

Compressibility of soils subjected to long-term acidic contamination

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ABSTRACT: Laboratory investigation was conducted to study the effects of acidic contamination on compressibility of two natural soils from the Tokyo area. A special container was designed to reproduce in the laboratory conditions the process of long-term soil-water-acid interaction that typically occurs during municipal and industrial waste storage. Solutions of sulfuric acid were used to leach high-quality soil samples for different time periods ranging from 1 to 6 months. The obtained results show that acidic contamination can significantly alter the compressibility of the soil, and the factors determining the degree of such changes are clay mineralogy, soil structure, and the duration of contamination.

1 INTRODUCTION

In response to the growing rate of soil and water contamination, a good deal of research has been conducted in the past few decades to evaluate the effect of chemicals on the properties of soil, as well as to refine the existing methods of field and laboratory investigation. The literature review indicates that the majority of studies have mostly focused on the influence of non-organic fluids on the geotechnical characteristics of soil while only few works were aimed at understanding the effect of acidic fluids on the behavior of soil.

Only recently, Wang and Siu (2006), Gajo and Maines (2007), Gratchev and Sassa (2009) have conducted systematic studies to determine whether the properties of soil can be affected by changes in pH environment. Although the aforementioned research has provided useful insights into the mechanisms of soil-water-chemical interaction, there are still some aspects of soil behavior that need to be addressed. For example, a number of studies have been performed on artificial soils or mixtures containing one dominant clay mineral such as montmorillonite or kaolinite. However, the majority of natural clays generally have a more complex mineral composition, and thus the effects of contamination on the properties of such soils still remain unclear. In addition, due to the low permeability of natural clays, it has become common practice to use remolded soils for testing, as it provides satisfactory results within a reasonable period of time. As a consequence, the role of soil microfabric and the duration of contamination have been generally neglected. To address these issues, a laboratory study was conducted at the University of Tokyo, Japan. Two natural soils with different mineral composition were collected from the Tokyo area and leached with acidic liquids for certain periods of time. A series of compression tests were performed to establish whether contamination could

alter the compressibility of soil. This article presents the obtained results.

2 REVIEW OF THE PREVIOUS WORKS

Recent studies (Imai et al. 2006, Wang and Siu 2006, Gajo and Maines 2007, Gratchev and Sassa 2009, and Gratchev and Towhata 2009) have shown that several major processes can occur in clay as a result of changes in pH environment; however, the degree to which each of them may affect the geotechnical properties of soil greatly depends on the mineralogy of clay fraction. Mitchell (1993) noted that the processes altering the diffuse double layer would have a significant impact on soils with montmorillonite/smectite while changes in particle orientation caused by chemicals should have a dominant effect on the properties of kaolinitic soils. A brief description of each process as well as its possible effect on the geotechnical properties of clay, including its compressibility, is given below.

When acid comes in contact with naturally consolidated clays, it may destroy/dissolve the chemical bonds or cementation between clay particles, forming relatively "loose" structures. Imai et al. (2006) leached a clay from the Osaka Bay with solutions of hydrogen chloride and reported that the clay structure originally cemented by calcium carbonates was partially destroyed by the acid as it dissolved the carbonate bonds between clay particles. Results of compression tests on the leached specimens indicated that the consolidation yield stress of soil decreased with decreasing calcium carbonate content.

It is believed that, when the environment becomes slightly acidic, hydrogen ions (H^+) engage in the exchange process with the cations from the diffuse double layer of clay particles. Due to its superior position in the Hofmeister series (Hof-

meister series describes how readily some cations are absorbed at a given concentration), proton H^+ ions would likely replace the commonly found exchanged cations such as Na^+ , Ca^{2+} , Al^{3+} , or Fe^{3+} . From clay colloid chemistry (Olphen 1991), it is known that this process would lead to an increase in double-layer thickness, resulting in a greater compressibility of soil. Results of compression tests conducted by Sridharan et al. (1986) on bentonite prepared in water with a specific type of ions lend support to this hypothesis. The investigators reported that bentonite "homonized" with ions of smaller valence had a higher compressibility.

As contamination continues, the amount of acid in the pore fluid may considerably increase, reaching the concentration at which the collapse of the diffuse double layer would occur (Mitchell 1993). Laboratory data show that this process will alter properties of soil such as hydraulic conductivity (D'Appolonia, 1980; Ruhl and Daniel, 1997; Kashir and Yanful, 2001), compressibility (Gajo and Maines, 2007) and plasticity (Sridharan et al., 1988; Gratchev and Sassa, 2009). However, it is believed that such changes in the diffuse double layer would affect montmorillonitic soils in a greater manner than kaolinitic soils (Mitchell 1993).

It has long been recognized that, in acidic medium, the charge on the edges of clay particles would become increasingly positive due to the adsorption of H^+ ions, resulting in flocculated, face-to-edge (F-E) structures (Olphen 1991). This mechanism is typically associated with kaolinite, whose variable charge is highly pH dependent (Mitchell 1993). Results of laboratory tests conducted on kaolinitic soils indicate that clays with an open-flocculated structure exhibit greater strength and permeability (Mitchell 1993, Sridharan and Prakash 1999). Such soils can also be expected to have a higher liquid limit because of the greater amount of water that is entrapped within large voids created by F-E associations (Sridharan et al. 1988). Wang and Siu (2006) presented experimental evidence, including results of Atterberg limits tests and SEM photographs, lending more support to this hypothesis.

It should be noted that the aforementioned mechanisms are dominant for the pH range of about 3-6, the range of values that is commonly encountered in natural systems. Olphen (1991), Mitchell (1993) noted that at extremely high concentrations of acid ($pH \approx 1$), significant changes in the mineral structure may occur (for example, dissolution of Al^{3+}), a process that would likely affect compressibility of soil. To avoid such an alteration of the mineral composition, only light concentrations of acid were used in this research.

From the above review, it can be inferred that the degree to which acidic contamination can affect compressibility of clay depends greatly on the mineralogy of clay fraction. Laboratory data obtained for montmorillonitic soils indicate that in acidic medium, high concentrations of H^+ -ions would suppress the diffuse double layer, causing decreases in the soil's compressibility as well as permeability and liquid limit. In contrast, kaolinite particles would likely form a flocculated structure with the dominant face-to-edge particle associations, resulting in greater compressibility, permeability and higher values of the liquid limit. It should be noted that although the effect of each mineral on the properties of clay in an acidic medium seems to be well-understood, the behavior of natural clays that contain both or several clay minerals still remains unclear. In addition, the soil structure also seems to have a large impact on the geotechnical properties of soil, especially in the case of consolidated clays with chemical bonds or cementation. However, this issue has been poorly studied as leaching and testing of high-quality soil samples typically require a lengthy time frame. This study seeks to investigate the process of long-term acidic contamination and its effect on the compressibility of natural soils with a variety of clay minerals.

3 SOILS TESTED AND LIQUIDS USED

High-quality samples of Yurakucho clay and Kawasaki mud were used in this research. The former was excavated from the depth of about 20 m at a construction site in Tokyo, Japan, cut into smaller blocks and carefully transported to the geotechnical laboratory of the University of Tokyo. The X-ray diffraction analysis showed that kaolinite was the major clay mineral. The results of Atterberg limits tests conducted on specimens with distilled water showed that the soil was plastic with the liquid limit (LL) of 50.9, and the plasticity index (PI) of 15.8.

The Kawasaki mud was dredged from the Tokyo Bay near Kawasaki-city, Japan. It contained montmorillonite and chlorite as the dominant clay minerals, and had the liquid limit and plasticity index of 62 and 32, respectively.

Distilled water ($pH=6.9$) and solutions of commercially available sulphuric acid (H_2SO_4) were used in this study. Sulphuric acid was diluted to the desired concentrations with pH values of 3.0 and 4.0. All pH measurements were performed by a pH meter (accuracy ± 0.01).

4 TEST PROCEDURE

4.1 Buffer Capacity

Buffer capacity is the property of a soil to absorb and/or desorb H^+ and OH^- ions, and thus it determines a resistance of soil to pH changes. In this work, the buffer capacity of studied soils was determined experimentally by titrating each clay with increasing concentrations of sulphuric acid (Yong et al. 1990). The acid solutions were first prepared at certain concentrations, then added to the soil at a ratio of 1:10 for soil: acid solution, using 4 g of dry soil and 40 g of acid solution. The pH of the soil solution was measured after the soil suspension sample was thoroughly shaken and allowed to stand for at least 24 h.

4.2 Specimen Preparation

To reproduce long-term soil-water-acid interaction, a special container as schematically shown in Fig. 1 was designed (Gratchev and Towhata, 2009).

The soil sample was sandwiched between two flat perforated PVC plates, which were 5 mm thick with circular holes of diameter 6 mm at 6 mm spacing. Filter paper was placed in between the sample and the plates in order to prevent loss of the soil. During leaching, the sample was kept confined under certain weight (≈ 4 kg) in order to minimize swelling. PVC pipes were used as supporting columns as shown in Fig. 1, providing an open area for an acidic liquid. Undisturbed samples of Yurakucho soil were carefully cut into smaller specimens with a height of about 2.2 cm, placed in the containers, and kept confined in order to minimize swelling. A liquid of certain pH was carefully poured into the container, resulting in the interaction between the soil and contaminated water. The container was covered with the lid in order to prevent vaporization of the fluid. After a certain period of time, the liquid was drained off the container, and its pH was measured by means of the pH meter. The experience acquired during testing showed that 7-8 days were sufficient for chemical equilibrium. Then, the container was filled again with the original solution, thus maintaining the desired value of pH throughout the tests. After the end of each time interval, a certain amount of soil was retrieved from the container, and compression tests were performed.

For the Kawasaki mud, a marine deposit with a high buffer capacity, acidic liquids with a low pH=3.0 were used to decrease the pH of pore fluids within reasonable periods of time. It is noted that in the case of Kawasaki mud, compression tests were conducted as the pH of pore fluid decreased by 0.2-0.4.

Consolidation was carried out in a standard 6 cm – diameter, and 2 cm- height, one-dimensional oedometer. Overburden stress was applied in increments, according to Japanese Test Standard (JIS A 1217), followed by unloading. The oedometer was covered with a polythene wrap during soil consolidation to minimize chemical vaporization of pore fluids.

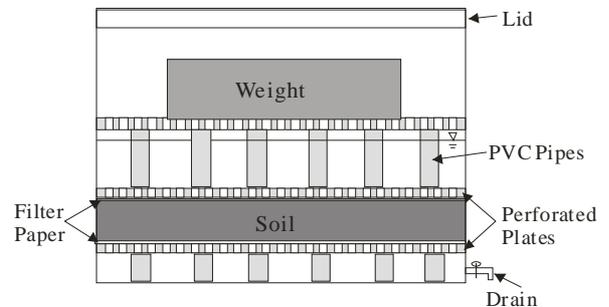


Fig. 1 A container used to leach soil with acidic water.

5 TEST RESULTS

5.1 Buffer Capacity

The titration curves of pH against sulfuric acid input for the clay suspensions are shown in Fig. 2. Also included in the figure is the titration curve of a blank (a solution in the absence of soil). The titration curve of the Kawasaki mud shows that the soil suspension can resist acid input with small changes in pH, decreasing gradually from 7.3 to 6.2. When the amount of acid reaches and exceeds $100 \text{ cmol } H_2SO_4 \text{ kg}^{-1}$ soil, the suspension pH drops to about 4. In the case of the Yurakucho clay, the suspension pH already decreases to a small value of 3.5 at relatively light concentrations of sulfuric acid. The obtained results indicate that the Kawasaki mud has a significantly higher buffer capacity than the Yurakucho clay, and thus greater amounts of acid would be needed to decrease the pH of pore fluids in this soil. It is noted that the high resistance of the Kawasaki mud to pH changes is likely due to their mineral composition, which includes montmorillonite, a mineral with a high cation exchange capacity (C.E.C.).

5.2 Yurakucho clay

5.2.1 The effect of long-term acidic contamination

Compression test results with distilled water, and solution of acidic water with pH=4.0 are presented in Fig. 3. As can be seen in this figure, the virgin compression curves for all specimens were placed in the order of the contamination time with the uppermost curve being with distilled water and the

lowest one with pH=4.0, after 6 months. At a given overburden stress, the void ratio decreased with an increase in the time of contamination. From this figure it can be inferred that the longer the soil is subjected to contamination, the greater changes in soil properties it undergoes.

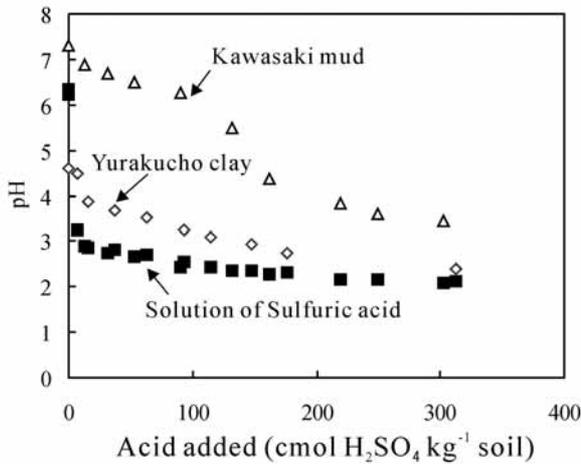


Fig. 2. pH-titration curves of the studied soils.

5.2.2 The effect of acidic concentration

The concentration of contamination seems to have a strong influence on the compressibility of Yurakucho clay as well. Results presented in Fig. 4 indicate that the specimen leached with acidic liquid of pH=3.0 had higher void ratios, suggesting that a significant increase in acidic concentrations (that is, a decrease in pH values) can cause the formation of a more open soil structure. Figure 5, which is plotted as pH against the compression index, seems to lend more support to this assumption. It is clear that the compression indices increased as the concentration of acid in the pore fluid also increased.

5.3 Kawasaki mud

Laboratory data from compression tests performed on specimens of Kawasaki mud at different pH values are presented in Fig. 6. As can be seen in this figure, the virgin compression curves for all soil specimens were placed in the order of pH values with the uppermost curve having pH=8.39 and the lowest one having pH=6.12. The test data also show that at a given overburden stress, the void ratio decreased with a decrease in pH.

Results of compression tests summarized in Fig. 7 show the reverse tendency, compared with that observed for the Yurakucho clay (Fig. 5). Unlike the Yurakucho clay, the compression index of the Kawasaki mud clearly decreases with a decrease in the pH. It is noted that similar results were reported by Meegoda and Ratnaweera (1994), Gajo and Maines (2007), who tested artificial mix-

tures and remolded clays with montmorillonite/smectite as the dominant clay mineral.

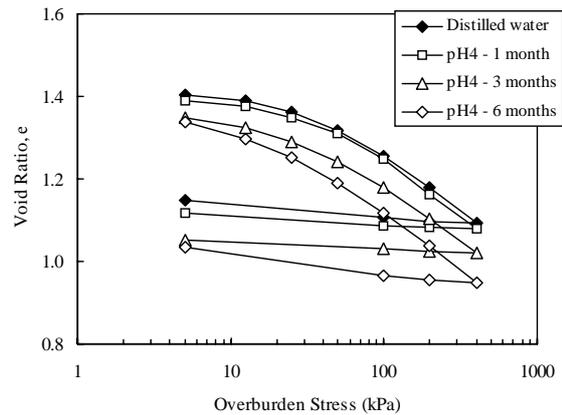


Fig. 3 Results of compression tests on Yurakucho clay leached with acidic liquid of pH=4.0 for 1, 3, and 6 months.

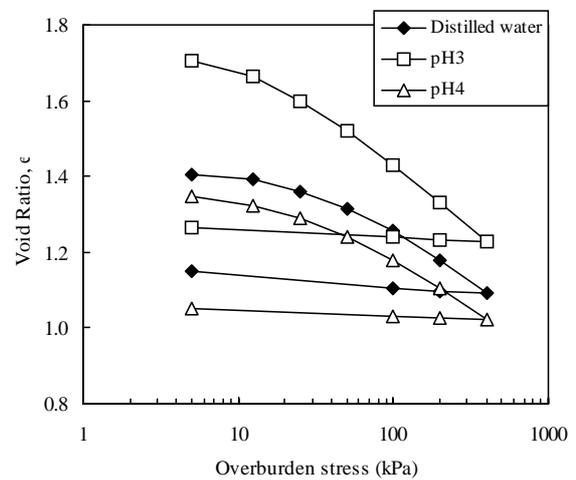


Fig. 4 Results of compression tests on Yurakucho clay leached with acidic water of pH=4.0, and pH=3.0.

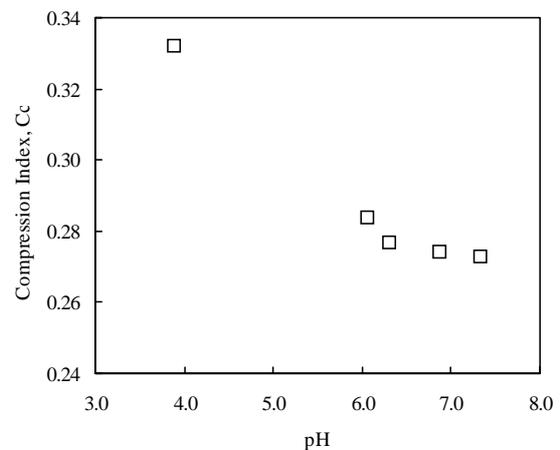


Fig. 5 Results of consolidation tests on Yurakucho clay plotted as pH against the compression index.

6 DISCUSSION OF TEST RESULTS

The results presented in Figs. 5 and 7 indicate that acidic contamination can have opposite effects on the compressibility of the studied soils. For the Yurakucho clay, it was found that a decrease in pH increased the compression indices of soil, while for the Kawasaki mud, the effect was opposite.

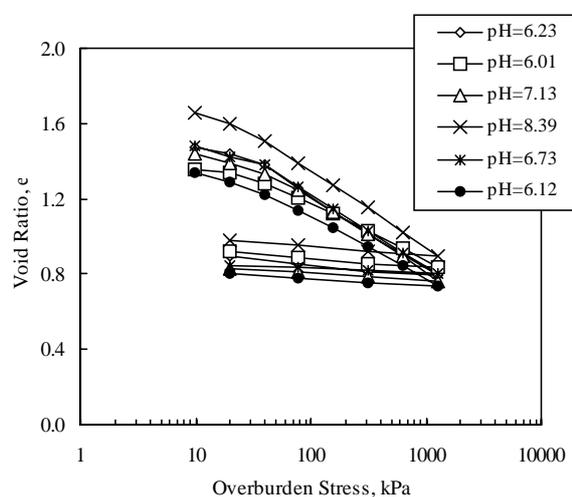


Fig. 6 Results of consolidation tests on Kawasaki mud leached with solutions of sulfuric acid.

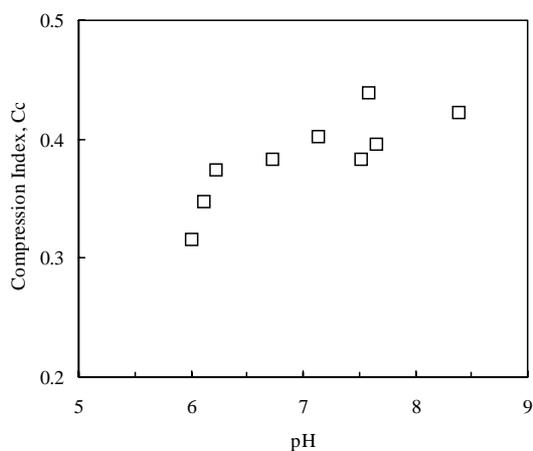


Fig. 7 Results of consolidation tests on Kawasaki mud plotted as pH against the compression index.

Form the available literature, it can be inferred that the influence of pH on the properties of fine-grained soils is complex and depends greatly on the mineralogy of clay fraction. Results of numerous studies on clay colloid chemistry suggest that changes in pH can generally affect soil structure through two major mechanisms: 1) by changing the charge on the edges of clay particles, resulting in flocculated or dispersed structures; and 2) by decreasing the thickness of the diffuse double layer. It is believed that the former prevails in kaolinite while the latter is of great importance for montmorillonite

Olphen (1991) and Wang and Siu (2006) noted that, in acidic medium, kaolinite particles form a flocculated structure with the dominant face-to-edge particle associations, resulting in a more open soil structure. As a result, the void ratio of kaolinitic soil is expected to increase, leading to a greater compressibility.

The results obtained for the Kawasaki mud, a soil that contains montmorillonite, can be interpreted on the basis of the double-layer theory (Olphen, 1991). It is believed that, in acidic medium, due to high concentrations of H^+ -ions, the diffuse double layer becomes suppressed, causing a decrease in void ratio. Gajo and Maines (2007) conducted a series of compression tests on bentonite leached with acidic fluids and pointed out that the collapse of the diffuse double layer resulted in a significant decrease of the soil's compressibility.

7 CONCLUSIONS

Laboratory investigation of two clays from the Tokyo area subjected to long-term soil-acid interaction was conducted in order to study the influence of contamination on the compressibility of the soils. Based on the obtained results, the following conclusions can be drawn:

- Long-term clay-water-acid interaction can lead to the alternation of compressibility of the studied soils;
- Clay mineralogy and the duration of clay-acid interaction appear to be the key factors determining the degree to which changes in the clay's properties can occur during contamination;
- For the Yurakucho clay, long-term acidic contamination resulted in higher compressibility, while for the Kawasaki mud, the effect was opposite.

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