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# Volume change and microstructure of calcareous soils contaminated with sulfuric acid

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ABSTRACT

Acid contamination of soils is a common problem within fertilizer and petrochemical industries. Soil properties could be altered due to acid contamination, especially the volume change within the vicinity of the contaminated soils. There are huge petrochemical and fertilizer industries in the industrial areas that produce or utilize different acids. Further, most of the soils used for road bases and under pavement and walkways are calcareous in nature. These carbonate soils are susceptible to large volume change when attacked by acids. This research aims to study the volume and microstructural changes using two calcareous soils, i.e. non-plastic and plastic marls, due to interaction with sulfuric acid at three different concentrations. The soil samples were prepared in high-density polyethylene (HDPE) molds that were placed in large containers and instrumented with linear variable differential transformer (LVDT) to measure the volume change. The morphology and composition analyses were studied utilizing scanning electron microscopy and X-ray diffraction. Results of this investigation indicate that the volume change and acid concentration were proportional except in the case with the non-plastic marl at high acid concentration. This was ascribed to the severe reaction of sulfuric acid with the carbonate minerals resulting in blocking the paths for further infiltration of the acid into the whole soil.



## 1. Introduction

The increase in industrial activities, especially chemicals and petrochemical, has resulted in environmental degradation of soils with unpredicted changes in their engineering behavior (Umesha et al., 2012). The extent of this change depends on various parameters including both type and concentration of the chemical contamination and the nature of the soils like particle size, mineralogical composition and specific surface area (Komnitsas et al., 1998). Contaminants have a substantial effect on the volume change behavior of soil, and in turn can have direct bearing on their geotechnical characteristics and affect the stability of buildings (Sivapullaiah et al., 2009). Contamination sources can be petroleum hydrocarbons, heavy metals, pesticides from industrial wastes, industrial-operation spills and leakages. Soil contamination with acid poses more challenges to the current geotechnical engineering practice because it comprises complex chemical reactions between soil and acid. Reports on acid contaminated soils revealed that dissolution of aluminosilicate of clay minerals with subsequent mineral formations leads to volume changes in soils (Sokolovich, 1995; Sivapullaiah et al., 2009). Stephenson et al. (1989) reported that the formation of gypsum resulted from the reaction of calcite with sulfuric acid has led to more than 18-inch (457-mm) upward movement of portions of electrolysis plant.

Among the acid contaminants, sulfuric acid is widely used in many industries such as copper leaching, inorganic pigment production, petroleum refining, paper production, and industrial organic chemical production. Rao and Reddy (1997) reported on the volume change characteristics of kaolinite contaminated with sodium phosphate/sulphate. Also, Sridharan et al. (1981) reported on heaving of soil due to acid contamination. Joshi et al., 1994 investigated the volume change in calcareous soils due to phosphoric acid contamination. They reported that the heave in soils was due to formation of brushite mineral in the presence of phosphoric acid. Al-Omari et al. (2007) investigated the effect of sulfuric and phosphoric acids on the behavior of a limestone foundation. They reported the use of chemical grouting by a two-shot injection of sodium silicate and calcium chloride to overcome the cyclic

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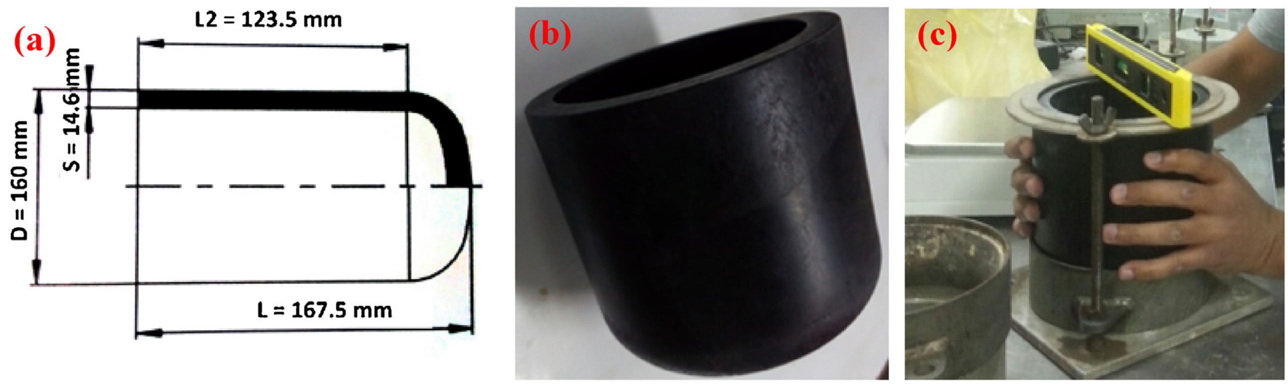


**Fig. 1.** (a) Dimensions of the end cap (mold), (b) HDPE end cap used as a mold to host the samples during acid contamination, (c) Leveling and fixing sample's mold in a CBR mold setup.

heave and settlement problem due to contamination of limestone foundation with acids.

Gratchev and Towhata (2009) investigated the effects of acidic contamination on the geotechnical properties of soils from coastal areas in Japan. They reported that acidic contamination could affect the compressibility of the clays, and the factors determining the degree of such changes were clay mineralogy, soil structure, and the duration of clay-acid interaction. The compressibility of kaolinitic soils typically increases due to soil-acid interaction (Sridharan et al., 1981; Gratchev and Towhata, 2008), while for bentonite the opposite effect can occur (Gajo and Maines, 2007). It has been reported that sulfuric acid-contaminated soils also reveal high swelling which resulted in distress to the structures built on them (Shekhtman et al., 1995). The influence of acid treatment on natural clay minerals was reported (Komadel, 2016; Zhang et al., 2012). Natural clays contain a variety of clay minerals, and thus the effect of contamination on the properties of such clays depends on the soil structure. Structures failures and tilting of acid storage tanks in factories such as in fertilizer industry due to accidental spillage or leakages of acids with the subsequent heaving of foundation soils were reported in several types of soils.

This work was concerned with the assessment of the volume and microstructural changes of two most common calcareous soils (M1 is the non-plastic marl, and M2 is the low plastic marl) due to interaction with sulfuric acid at three different concentrations.

## 2. Experimental

### 2.1. Soil procurement and characterization

Two different types of marl were collected from two sites in the Eastern Province of Saudi Arabia, whereby the two soils were being utilized for constructional purposes. Samples of the non-plastic marl (M1) and the low plastic marl (M2) were sieved through ASTM Sieve #4 (4.75-mm opening) and allowed to dry in the open air for few days to expel the moisture. Thereafter, they were stored in large plastic drums till testing. The soil samples were then subjected to soil characterization tests that included grain size analysis, specific gravity, consistency limits and standard Proctor compaction.

The specific gravity was determined in accordance with ASTM D 854. The average value of triplicate samples for each soil was used as a representative value. Sieve analysis test was conducted as per the ASTM D 422 using both dry and wet analyses. For the particles passing through sieve No. 200 (75 μm opening size), the hydrometer test was conducted in order to determine the silt and clay content in each of the two soils.

The liquid limit and plastic limit tests were conducted on the two marl soil samples according to ASTM D 4318. For M1 soil, it was not possible to get the number of blows for the liquid limit test and, hence, the liquid limit was reported to be nil. In addition, the soil samples could not be rolled to a thread of 1/8 in.(3.18 mm), therefore, the soil was classified as non-plastic. Standard Proctor compaction test was conducted in accordance with ASTM D 1557 to determine the maximum dry unit weight and optimum moisture content of the two soils.

### 2.2. Testing program for acid contamination

In order to study the effect of acid on calcareous materials in the field, several steps were simulated in the laboratory. Firstly, the soil samples had to be prepared in suitable molds that could resist the acid without any corrosion or deterioration. Then, the molds had to be placed and leveled in a proper container/bath that had to be made of acid-resistant material and accommodate four soil specimens. After that, linear variable differential transformers (LVDTs) had to be mounted on the samples, and the acid had to be poured gently in the bath just after the data logger was initiated to measure the volume change of the soil using the LVDT's. The inundating solution is introduced into the sample from the bottom.

The soil baths were placed within a vacuum hood to prevent any fume or acid dispersion into the laboratory. The volume change was monitored for a period of two weeks from the starting time of contamination. This duration was selected based on the behavior of expansion in the preliminary round of contamination where the swelling was observed to cease after two weeks. Subsequently, the soil samples had to be assessed/analyzed in terms of microstructure and mineralogical changes.

### 2.3. Preparation of soil samples

Metallic molds could not be used in this experiment because the metal would not resist aggressive acids and corrosion was noted within few days even if stainless steel mold was used (Alshammari, 2017). For that reason, a proper sample mold (i.e. end cap of a mold), made from high-density polyethylene (HDPE), was selected for this experimental program. The mold has a length (L) of 167.5 mm, with 160 mm diameter (D) and 14.6 thickness (S), as shown in Fig. 1. However, the length from the opening to the point before the curvature (L2) is 123.5 mm. The mold has very small curvature at the bottom with 12 holes of 2.5 mm diameter were drilled in the bottom in a direction parallel to the length of the mold to allow the acid to infiltrate inside the mold. Moreover, to fit the soil sample in a cylindrical shape without the end curvature, polypropylene pellets were filled inside the curvature of the mold and they were encapsulated by a geotextile made of polypropylene.