

Compressibility of natural soils subjected to long-term acidic contamination

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Abstract.

This paper presents the results of a systematic analysis aimed at establishing whether acidic pore fluids can affect the properties of natural soils, in particular their compressibility. Marine deposits with different mineral composition and undisturbed soil structure were collected for this research from three coastal areas in Japan. Pleistocene clays from the Osaka and Ariake Bays were obtained from boreholes at a depth of more than ten meters while the Kawasaki mud, a relatively young deposit of Holocene, was dredged from the bed of the Tokyo Bay. Soil specimens were placed in special containers, which were designed to reproduce the process of long-term soil-water-chemicals interaction, and leached with solutions of sulfuric acid for different periods of time, ranging from one to nine months. At the end of each time interval, standard compression tests were performed to study the behavior of soil in an acidic environment. It was found that clay mineralogy and soil structure had a significant effect on the compressibility of clays at low pH. In the case of the Osaka and Ariake clays, the compressibility significantly increased with a decrease in pH values, a finding that was primarily attributed to changes in the soil's structure. In contrast, the effect of acidic leaching on the properties of Kawasaki mud was observed to be the opposite. Laboratory data showed that in acidic medium the compressibility of soil decreased presumably due to the collapse of the diffuse double layer.

KEY WORDS: acid; contamination; clay; compressibility; buffer capacity; clay mineralogy; soil structure.

INTRODUCTION

In mountainous countries like Japan where the land available for construction is generally limited to coastal areas, new development projects often face the problem of soil contamination caused by mismanagement of the land in previous years. Although geotechnical practice indicates that engineering constructions built on such soils can experience sufficient deformations due to changes in the soil's compressibility (Sridharan et al. 1981; Rao and Rao 1994), geotechnical engineers are faced with the dilemma whether to give this factor a special consideration or not, mostly due to the lack of standard regulations concerning this type of soil.

The research conducted in the past few decades shows that changes in the pore water chemistry may significantly alter the compressibility of soil. Bolt (1956) was probably the first to present results of a systematic analysis regarding effects of physicochemical factors on the soil's compressibility. Later, Rosenqvist (1959), Mitchell (1960) showed that the compression indices of clay were controlled by the characteristics of ions such as the type, valence, and concentration in the pore fluid. Subsequent to these studies, Olson and Mesri (1970), Sridharan and Rao (1973) carried out compression tests on bentonite and kaolinite with various chemical liquids as pore fluids and noted the important role of clay mineralogy in the soil behavior. More recently, Meegoda and Ratnaweera (1994), Chen et al. (2000), Wang and Siu (2006b), Sunil et al. (2006), Gajo and Maines (2007) have provided useful insight into the effects of the dielectric constant of organic pore fluids, pH environment, and electrolyte concentration on the compressibility of clays.

Although the aforementioned studies have identified several important factors that may affect compressibility of soil, there remain some aspects of soil-water-chemical interaction that need to be addressed: 1) the majority of studies have focused on the effect of organic fluids on the compressibility of clay while only little attention was devoted to the behavior of soil at different pH environments. In light of the growing rate of acidic contamination (Kamon et al. 1997; Golab and Indraratna 2009), additional research is needed to clarify the influence of pH on the geotechnical properties of soil. 2) The previous research regarding the pH-dependent properties of soil has generally been concerned with clay mixtures, which typically contain only one dominant clay mineral, either montmorillonite or kaolinite. However, naturally occurring clays have a more complex mineral composition that may include a variety of clay minerals. 3) Due to the low permeability of tested clays, it has become common practice to use remolded soils during laboratory investigation, as it provides satisfactory outcome within a reasonable period of time. As a result, the effects of soil microfabric and the duration of contamination have been neglected. This study seeks to shed light on these issues by providing

laboratory data regarding the compressibility of natural undisturbed clays that were subjected to long-term acidic contamination.

In this work, soil samples from three coastal areas in Japan were used. X-ray analysis, Atterberg limits and grain size distribution tests were carried out to determine the index properties of the studied soils. The buffer capacity was determined experimentally by measuring the pH change when strong acid was added to the solutions. To reproduce the process of long-term contamination, the specimens were leached with acidic liquids for specific periods of time ranging from one to nine months. A series of compression tests were performed at the end of each time interval in order to establish the relationship between pH and compressibility of soil. This article reports and discusses the obtained results.

REVIEW OF THE PREVIOUS WORKS

Although a substantial amount of research has been conducted in the past few decades to investigate the effect of pH on the rheological characteristics of clay suspensions (Benna et al. 2001; Jozefaciuk 2002), it was not until recently that Imai et al. (2006), Wang and Siu (2006a,b), Gajo and Maines (2007), Gratchev and Sassa (2009), Gratchev and Towhata (2009) performed systematic studies aimed at determining whether pH can influence the properties of soils that are typically encountered in geotechnical practice. These recent works, which elaborated and clarified the results of previous research (D'Appolonia 1980; Kamon et al. 1997; Ruhl and Daniel 1997; Kashir and Yanful 2001), have provided valuable insight into the role of pH in determining the geotechnical properties of clay. It has been 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), Sridharan and Prakash (1999) 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. It is noted that the focus of this review is directed towards the behavior of soil in an acidic environment ($\text{pH} < 7$). The reader who is interested in the effect of an alkaline medium on the properties of clay is referred to Wang and Siu (2006a,b), Gratchev and Sassa (2009).

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 (Hofmeister 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), cyclic strength (Gratchev et al. 2006; Gratchev et al. 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; Ma and Eggleton 1999; Li and Li 2000). 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 (2006a) 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.

SOILS TESTED AND LIQUIDS USED

The marine soils used in this research were kindly provided by the Port and Airport Research Institute, Japan. Undisturbed soil samples of the Osaka Bay clay were collected from a borehole at the elevation of -44 m (the water depth was about 20 m) during the second construction phase of the Kansai International Airport in Osaka. The geotechnical properties of this clay have been studied by Watabe et al. (2002), Tanaka and Locat (1999), who pointed out that the liquid limit (LL) and plasticity index (PI) of the Osaka clay varied with the elevation: LL was estimated to be in the range of 75-90, and PI in the range of 50-55. The soil samples tested in this work had a liquid limit of 66.4, and a plasticity index of 34.8 (Table 1). X-ray analysis conducted at the Analysis Center Co., Ltd., Japan, revealed the presence of clay minerals such as smectite, chlorite, kaolinite and illite. In addition, this clay contained a high amount of calcite, which comprised about 2% of the total mineral composition.

Borehole samples of the Ariake Bay clay were obtained at a depth of 11 m. Sridharan et al. (2002) studied in detail the mineral composition of this clay and reported that smectite, kaolinite, illite, and vermiculate were the dominant clay minerals. Egashira and Ohtsubo (1983) also noted that the smectite of Ariake clay may contain as much as 10% Fe_2O_3 , and that the clay generally exhibits little intracrystalline swelling. The liquid limit and plasticity index were estimated to be 82.0 and 30.6,

respectively (Table 1).

The Kawasaki mud was collected in bulk from the Kawasaki Port in Tokyo Bay, and stored as slurry in a sealed container in the laboratory. This slurry was used to form samples of the same moisture content, which were then leached with acidic fluids for different periods of time. The mineral composition included montmorillonite and chlorite. The liquid limit and plasticity index were 58.4 and 25.6, 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 produce acidic liquids with pH=3.0. All pH measurements were performed by a pH meter (accuracy ± 0.01).

EXPERIMENTAL PROCEDURE

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, and 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.

Specimen Preparation

To reproduce the process of long-term soil-water-chemicals interaction, a special container as schematically shown in Fig. 1 was designed. 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 soil specimen and the plates in order to prevent loss of the soil. The specimen was kept confined under the pressure of 10 kPa to ensure the same stress conditions during leaching. PVC pipes were used as supporting columns as shown in Fig. 1, providing an open area for an acidic liquid.

Undisturbed borehole samples of Osaka and Ariake clays with a diameter of 10 cm were carefully cut into smaller specimens with a height of about 2.2 cm, placed in the containers, and leached with an acidic liquid of pH=3.0 for different periods of time. In the case of the Kawasaki mud, the soil with moisture content close to its liquid limit was used. The container was covered with the lid to prevent vaporization of the fluid. After seven to eight days, a period of time that proved to be sufficient for

chemical equilibrium, the liquid was removed from the container, and the liquid's pH was measured. After the measurement, the container was re-filled with the original solution of pH=3.0 to maintain the desired value of pH throughout the test. Figure 2 shows changes in the liquid's pH recorded over a 300-day period for specimens of the Osaka Bay clay. As the pH values of the liquid decreased, a specific amount of soil was retrieved from the container and compression tests were performed.

Compression tests were carried out in a standard 6 cm – diameter, and 2 cm - height, one-dimensional oedometer. Overburden stress was applied in increments of 9.8, 19.6, 39.2, 78.5, 157, 314, 628, and 1256 kPa, according to the 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. The compression index C_c , or the slope of the virgin compression curve, was computed as follows: $C_c = (e_{157\text{kPa}} - e_{1256\text{kPa}}) / (\log(1256\text{kPa}/157\text{kPa}))$.

TEST RESULTS

Buffer Capacity

The titration curves of pH against sulfuric acid input for the three clay suspensions are shown in Fig. 3. Also included in the figure is the titration curve of a blank (a solution in the absence of soil). The Ariake clay suspension has a slightly lower initial pH (6.8) than the Osaka Bay clay (pH 7.0), and the Kawasaki mud (pH 7.3). The titration curve of the Kawasaki mud shows that the soil suspension can resist acid input with small changes in pH, dropping slowly from a pH value of 7.3 to a value of 5.8. When the amount of acid reaches and exceeds 100 cmol $\text{H}_2\text{SO}_4 \text{ kg}^{-1}$ soil, the suspension pH drops gradually to 4. A similar tendency is observed for the Osaka Bay clay with slightly lower values of pH (about 3) at higher acidic amounts. In the case of the titration curve of the Ariake clay, the suspension pH drops from about pH 6 to 3.3 when acid input exceeds 50 cmol $\text{H}_2\text{SO}_4 \text{ kg}^{-1}$ soil. Taken as a whole, the titration curves show that the Kawasaki and Osaka Bay clays have a slightly higher resistance to pH change than the Ariake clay. High resistance of the soils to pH changes is likely due to their mineral composition, which includes smectite, a mineral with a high cation exchange capacity (C.E.C.). In the case of the Osaka Bay clay, the carbonates neutralize H^+ , a process that also contributed to the high buffer capacity of the soil.

In summary, the results indicate that the studied soils have high buffer capacities and thus relatively high amounts of acid would be needed to substantially decrease the pH of pore fluids in these clays.

Test Results with Osaka Bay Clay

Compression tests with different pH values of the pore fluid were performed on eight specimens of

the Osaka Bay clay (Fig. 4). Two clusters of the compression curves can be recognized: the first cluster consists of four curves with pH values in the range of 6.65 to 7.09, including the specimen with distilled water (pH=7.09). The second cluster (solid symbols in Fig. 4) includes pH values in the range of 5.78 to 6.07. It is noted that the specimens with lower pH values underwent acid contamination for a longer period of time.

Laboratory data indicate that the duration of acidic leaching has a strong influence on the compressibility of the Osaka Bay clay. It is evident from Fig. 4 that the specimens in the second cluster had greater values of initial void ratio, which may be the result of the longer exposure to the acidic contamination. In addition, Fig. 5 shows that the compression index (C_c) can significantly increase as the environment becomes more acidic. It is clear from this figure that the compression indices of clay specimens with pH values in the range of 6.65 to 7.09 only slightly differ from that of distilled water ($C_c=0.474$) while at the lower pH values the compression index of the same clay greatly increased to 0.722 ~ 0.756.

Test results with Ariake Bay Clay

Due to the limited amount of the Ariake Bay clay available for this research, only four tests with different pH values were carried out. As can be seen in Fig. 6, the virgin compression curves for all four soil specimens were placed in the order of pH values, with the uppermost curve having pH=5.71 and the lowest two having pH in the range of 7.04 to 7.58. For reference, the specimen in distilled water has a pH value of 7.58. The test data show that at a given overburden stress, the void ratio decreased with an increase in pH. These results are in agreement with those obtained for the Osaka Bay clay. Another similarity is that the compression index of the Ariake clay also increases with a decrease in the pH (Fig. 7).

Test results with Kawasaki Mud

The results of nine compression tests with the Kawasaki mud demonstrate the opposite tendency in comparison with the test data of the Osaka (Fig. 5) and Ariake (Fig. 7) clays. As can be seen in Fig. 8, the uppermost virgin compression curve has the highest pH value of 8.39, while the lowest two values are 6.01 and 6.12. In addition, unlike the Osaka and Ariake clays, the compression index of the Kawasaki mud clearly decreases with a decrease in the pH (Fig. 9). This observation is in qualitative agreement with the double-layer theory (Olphen 1991). According to the theory, the thickness of the double layers decreases with an increase in concentration of ions (in this case H^+ ions), leading to a

decrease in void ratio. It is also noted that similar behavior was reported by Meegoda and Ratnaweera (1994), Sridharan et al. (1986), Gajo and Maines (2007) for artificial mixtures and remolded clays.

DISCUSSION

It has long been recognized that compressibility of clay is governed by mechanical and physicochemical factors. However, the results obtained in this study indicate that the physicochemical factors may have more pronounced impact on the consolidation properties of clays when pH values of the pore fluid become low. As the specimens of each tested soil are assumed to have had the same microfabric prior to acidic leaching, it can be hypothesized that the observed changes in compressibility were mainly due to the soil-water-acid interaction.

It is interesting to note that acidic contamination can have opposite effects on the compressibility of marine soils. For example, in case of the Osaka (Fig. 5) and Ariake Bay clays (Fig. 7), it was found that a decrease in pH correlated with increases in compression indices, while for the Kawasaki mud (Fig. 9), the effect was the opposite. Analysis of the available literature suggests that the observed difference may be attributed to two main factors, that is, clay mineralogy and soil structure. Unlike the Kawasaki mud, both the Osaka and Ariake clays have developed chemical bonding, including cementation, over a long geological history. In the case of the Osaka Bay clay, the bonding agent is assumed to be calcium carbonate (Imai et al. 2006), while for the Ariake clay, it is iron oxide (Torrance and Ohtsubo 1995). It can be hypothesized that during leaching acidic water would dissolve the cementation, forming looser structures with larger voids, thus resulting in the greater compressibility of the clays. In comparison, for the non-consolidated deposit of Kawasaki mud, the effect of structural bonds is expected to be less pronounced, and its behavior would be determined by the mineral composition and diffuse double layer (Mitchell 1993). According to Gajo and Maines (2007), the compressibility of soils with smectite/montmorillonite which are consolidated from slurry would decrease in an acidic environment presumably due to the collapse of the diffuse double layer.

It is interesting to note that although all three tested soils contain smectite/montmorillonite, the expected decrease in compressibility due to the collapse of the diffuse double layer was observed only for the Kawasaki mud. It is believed that, in addition to earlier discussed changes in the soil structure of the Ariake and Osaka clays, the kaolinite mineral, which is only found in these two clays, might also contribute to the alteration of clay microfabric. As shown by Wang and Siu (2006a,b), in an acidic medium, kaolinite tends to form more open flocculated arrangements that would determine greater compressibility of soil.

CONCLUSIONS

In this work, three marine deposits, namely Osaka Bay clay, Ariake Bay clay, and Kawasaki mud, were subjected to long-term acidic leaching in order to study the influence of contamination on the compressibility of natural soils. Based on the obtained results, the following conclusions can be drawn:

- The marine soils used in this research have high buffer capacities, a property that is attributed to their mineral compositions;
- Long-term clay-water-acid interaction can lead to considerable changes in compressibility of the studied soils;
- Clay mineralogy, soil structure, including cementation, and the duration of acidic contamination appear to be the key factors determining the degree to which changes in the clay's compressibility can occur during leaching;
- For the Osaka and Ariake clays, long-term acidic contamination resulted in higher compressibility, likely due to changes in soil's structure.
- For the Kawasaki mud, the opposite effect was observed, that is, the compressibility of soil decreased in an acidic medium as a result of a decrease in the diffuse double layer thickness.

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FIGURE CAPTIONS

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

Figure 2. Changes in the liquid's pH recorded over a 300-day period for the Osaka Bay clay. As a part of leaching procedure, the liquid was replaced with the original solution of sulfuric acid (pH=3.0) every 7-8 days.

Figure 3. pH-titration curves of clays studied.

Figure 4. Influence of pH on void ratio-overburden stress relationship obtained for the Osaka Bay clay.

Figure 5. Influence of pH on Compression Index of the Osaka Bay clay.

Figure 6. Influence of pH on void ratio-overburden stress relationship obtained for the Ariake Bay clay.

Figure 7. Influence of pH on Compression Index of the Ariake Bay clay.

Figure 8. Influence of pH on void ratio-overburden stress relationship obtained for the Kawasaki mud.

Figure 9. Influence of pH on Compression Index of the Kawasaki mud.