluid (Fig. 5) demonstrate the opposite tendency when compared with the test data of the specimens with NaOH solution (Fig. 2). Figure 5 shows that a swell of about 10%, 45% and 51%, respectively was observed in specimens inundated with 1N, 4N and 8N H_2SO_4 solutions. Unlike the specimen with 1N NaOH, the percentage swell with 1N H_2SO_4 decreased when compared with that of water. However, at higher concentration the percentages swell increased, more than water, with increase in concentration of H_2SO_4. These variations in the percentage swell with different concentrations of H_2SO_4 are assessed by thoroughly investigating for mineralogical and microstructure changes.

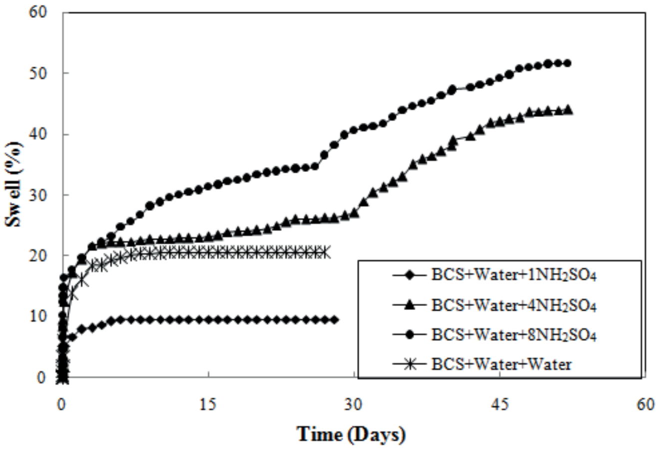

Fig. 5 Influence of pore fluids water and H_2SO_4 solutions on free swell behaviour of black cotton soil

4.5 XRD results after swell test with H_2SO_4 solution

The X-ray diffraction patterns for samples taken after swell test of natural black cotton soil inundated with water and varying concentrations of H_2SO_4 are shown in Figure 6. From the XRD pattern of soil inundated with 1N H_2SO_4 it was established that mainly sulphate based minerals, namely *Bassanite* (Peaks at 6.00, 3.00 and 2.80 [Å]) and *Gypsum* (Peaks at 7.63, 3.06 and 2.87 [Å]) which are calcium sulphate hydrate minerals, are formed. With an increase in concentration to 4N H_2SO_4 a new mineral (Peaks at 4.48, 4.39 and 3.46 [Å]), namely, *Alunogen* called as aluminium sulphate hydrate along with *Bassanite* is formed. Further increase in concentration to 8N H_2SO_4 showed peaks pertaining to *Alunogen* and *Gypsum* minerals.

4.6 SEM and EDAX results after swell test with H_2SO_4 solution

The SEM images along with EDAX of the soil specimens interacted with water, 1N H_2SO_4, 4N H_2SO_4 and 8N H_2SO_4 solutions are shown in Figure 7. The SEM image of BCS interacted with 1N H_2SO_4 solution (Fig. 7b) indicates that it has undergone some typical morphological changes upon interaction with the acid solution compared to the image (Fig. 7a) obtained after interaction with water. The particles with *needle like crystal* (Bassanite structure) morphology from image (Fig. 7c) show the extreme morphological changes with 4N H_2SO_4 solution. From the SEM image of soil after interaction with 8N H_2SO_4 solution (Fig. 7d) it has been observed that particles with a *six-sided thin prismatic plate* (Alunogen structure) morphology can be seen from the SEM image, which is an indication of morphological changes due to soil-acid reaction.

5 Discussions

It has long been recognized that the swell behaviour of soil is significantly affected by the nature of the pore fluid without any change in the external load. It is interesting to mention that acid and alkali aqueous solutions can have opposite effects on the free swell behavior of black cotton soil. At lower concentrations (1N) for sulfuric acid, the free swell for the soil was lower than that of distilled water, but for sodium hydroxide the opposite effect was observed i.e., maximum free swell. Similarly, at higher concentrations (4N and 8N) for sulphuric acid, the maximum free swell was observed; whereas, for sodium hydroxide (4N and 8N), the free swell was lower than that of distilled water. These variations in percent swell are clearly discussed below.

High percent of free swell (about 30%) observed in natural black cotton soil upon interaction with water was mainly due to the presence of volkonskoite, a smectite group mineral, which expands and increases the thickness of the diffused double layer near the negative surface of clay mineral particles due to hydration of cations present in the interlayer spacing. The percent swell of soil with 1N NaOH solution increased to about 54% compared to actual soil. This increase in swell was mainly due to the large scale formation of hydrous sodium carbonate bicarbonate, i.e. *Trona,* in the interlayer spacing which can be clearly observed from XRD pattern obtained (Fig. 3). Formation of *Trona* along with dissolution of components of bentonite namely smectite, opaline silica and quartz upon interaction with 1M sodium hydroxide were observed [16]. EDAX analysis of the specimen after swell test showed a decrease in silica percentage and increase in alumina percentage (Table 2). This is mainly due to alkali attack on the actual minerals of the soil. From XRD it can be observed that some peaks pertaining to *Microcline* (Fig. 4b) disappeared upon releasing silica while *Volkonskoite* mineral opens up octahedral sheet exposing alumina. The decrease in percent swell to 19% and 12%, respectively, with an increase in NaOH concentration to 4N and 8N was due to the dissolution of actual mineral components leading to formation of a new mineral, *Tosudite* (Fig.4b) in case of 4N and *Sodalite* (Fig.4c) in case of 8N, which stabilizes the soil and prevents high swelling. Based on these studies, it can be concluded that sodium hydroxide at higher concentrations can be used for stablizing the expansive soil. Similar observations were highlighted by Mulyukov [10].

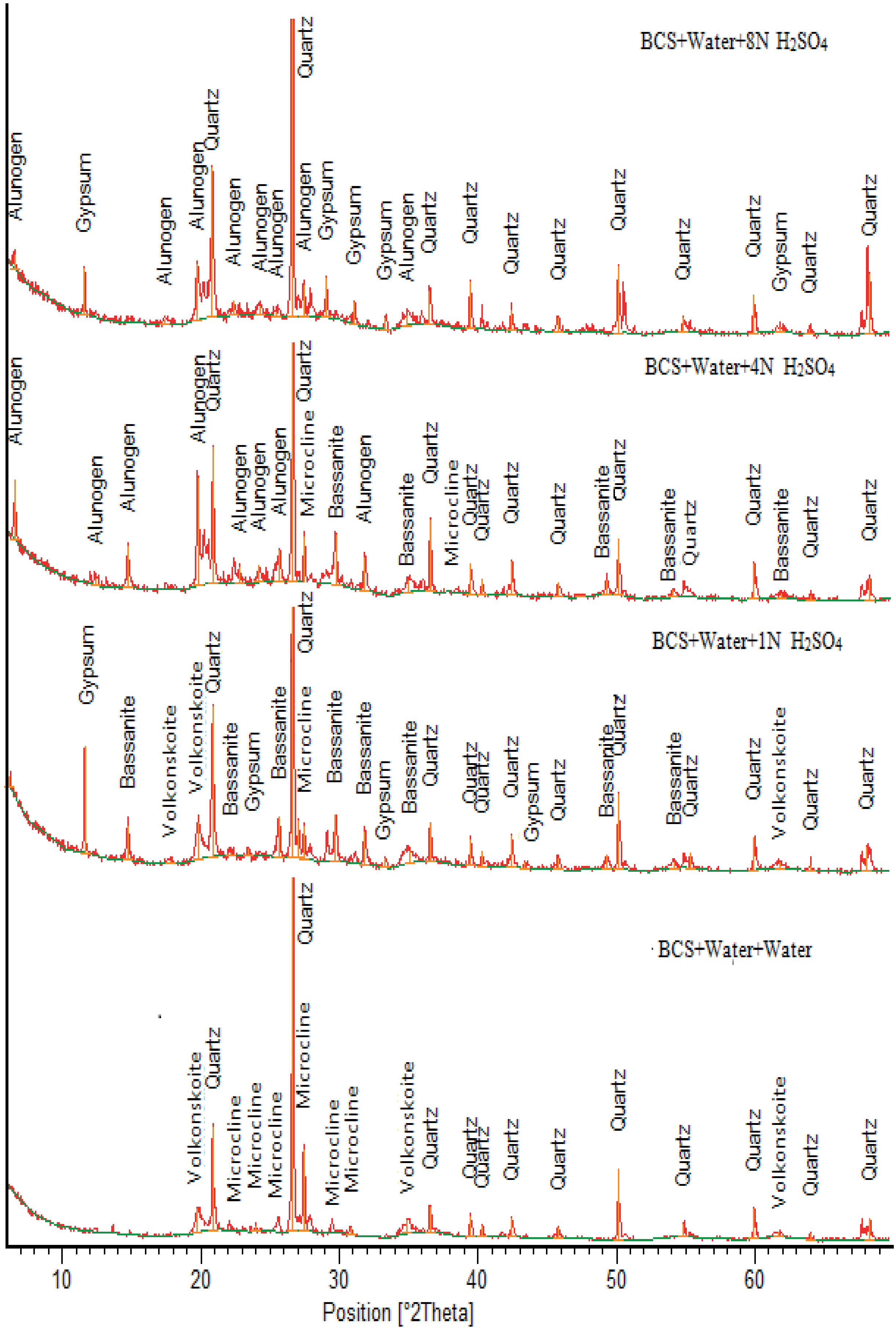

Fig. 6 X-ray diffraction patterns of black cotton soil after free swell with Water and H_2SO_4 solutions

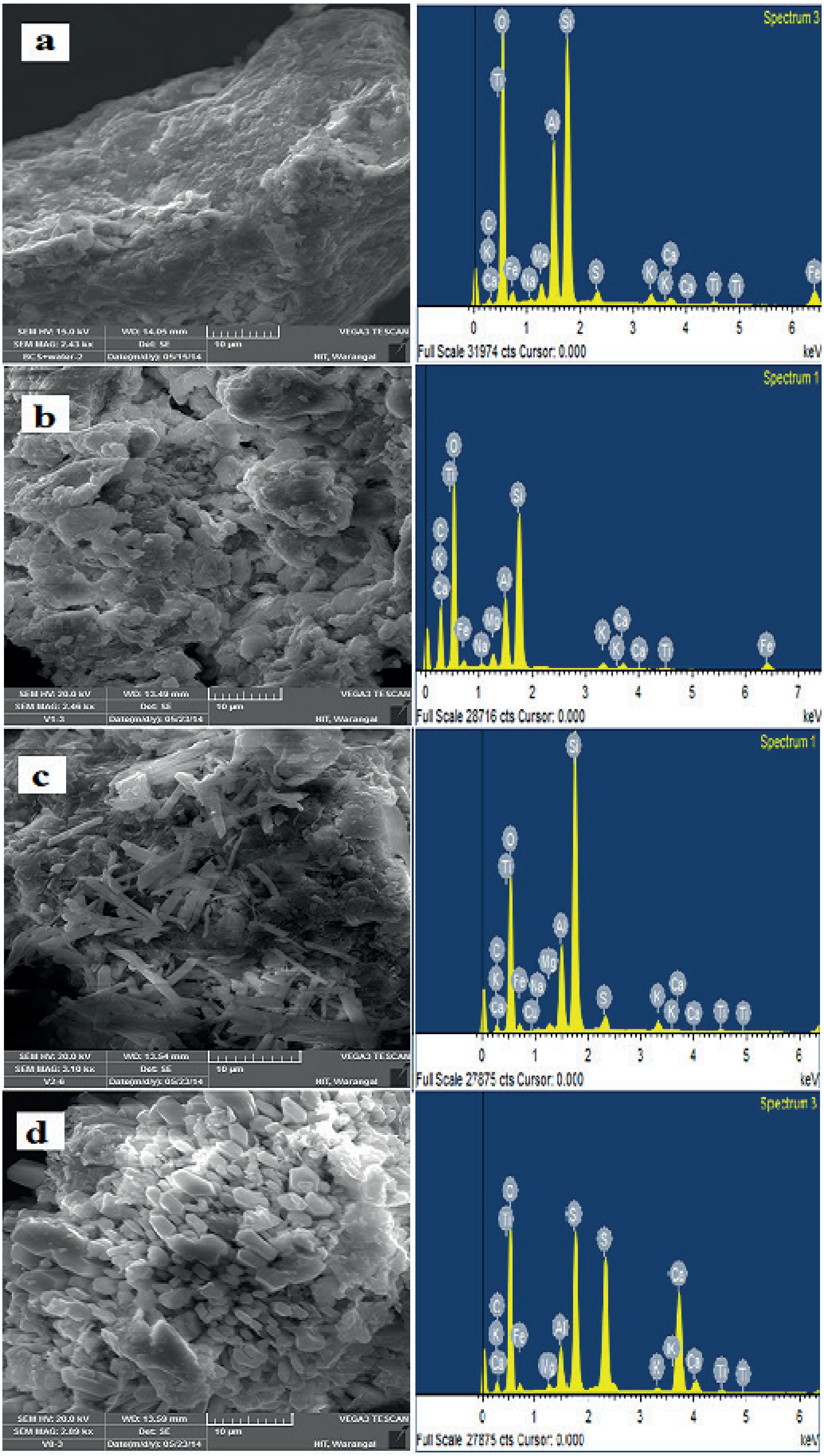

Fig. 7 Scanning electron microscopy and Energy dispersive analysis of X-ray images of black cotton soil after free swell with (a) Water (b) 1N H_2SO_4 (c) 4N H_2SO_4 and (d) 8N H_2SO_4

Table 2 Si/Al ratio as determined by EDAX analysis of black cotton soil inundated with water and NaOH solutions

Conc. of NaOH (N)	Si (Wt %)	Al (Wt %)	Si/Al
0	27.36	4.23	6.46
1	18.63	7.08	2.63
4	15.73	6.1	2.58
8	16.26	11.06	1.47

The decrease in swelling (10%) with 1N H_2SO_4compared with water is mainly due to the formation of new minerals as a result of exchange reaction (hydrogen ions replacing the calcium which combines with sulphates) which may chemically stabilize the soil. XRD data for 1N H_2SO_4, clearly confirms the formation of calcium sulphate hydrate minerals such as *Bassanite* and *Gypsum*. Further, with respect to acid attack on the actual mineral components, similar observation as explained with alkali solution can be made from EDAX analysis (Table 3).

Table 3 Si/Al ratio as determined by EDAX analysis of black cotton soil inundated with water and H2SO4 solutions

Conc. of H2SO4(N)	Si (Wt %)	Al (Wt %)	Si/Al
0	27.36	4.23	6.46
1	17.89	9.32	1.92
4	21.89	5.46	4.01
8	24.12	6.03	4.00

High swelling (45%) with 4N H_2SO_4 is mainly due to the combination of escaped aluminium (from lattice) with acid anion i.e. sulphate to form aluminium sulphate hydrate mineral (*Alunogen*), which heave more actively suppressing the stabilizing effect of *Bassanite* (Fig. 7c). Although swelling further increased (51%) with an increase in concentration to 8N H_2SO_4, due to the formation of *Alunogen*, the percentage increase in swell was reduced due to the stabilizing effect of *Gypsum* mineral formed (Fig. 7d).

From experimental results, it is established that in contrast to normal swelling which is due to increase in moisture content, in case of pollutant interacted soil, an increase or decrease in free swell is due to the synthesis of new mineral formations in the pore space of the soil.

6 Conclusions

Based on the obtained experimental results, the following conclusions can be drawn:

1. The free swell in black cotton soil is due to the high clay content and presence of swelling mineral Volkonskoite.
2. Prolonged interaction with both acid and alkali solutions lead to significant changes in free swell behaviour of black cotton soil.
3. In case of alkalis, free swell is higher, more than that of water at lower concentration and decreased, less than that of water, at higher concentrations. Whereas in case of acids the opposite trend is observed. These variations in free swell are mainly due to the influence of pore fluid on the mineral composition of soil.
4. Occasionally sodium hydroxide can be recommended as a stabilizing reagent in expansive soils based on its mineralogical composition.

Acknowledgments

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