Effects of Grain-Size Distribution, Iron Oxide, and Organic Matter on Sedimentation and Self-Weight Consolidation of Thoroughly Disturbed Soft Marine Clay

KONDO Fumiyoshi* and TORRANCE J. Kenneth**

*Faculty of Agriculture, Saga University, 1 Honjo-machi, Saga 840-8502, JAPAN.
**Department of Geography and Environmental Studies, Carleton University, 1125 Colonel By Drive, Ottawa, ON K1S 5B6, CANADA.

Abstract

The sedimentation and self-weight consolidation processes of thoroughly disturbed soft marine clay are relevant both to land reclamation work and to sediment accumulation on tidal flats of coastal lowland plains. The influences of grain-size distribution, iron oxide content and organic matter content on these processes and the final water contents of a low-activity mud were investigated at water contents from 500 to 6,000% over salinities ranging from 30 to 0.5g/L. Particle size distribution had a large effect; specifically, the lower the clay content the more rapid the differentiation of an upper sediment boundary and the self-weight consolidation of the accumulated sediment. Iron oxide addition had little effect on these characteristics for this low activity clay. Peroxide destruction of the original organic matter suggested that it had a mildly flocculating effect; the addition of 5% organic matter, in the form of fulvic and humic acids, had a dramatic dispersing effect and inhibited sediment accumulation and consolidation for all conditions tested. The experiments indicate that use of dredged mud with low organic matter content, lesser clay contents and lesser swelling clay content, applied at the lowest water content that is feasible, will provide for the most rapid sedimentation and self-weight consolidation in reclamation projects.

Key words: Grain-size distribution, Iron oxide, Organic matter, Sedimentation mode, Final average water content, Land reclamation

1. INTRODUCTION

The dewatering behavior of disturbed soft clay is among the most important factors for planning of land reclamation projects using dredged mud. The literature on dredging indicates that dewatering, with consequent volume decrease, occurs by two processes: first by sedimentation during which the particles sediment individually, or as floccules, through the water phase; and second by self-weight consolidation during which the weight of the accumulated sediment forces water from the accumulated material. Theoretical investigation of the sedimentation and self-weight consolidation behavior has been conducted (Kynch, 1952; McRoberts and Nixon, 1976; Mikasa, 1963; Mikasa and Takada, 1984), but influences of the properties of the dredged material on the processes have not yet been investigated. This work addresses these influences.

The sedimentation and self-weight consolidation processes are relevant not only to land reclamation work but also to sediment accumulation on tidal flats of coastal lowland plains, such as the Ariake Bay area, western Kyushu, Japan. Extensive areas of the clay-rich sediments of these wide tidal flats surrounding Ariake Bay, subsequently called Ariake clays, have been developed as reclaimed paddy farming land, with or without the addition of dredged materials. Integrated information on the geology, mineralogy and geochemistry of Ariake clays, and their correlation with geotechnical behavior, was reported by Ohtsubo et al. (1995). The Ariake clays of the tidal flats, that accumulated during and after post-glacial sea-level rise, are dominated by a low- to non-swelling smectite and can develop the geotechnical behavior known as quick clay. In this context, they are similar to the non-smectitic, sensitive marine deposits (commonly known as Leda clay) in eastern Canada (Ohtsubo et al., 1982; Torrance, 1984; Torrance and Ohtsubo, 1995).

Recent laboratory sedimentation and self-weight consolidation experiments with Leda clay from eastern Ontario, Canada demonstrated that its settling pattern was influenced by its physical, chemical and mineralogical properties (Kondo and Torrance, 2003). The sedimentation/self-weight consolidation modes and
final water contents of Leda clay are also strongly affected by smectite, salinity and water content (Kondo and Torrance, 2005). The current paper reports the influence of iron oxide content, organic matter content and grain-size manipulation on the sedimentation/self-weight consolidation modes and final water contents of the same Leda clay.

2. MATERIALS AND METHODS

2.1 Stock Material

The Leda clay material used was collected from the site of the Lemieux landslide of 1993 on the South Nation River in eastern Ontario, Canada (Evans and Brooks, 1994). An unoxidized sample was collected from about 1 m below the recently erosion-exposed surface near the base of an intact block that had been displaced by the landslide. The material is estimated to have been approximately 20 m below the original land surface. Its in-situ properties were: water content = 42%; liquid limit = 33%; plastic limit = 20%; sand (>75 μm) = 0.2%, silt (2-75 μm) = 41%, clay (<2 μm) = 59%. The mineralogy was dominated by quartz, feldspars, illite, chlorite and amphibole, as is typical of the Leda clays. It contained about 0.5% organic matter.

This stock material was modified by addition or removal of components, as described below, in order to test the nature of the influence of various material factors on the sedimentation and self-weight consolidation process. In order to have a consistent chemistry, all samples were Na-saturated. Na-saturation was achieved for each experimental sequence by washing the appropriate material once with 1 M (58.5 g/L) NaCl and then twice more with 30 g/L NaCl (as salinity relative to average sea water level). The Na-saturated Leda clay serves as the control material. The results of sedimentation experiments for the control material have been reported (Kondo and Torrance, 2005).

An experimental sequence was also conducted in which the Na-saturation step was omitted and the untreated, natural Leda clay material was introduced directly into a 30 g/L NaCl sedimentary environment for the first step of the experimental sequence. This we call the natural material.

2.2 Grain-size Manipulation, Iron Oxide Addition and Organic Matter Treatment

The natural material contained 59% clay. Materials with lesser and greater clay (<2 μm) contents were produced by the following procedures. A large amount of natural material was mechanically dispersed and allowed to settle in sedimentation cylinders for 7 hours, after which the suspended material remaining in the top 10 cm was decanted. The dispersion, sedimentation and decantation procedure was repeated 4 times to produce material with 38% clay. Material with 79% clay was prepared by adding the clay-sized particles removed in the above step to an appropriate amount of the stock material. In these clay-depleted and clay-enriched materials, the mineral suite present is unchanged, but the overall abundance of primary minerals is increased in the clay-depleted (silt-enriched) material and the overall abundance of phyllosilicates increased in the clay-enriched material (Torrance, 1985; Kondo and Torrance, 2003). Na-saturation was carried out on each material.

Iron oxide addition experiments were conducted by adding goethite to the natural material to produce the materials with 5% and 10% iron oxide. This goethite was well-aged, having been prepared in 1986 by bringing a 0.17 M FeCl₃ solution to pH 12 by addition of 2 M NaOH (Torrance, 1990). After goethite addition, the sample was washed with 30 g/L NaCl solution 5 to 6 times to decrease the pH value and to attain Na-saturation. This addition of crystalline iron oxide was intended to mimic the situation of crystalline iron oxides co-sedimenting with the other minerals during sediment accumulation; this experiment does not mimic the situation of large quantities of iron oxides present as surface films on other mineral particles, as is common in natural soils.

Organic-free soil was prepared by hydrogen peroxide (H₂O₂) treatment. 310 mL of 30% H₂O₂ and 2 L of deionized water was added to 2,500 g of the natural material to attain a suspension with approximately 3% H₂O₂. The bubbling contents were stirred occasionally to prevent overflowing of soil from the container. After frothing ceased, another 200 mL of 30% H₂O₂ was added and stirred occasionally until frothing again ceased. Organic-enriched soil (5% organic matter) was produced by adding soil organic matter to the natural material. This organic matter was extracted from peat soil by centrifugal washing using 0.1 N NaOH as extractant. This procedure extracts both the acid- and base-soluble fulvic acids and the base-soluble humic acids; these are the most chemically active portions of the organic fraction of soils and sediments (Schnitzer, 1982). In order to encourage association of the organic matter with the mineral soil material, the pH value after addition was adjusted to 4 using 1 N HCl. Two washings with 30 g/L NaCl were conducted, followed by neutralization of the pH using 0.5 M NaOH, and two more washings with the NaCl to establish Na-saturation and near-neutral pH.

2.3 Sedimentation Procedure

The sedimentation experiments were conducted using 6-cm diameter, 26-cm high acrylic cylinders that were filled to 20 cm with the appropriate suspensions. Test
series were conducted at water contents ranging from 500 to 6,000%, with each series being conducted over salinities (NaCl) ranging from 30 or 15g/L to 0.5g/L (30, 15, 8, 4, 2, 1 and 0.5g/L). The 30g/L experiment was not done with the materials of grain-size manipulation, iron oxide addition and peroxide treatment. After thorough mixing with a plunger, the sedimentation and self-weight consolidation processes at each salinity were monitored for 14 days. To decrease the salinity to the next lower value in each constant-water-content sequence, an appropriate amount of clear supernatant solution was removed by siphon or centrifuge (when necessary) after each test was completed. Deionized water was then added to re-establish the original water content and obtain the next desired salinity. The thorough mixing and monitoring was repeated. For all experiments, the salt concentrations were monitored by measuring the electrical conductivity of the suspensions. The initial water contents in all cases are the values corrected for salinity (Imai et al., 1979). All experiments were conducted in the laboratory with a constant temperature of 21°C.

The settling patterns of the clay suspensions were classified into four types: dispersed free settling (DFS – the particles settle independently, according to size), flocculated free settling (FFS – the particles flocculate and the flocules settle independently, according to size), zone settling (ZS – the particles flocculate, and the flocules all settle at the same rate) and consolidation settling (CS – the particles flocculate as a single mass that occupies the whole column and this mass experiences self-weight consolidation) (Imai, 1980). The classification of settling types was done on the basis of naked-eye observations. In all modes of settling, self-weight consolidation commences as soon as a sedimentary pack commences to accumulate.

3. RESULTS AND DISCUSSION

The experimental results are presented as a series of figures (Figs.1-8) that include graphs of “sediment thickness” versus time for a series of salinities (15, 8, 2 and 0.5g/L), with the settling pattern exhibited by the individual treatments (pretreatment, salinity and water content) being indicated at the right side of each graph. The “sediment thickness” recorded in these figures represents the interface between the zone of water that was free or almost free of particulate material and the zone of high particulate concentration. The first point plotted for each water content represents the earliest time that a sediment interface between clear (or almost clear) water and settling flocs or accumulated sediment could be identified. For CS, ZS and FFS behaviors, it always represents an upper boundary of settling or self-weight consolidating flocs; for DFS behavior, it represents the upper level of particles that have accumulated on the bottom of the cylinder.

The relationships between maximum settling rate and salt concentration are also presented in Fig.9. The maximum settling rate was calculated by least squares method from the maximum gradient of the time-sediment thickness curve. Then the interpretation
and comparison of the results of the different treatments are facilitated.

3.1 Sedimentation Modes of Na-saturated and Natural Leda Clay Material

The time sediment thickness curves for the Na-saturated and natural materials at selected salt concentrations are presented in Fig.1 and Fig.2. The relationships between maximum settling rate and salt concentration for these materials are also presented in Fig.9(a) and Fig.9(b). The Na-saturated results represent the control response to which the other treatments are compared. The natural material response differs somewhat from the Na-saturated control material in that the ZS-FFS and CS-ZS boundaries are both shifted to higher water contents, and CS behavior continues to lower salinity (Kondo and Torrance, 2003; 2005). The difference in the behavioral boundaries is explainable by an increased flocculating power of the sedimentation environment associated with the displacement into solution of adsorbed Ca, Mg and K cations from the natural material by cation exchange with Na from the NaCl sedimentation environment. A second difference is that, while the time-sediment thickness curves were never completely identical for any of the sedimentary conditions, the curves for the natural material (relative to the Na-saturated material) tended to be shifted upward (time delay) at lower water contents and downward (earlier) at higher water contents, with the boundary between the upward and downward shifting tendency being at approximately 2,500% water at 15g/L, 1,500% water at 8g/L, 750% water at 2g/L, and not present at 0.5g/L. The upward shift is believed to be related to the increased flocculating power generating fluffer, lower density flocs that will settle more slowly in ZS mode, and consolidate more slowly in CS mode. The downward shift at higher water contents probably also relates to slightly larger flocs, but in this case settling is more rapid because, unlike at lower water contents, the flocs are sufficiently separated that they do not significantly interfere with each other’s settlement.

3.2 Sedimentation Modes of Grain-size Manipulation Experiments

Time-sediment thickness curves for the clay-depleted (38%), clay-augmented (79%) and the control material are presented in Figs.3, 4 and Fig.1 respectively. As was anticipated, the particle size distribution has a large effect on the settlement-time response and maximum settling rate (Fig.9(c) and Fig.9(d)) of thoroughly disturbed sediments at various salinities and water contents.

The effect of clay depletion (from 59 to 38%) was to shift the time-sediment thickness curves toward more rapid differentiation of an upper sediment boundary and for the sediment below the differentiated boundary to be more compact at comparable times. The CS behavior, which was exhibited at higher salinities in the control material, was not present; slight shifts in ZS/FFS boundaries occurred at the higher salinities.
Augmentation of the clay content (from 59 to 79%) had the effects of shifting sediment mode boundaries and the time-sediment thickness curves. The latter shifted toward: slower differentiation of the upper sediment boundary at the 15 and 8g/L salinities for all water contents; more rapid differentiation of the upper sediment boundary at 2g/L salinity for high water contents; and faster differentiation of the sediment boundary at all water contents at 0.5g/L. The shapes of the time-sediment thickness curves at 79% clay suggest that slower differentiation of the upper boundary at the higher salinities is caused by quick development of full volume gelation (the start of CS mode sedimentation) at water contents above 2,500% at 15g/L and above 1500

**Fig.3** Time-sediment thickness curves for material of 38% clay content (Initial water contents: ● = 500%, ○ = 750%, ○ = 1,000%, ▲ = 1,500%, △ = 2,000%, ▽ = 2,500%, ▼ = 3,000%, □ = 4,000%, ◆ = 6,000%)

**Fig.4** Time-sediment thickness curves for material of 79% clay content (Initial water contents: ● = 500%, ○ = 750%, ○ = 1,000%, ▲ = 1,500%, △ = 2,000%, ▽ = 2,500%, ▼ = 3,000%, □ = 4,000%, ◆ = 6,000%)
% at 8g/L, with the CS mode breaking down to ZS mode after some time for some of the water contents. While such behavior is most common in the clay-augmented experiments, it is not unique to this material; the characteristic of having a thickness of approximately 20cm for 100 to 500 minutes and then experiencing a “catastrophic” decrease in thickness is exhibited under one or more conditions with all the other materials.

3.3 Sedimentation Modes of Iron Oxide Addition Experiments
Leda clay from the South Nation River area of eastern Ontario has a citrate-dithionite extractable iron

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**Fig. 5** Time-sediment thickness curves for 5% iron oxide added material (Initial water contents: ● = 500%, ○ = 750%, ○ = 1,000%, ▲ = 1,500%, △ = 2,000%, ▼ = 2,500%, ▽ = 3,000%, □ = 4,000%, ○ = 6,000%)

**Fig. 6** Time-sediment thickness curves for 10% iron oxide added material (Initial water contents: ● = 500%, ○ = 750%, ○ = 1,000%, ▲ = 1,500%, △ = 2,000%, ▼ = 2,500%, ▽ = 3,000%, □ = 4,000%, ○ = 6,000%)
oxide content of approximately 0.6% on a dry-weight basis (Seekings, 1974), most of which is present as very fine particles of hematite (Torrance et al., 1986). The amounts of goethite added for these experiments greatly augments the iron oxide content, although the added oxide probably does not become as intimately associated with the other mineral particles as that originally present.

The time-sediment thickness curves for the materials with 5% and 10% iron oxide added are presented in Fig.5 and Fig.6, respectively. Compared to the Na-saturated material, there are only very minor differences in the ZS/FFS boundaries at salinities above 8g/L in the sedimentation modes for both iron-oxide-enriched samples. The time-sediment thickness patterns and maximum settling rates (Fig.9c and Fig.9f)) are also relatively unaffected. These results indicate that even 10% co-sedimented particulate iron oxides have little effect on the sedimentation characteristics of low activity clays.

The reader is cautioned that iron oxides formed in soil under oxidizing conditions commonly become closely associated with and form “films” that partially cover the particle surface. In rheological experiments involving manipulation of iron oxide contents, Torrance (1990) found that it required about 2% of added particulate iron oxide to restore the yield stress/water content behavior of a South Nation Leda clay from which iron oxide had been removed by citrate-dithionite extraction to the behavior exhibited by the untreated material with its 0.6% iron oxide present as discontinuous surface films. The addition of 8% particulate iron oxide greatly affected the rheological behavior. The differences in the effects of iron oxide addition for sedimentation behavior and rheological behavior is probably related to the water contents, which were much lower (40 to 200%) for the rheological experiments. Our belief is that 5 to 10% of iron oxides, if present as surface films on clay mineral particles, would have substantially greater effects on sedimentation and settlement behavior than observed in these experiments.

3. 4 Sedimentation Modes of Organic Matter

Treatment Experiments

Time-sediment thickness curves for materials from which the organic matter was removed using hydrogen peroxide, and for materials to which 5% organic matter (as fulvic and humic acids extracted from a peat soil) was added are presented in Fig.7 and Fig.8, respectively.

In the case of organic matter removal by hydrogen peroxide (Fig.7 and Fig.9g), sedimentation modes (relative to the control material in Fig.1 and Fig.9(a)) were unchanged at 15 and 8g/L salinity, and only very minor changes in the position of individual sedimentation curves occurred. At 2g/L, sedimentation of the 500% water content suspension was delayed, but the development of an upper boundary for the sedimenting material was advanced for water contents above 1500%. The greatest effect is observed at 0.5g/L salinity where the development of the upper boundary for the sedimenting material is advanced for all experimental conditions, and ZS behavior still occurred at the lowest water content. At low salinity, with the exception of the lower water content experiments at 2g/L, the effect of oxidation was to enhance the rate of settlement of particles from suspension. This might be interpreted as indicating that the organic matter originally in the soil was having a mildly dispersing effect, but a complicating factor is present. The complication is the possibility that the oxidizing agent affected the minerals present, through the oxidation of some structural iron, and thereby lowered the cation exchange capacity (as happens with the low-activity, high-iron smectites of Ariake Bay, Japan (Torrance, 1999)). The effect of oxidation on the smectitic Ariake Bay sediment is to enhance swelling (and dispersion) at low salinities; in the non-swelling experimental materials, the effect of oxidation of structural iron (if such occurs) would be to decrease dispersing forces and enhance settlement at low salinities. Despite this possible complication, it seems most probable that, when the salinity was sufficiently low as to not induce relatively quick flocculation, the organic matter originally present had a mildly dispersing effect that retarded both flocculation and the sedimentation process.

The addition of 5% organic matter by weight, in the form of fulvic and humic acids extracted from a muck soil, has dramatic effects of the sedimentation behavior for all combinations of suspension water content and water salinity (Fig.8 and Fig.9(h)). At 15g/L, the boundaries for sedimentation mode are shifted upward and the development of an upper boundary for the accumulating sediment is delayed. The effects become extreme for 8g/L salinity and by 0.5g/L no sedimentation data was obtainable because it was not possible using the experimental protocol to prepare any samples with the desired starting conditions. Three starting conditions could be met for the 2g/L material. The dispersing effect of the adding 5% organic matter, as fulvic and humic acids, was sufficiently strong to seriously disrupt the flocculating ability of salinities as high as 15g/L. The observed dispersing effects were stronger than anticipated, and are suggestive that even modest amounts of soluble organic matter can have major consequences for geotechnical behavior.

3. 5 Final Average Water Contents

The final average water contents of the accumulated sediment are important factors to be considered in the
planning of reclamation and the use of reclaimed land. **Fig.10** show the relationships between final average water content and salt concentration of the sediments. The water contents were calculated from the known weight of the suspended material and the final sediment thickness in each cylinder.

For the Na-saturated control material (**Fig.10(a)**) and the natural material (**Fig.10(b)**), the least compact sediments (the highest water contents) at the end of two weeks, are exhibited by sediments accumulated at high salinity from high initial water content suspensions. The sediment water contents generally decreased along both the initial water content and the salinity axes. The change along the salinity axis was dramatic for high

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**Fig.7** Time-sediment thickness curves for H₂O₃ treated material (Initial water contents: ⬜= 500%, ○= 750%, ○=1,000%, ▲=1,500%, △=2,000%, ▼=2,500%, ▽=3,000%, □=4,000%, △=6,000%)

**Fig.8** Time-sediment thickness curves for 5% organic matter added material (Initial water contents: ⬜= 500%, ○= 750%, ○=1,000%, ▲=1,500%, △=2,000%, ▼=2,500%, ▽=3,000%, □=4,000%, △=6,000%)
initial water contents but was relatively small for the lower initial water content experiments. There was also little effect of initial water content differences at low salinities. The natural material response differs from the Na-saturated material in that the final water contents are relatively higher along both the initial water content and the salinity axes. The differences between these two cases are explainable in terms of increased tendency to flocculation in the natural material. For both materials, it can be stated the primary condition to satisfy in order to rapidly accumulate 'low' water content sediment is to begin with a low water content suspension.

Relative to the Na-saturated material, the clay-depleted material (Fig.9(c)) developed a much lower water content sediment under all experimental conditions. The patterns of change with different initial water contents and salinity are similar, but of lesser magnitude that for the natural sample. In practical terms, the effects of salinity on final water content are probably of little importance if the initial water content of the suspension is low. At high initial water contents, the lower salinity sedimentation environments yielded sediments with substantially lower water contents.

The clay-enriched material (Fig.9(d)), follows a substantially different pattern with sediment water contents being particularly high for high salinity, high initial water content materials and for low salinity, low initial water content materials. The most compact sediments occur for low salinity, high initial water content materials, but, in practical terms, the high salinity, low initial water content materials would yield most rapid sediment accumulation. Over the clay content sequence of experiments, it becomes clear (as expected) that increasing clay contents lead to higher
volume sediments regardless of sedimentation conditions.

Relative to the Na-saturated material, enhanced iron oxide contents (Fig.10(e) and Fig.10(f)) change the patterns only slightly over most of the range of conditions. The main effect is to cause a substantial increase in the sediment volume for the low salinity (1g/L), low initial water content conditions. This effect increased as the amount of iron oxide increased from 5 to 10%.

The peroxide treatment (Fig.10(g)), that removed the approximately 1% organic matter naturally present in the original material, did not substantially change the pattern of the results, but decreased the final water content of the sediments over the complete range of experimental conditions. Most of this effect is attributed to the organic matter removal because it is unlikely that the peroxide treatment had significant effects on the mineralogy in the non-swelling soil materials used in these experiments.

The addition of 5% organic matter (Fig.10(h)), in the form of humic and fulvic acids, had dramatic effects. Particle settlement was greatly inhibited at low salinities for all initial water contents. Where data is lacking in Fig.10(h), it is because the dispersing effects of the added organic matter precluded the preparation, using the experimental protocol, of the sample for the desired experimental conditions.

While sediment accumulates under the higher salinity conditions, the dispersing actions of the humic and fulvic acids are very strong and present problems for the accumulation of low water content sediments. The action in this case is dominantly a chemical effect; less soluble organic materials (larger molecules or fragments) would also be expected to be
problematic but more probably because of physical effects they would have on the system.

4. CONCLUSIONS

The experimental results demonstrate that grain size distribution, iron oxide content and organic matter content all influence the sedimentation and self-weight consolidation modes and final water contents of Leda clay suspensions. The greatest change in behavior was caused by a 5% by weight augmentation of the organic matter content from its original value of 0.5%. The dispersing activity of the added organic matter acted to inhibit the processes of flocculation, sediment accumulation and self-weight consolidation over the full range of experimental conditions. Differences in grain size distribution were also significant with the rates of sediment accumulation and self-weight consolidation decreasing substantially as the clay content increased from 38 to 79%. The presence of differing amounts of iron oxide (up to 10% goethite), as discrete particles, had little influence on any of the processes.

The results indicate that the most desirable mud to dredge for rapid accumulation of sediment with low water content in land reclamation projects is one with low organic matter content and lesser clay content, and with less swelling clay present. The general condition for most rapid accumulation and self-weight consolidation is to start with as low water content is possible and with his sedimentation environment that is saline to brackish; a low salinity sedimentation environment generally leads to slower accumulation and slower consolidation.

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