PORE SIZE DISTRIBUTION AND HYDRAULIC CONDUCTIVITY
CHARACTERISTICS OF MARINE CLAYS
Hiroyuki Tanaka, Port and Airport Research Institute, Yokosuka, Japan

ABSTRACT
Using intact clayey soil samples, which were recovered from various areas and depths, Pore Size Distribution (PSD) was measured by Mercury Intrusion Porosimetry (MIP). In addition, natural deposit samples, the artificial mixture soils, namely Singapore clay with Toyoura sand or diatomite, are studied in order to know the influence of grain size component on PSD. It was found that characteristics of the PSD are represented by the mean of pore size ($D_{50}$). As an application of PSD characteristics to practical geotechnical engineering, hydraulic conductivity ($k$) is attempted to be related with the PSD.

RÉSUMÉ
Des échantillons intacts d’argile ont été obtenus de différentes régions et profondeurs. Sur ces échantillons, on a mesure la courbe granulométrique (PSD) ainsi que la distribution des pores à l’aide d’un porosimètre au mercure (MIP). En plus des échantillons naturels, des mélanges artificiels ont été réalisés avec des sols de Singapour, du sable de Tyoura ou de la diatomite. Il a été trouvé que les caractéristiques de distribution des pores était possible en utilisant la distribution moyenne des pores ($D_{50}$) obtenue par le PSD. Une application à la géotechnique, reliant les résultats obtenus avec le PSD et la conductivité hydraulique est proposée.

1. INTRODUCTION

The hydraulic conductivity ($k$), especially for clayey soils, has been indirectly used as consolidation coefficient ($c_v$) for prediction of the rate of settlement. However, recently with increasing concern of environmental problems, the value of $k$ itself has become a very important parameter in seepage problems of polluted fluid.

It is well established from previous studies that the $k$ value decreases with a decrease in void ratio ($e$) and there is a unique relation between $e$ and $k$ of a soil, regardless of its stress history. However, if the relation of $e$ and $k$ is compared with various soils, it is easily understood that $k$ is not governed by only $e$. For example, though $e$ for sandy soil is in general smaller than that for clayey soil, $k$ value for sandy soil is definitely larger than that for clayey soil. It is anticipated that the $k$ value cannot be not correlated with only $e$, but also governed by additional factors, especially the size of voids. In this paper, Pore Size Distribution (PSD) was measured using Mercury Intrusion Porosimetry (MIP) to find a correlation between $k$ value and PSD.

Soil samples used in this study were natural clays recovered from various areas in the world. The studied soil samples are not only Holocene deposited, but also Pleistocene clays retrieved from depths as deep as about 200 m. In addition to these natural deposited soil samples, artificially mixed soils with sand or diatomite soil are used to examine the influences of grain size components on the PSD as well as the $k$ value.

2. SOILS USED IN THE STUDY AND TESTING METHOD

2.1 Naturally deposited soils

Geotechnical index properties of soils used in this study are given in Table 1. Ma 13, Ma 11, Ma 9 and Kyoto clay belong to the Osaka Group, which are numbered according to deposited are. For example, Ma 13 is the youngest Holocene clay deposit and found at the sea bottom. Other numbered clays including Kyoto clay are Pleistocene clays. All intact samples, except for Pleistocene clays, were recovered by the fixed piston sampler specified by the Japanese Geotechnical Society (JGS). Since the Pleistocene clays were very stiff, the rotary core sampler was used to retrieve them. London clay is well-known clay in geotechnical engineering literatures and was taken from the Heathrow airport, London, UK. Only this clay is Tertiary deposit.

2.2 Artificially mixed soils

To study influence of grain size component, artificially mixed soils with Toyoura sand and diatomite were prepared. As a base material, Singapore clay was used. Their main properties including the grain size distribution are indicated in Table 2 and Fig. 1. The Toyoura sand is a well-documented material in Japan because researchers of the JGS have extensively used it in their study as a standard granular material. Its grain size distribution is shown in Fig. 1. While the diatomite soil used in this study was recovered from the Hiruzenbara site, Okayama prefecture, Japan (Shiwakoti et al., 2002). Although the particle size of the diatomite soil is mainly silt, as shown in Fig. 1, its void ratio or water content is very large because of large pores in diatom particles, which can hold a large volume of water. As a result, if the diatomite soil is mixed with usual soil, its index properties such as liquid limit ($w_l$) or plastic limit ($w_p$) increase in spite of decrease in clay particle content. This
tendency is completely opposite to the soil mixed with the Toyoura sand. Singapore clay was mixed with the Toyoura sand or the diatomite soil, using a mixer at high water content. The reason for selection of Singapore clay as a base material is that it does not contain diatom unlike Japanese clays. The mixed soils were prepared at several ratios: namely, 0, 25 %, 50 % and 75 % by dry weight of the Toyoura sand or the diatomite soil to the Singapore clay. Hereafter, the mixed soils are named such as S+Sa(50). The first letter of S indicates the name of Singapore clay. The later letter of Sa and D means the mixing material of the Toyoura sand and the diatomite soil, respectively. The number in the blanket indicates the mixing ratio as explained above.

The mixed soil was poured into a cylindrical consolidation cell having the diameter of 10 cm. The final consolidation pressure was 200 kPa and the completion of consolidation was confirmed by the so-called 3t method: i.e., the duration for the consolidation is three time longer than the time for getting the primary consolidation defined by the bending point in the settlement curve plotted against time in the logarithm scale. Then the consolidation pressure was removed, and the reconstituted sample was extruded from the cell.

2.3 Mercury intrusion porosimetry
For the Mercury Intrusion Prosimetry (MIP), every void in the specimen should be replaced by air: i.e., the specimen needs to be dried. In this study, the dry freezing method was employed to avoid changes in the pore size distribution in the process of drying due to the volume change. The specimen was cut into a cubic about 5 mm, which was then submerged into the liquid nitrogen to freeze it. To dry the frozen specimen, it was set in the special chamber under the vacuum (less than –90 kPa) and low temperature (-10 °C). In this process, water in the voids was sublimated without any volume change of the specimen.

The dried specimen was then mounted in the apparatus of the MIP. Mercury was intruded into the voids by applying pressures. The diameter of voids is calculated according to the capillary force theory. The maximum pressure of mercury intrusion is 225 MPa, which corresponds to the pore size diameter of about 0.005 µm. It is usual that the void ratio of the specimen is calculated from the water unit weight of the soil (γw) and unit solid weight of the soil particle (ρs). The calculated e in this manner should agree with the total sum of pores measured by the MIP. However, if pores in the specimen get shrunken in the process of the specimen preparation or if cracks are developed in the specimen, then the total pores measured by the MIP are not identical to the e obtained by the above conventional calculation method. Therefore, if the discrepancy between these two values of e is found to be more than 10 %, then such test data were not used in this study.

It should be kept in mind that the diameter of pores measured by the MIP is somewhat different from our
impression brought by the SEM observation. In addition to the assumption of the pore shape as cylinder, the pore diameter obtained by the MIP corresponds to that at the entrance of pores, but not the inside diameter of the pores, as discussed by Lapierre, et al. (1990).

3. PORE SIZE OF INTACT NATURAL SOILS

3.1 Pore size distribution (PSD)

Figure 2 shows typical examples of Pore Size Distribution (PSD) for intact natural soils. The vertical axis in the figure is plotted as \( \frac{dV}{d\log D_p} \), where \( D_p \) is the entrance pore size diameter and \( dV \) is the volume of pores whose diameter is \( D_p \), expressed in terms of the unit dry weight of soil. Therefore, the area bounded by certain \( D_p \)s indicates the volume of pores between these \( D_p \)s, \( dV \). If the volume of pores with \( D_p \) less than 0.005 \( \mu m \) is ignored, then the total area bounded by the measured curve by the MIP corresponds to the void ratio, \( e \).

Figure 2 is somewhat inconvenient to characterize the PSD, and it is necessary to show the PSD in terms of a more representative parameter. Figure 3 shows the relation between \( D_p \) and the cumulative volume normalized by the dry soil weight, where the cumulative pore at the infinitive large \( D_p \) corresponds to the total void volume, i.e., \( e \). In this figure, three pore diameters are defined as \( D_{p30} \), \( D_{p50} \) and \( D_{p60} \). For example, the \( D_{p30} \) is the diameter where 30% of the total cumulative pore is attained, as shown in the figure.

Figure 4 shows how the ratio of \( D_{p30}/D_{p50} \) or \( D_{p60}/D_{p50} \) varies with \( D_{p50} \). It can be seen that these ratios are scattered in some narrow ranges and do not change with \( D_{p50} \). This means that the pattern of the PSD curve is nearly the same and the PSD can be represented in term of \( D_{p50} \). It should be kept in mind, however, that since these ratios are somewhat scattered, there can be some doubt whether the \( D_{p50} \) truly represents the PSD, because pores with large \( D_p \), even if their volume is small, may control entire hydraulic conductivity. This point will be again later discussed in more detail.

Figure 5 shows how the \( D_p \) changes due to consolidation, using the newly defined \( D_{p50} \). The specimen was
consolidated in a conventional oedometer cell. With the increase in consolidation pressure ($p'$), the value of $D_{p50}$ becomes smaller and a linear relation exists between them in logarithm scales. However, the relation of $p'$ and $D_{p50}$ varies with samples. For example, even after the specimens were consolidated at 10 MPa, $D_{p50}$ of the Tokyo clay is 4 times larger than that of the Kyoto clay. This test result indicates that the variation in the $D_{p50}$ cannot be explained only by the consolidation pressure, though it is recognized that the magnitude of $p'$ is a very important factor in governing the PSD.

3.2 Relation between $D_{p50}$ and $e$

Figure 6 shows the relation between $e$ and $D_{p50}$. The relation for Louiseville clay is referred from a literature (Lapierre et al., 1990) and plotted in this figure. It is observed, as a general trend, that with the decrease in $e$, the $D_{p50}$ slightly decreases. However, there is a large scatter in this relation. For example, relations for Bangkok, Bothkennar and Pusan clays are located in the upper part of the relation for the Osaka bay clays. This means that these clays possess relatively larger pore diameters compared with that for Osaka bay clays, even at the same void ratio. The relations for Louiseville, Singapore and Drammen clays are located between them.

Figure 7 shows the relation between $e$ and $D_{p50}$ in the process of consolidation, where the data is using the same as in Fig. 6. As $e$ decreases due to consolidation, so does the $D_{p50}$. The range of these traces in $e$ and the $D_{p50}$ relation moves in the range of the relation measured at the intact condition. It is very interesting in noting that although the void ratio after consolidation to 10 MPa is nearly the same for all soils ($e$ is nearly equal to 0.5), the values of $D_{p50}$ for these clays are considerably different from each other, as mentioned earlier.
3.3 Relation between hydraulic conductivity and pore size

Figure 8 shows relation with e and k, which was measured by the Constant Rate Strain (CRS) oedometer test. As a general trend, k decreases with decrease in e. However, some scatter exists between them. For example, the k value for Tokyo and Drammen clays is relatively large in spite of their small e.

Until now, several researchers have tried to establish the correlation between the PSD measured by MIP and k value (for example, Garcia-Bengochea et al., 1979; Juang and Holts, 1986). As already discussed, it is found that the correlation between the PSD measured by MIP and k may be simply related by \( nD_{50}^2 \). The relation between \( nD_{50} \) and k is shown in Fig. 9, where the relations for both intact and consolidated states are plotted. In the figure, the relatively good correlation is seen between k and \( nD_{50}^2 \), except for Bothkennar and Bangkok clays. The k value for these clays is somewhat small in spite of large \( nD_{50} \) parameter. Since these clays have relatively large \( D_{50} \) compared to e, as shown in Fig. 6, the reason for large \( nD_{50}^2 \) is large \( D_{50} \), not n. Therefore, it can be said that even though Bothkennar and Bangkok clays have large pore diameter, their conductivity is small, probably due to lack of continuity of pores or different shape of pores.

4. PORE SIZE OF ARTIFICIALLY MIXED SOILS

Figures 10 (a) and (b) show the PSD for diatomite and sand mixtures in Singapore clay with different mixing ratios, respectively. In case of diatomite mixture, void ratio as well as \( D_{50} \) increases monotonously. Comparison of the two figures also reveals the tremendously large volume of voids in diatomite mixture. On the other hand, the PSD for mixture of Singapore clay and Toyoura sand looks more complicated as shown in Fig. 10 (b). Unlike the mixture of diatomite, with the increase in sand content, the void ratio decreases (this trend can be more clearly seen in Fig. 11). However, the \( D_{50} \) increases with increase in sand content, although the shape of the PSD is not so smooth as that of the diatomite mixture. When the content of sand become 75 %, the shape of the PSD suddenly changes and the pore size is distributed in a relatively narrow range. This may be caused by the development of aggregates, which consist of bridges of sand particles glued by small quantity of clay. As already mentioned, the shape of the PSD is not so much different for most natural soils because \( D_{60}/D_{50} \) is nearly constant as shown in Fig. 4. It should be kept in mind, however, that this finding cannot be applied to the sand mixture soil.

Figure 11 shows the relation between \( D_{50} \) and e, for naturally deposited soils and artificially mixed soils. Again this figure clearly shows that in case of sand mixture, void ratio decreases with increase in sand content. The opposite trend is observed in the diatomite mixture, since diatomite has large voids in its particle. However, the \( D_{50} \) of sand and diatomite mixtures increases with increase in their content. Especially it is likely that sand content plays an important role to increase the \( D_{50} \) in spite of small change in e, compared with the diatomite mixture. From this investigation, the reason for relatively large \( D_{50} \) for the Bothkennar clay is inferred due to the large content of silt and sand particles, as shown in Table 1; on the other hand, clay content of Singapore and Louiseville clay (for Louiseville clay, it is about 80 %, according to Lapierre, et al., 1990) is quite large, therefore, their \( D_{50} \) is small.
However, the reason for large $D_{50}$ of Bangkok clay as well as Osaka clays cannot be explained only by grain component. It may be concluded that though the grain component is an important factor in governing the PSD, there must be other equally important factors.

Figure 12 shows the relation between $k$ and $e$, measured by the CRS test for the artificially mixed soils. In the curve of $k$ and $e$ relation, there is a bending point indicated by an arrow. The sample was reconstituted under the consolidation pressure of 200 kPa. When $p'$ is smaller than the reconstituted pressure, the measurement of the pore water pressure is not accurate enough to calculate reliable value of $k$. Therefore, the relation between $k$ and $e$ should be considered only in the part of the left hand side from the arrow. It is very interesting to note that as void ratio increases or decreases due to increase in content of diatomite or sand respectively, the value of $k$ increases. This fact also confirms that $k$ cannot be determined only by void ratio.

Since the PSD of the mixture was measured after pre-consolidating at 200 kPa, the $k$ value of the sample may correspond to that indicated by the arrow in Fig. 12. Similarly, as in the case of the natural soils, the relation between $k$ and $nD_{50}^2$ for artificially mixed soils is plotted in Fig. 13. A good relation exists between $k$ and $nD_{50}^2$ in spite of so large scatter of $k$ and $e$ relation. However, the relation for the artificial mixtures is located in the same range as that of the Bangkok and Bothkennar clays, which are located in relatively upper part compared to other natural clays. The reason for this discrepancy may be attributed to 1) the representation of PSD as $D_{50}$ and 2) derivation of the pore diameter measured by MIP: i.e., assumption that the shape of pores is cylindrical. As already seen in Fig. 10 (b), the pattern of the PSD for sand mixture is considerably different compared to that of natural soils or diatomite mixture. The pattern of the PSD for even natural soils are slightly different from each other, if we carefully examine the ratio of $D_{50}/D_{50}$ or $D_{50}/D_{50}$ in Fig. 4. In addition, as pointed by Lapiere et al. (1990), the shape of real pore is somewhat different from cylindrical, which has been assumed to obtain the $D_{0}$ from MIP. This fact is easily recognized when we observe microstructure of soil by Scanning Electron Microscope.

4. CONCLUSIONS

Pore size distribution (PSD) for clayey soils is measured by Mercury intrusion porosimetry (MIP). To study the influence of grain composition, the PSD was measured in artificially mixed soils consisting of Singapore clay + Toyoura sand and Singapore clay + diatomite soil. The following conclusions are obtained:

1) From the relation between cumulated pore volume and pore diameter ($D_{p}$), the $D_{p30}$, $D_{p50}$ and $D_{p60}$ are defined by the corresponding $D_{p}$ at the cumulated volume with 30, 50 and 60 % of the total volume, respectively. It is found that the ratio of $D_{p30}/D_{p50}$ or $D_{p60}/D_{p50}$ is nearly identical for every soil. This means that the shape of the PSD can be considered to be not so different from each other, and the characteristics of the PSD can be represented by $D_{p50}$, the mean diameter of pores.

2) The value of $D_{p50}$ decreases with the increase in consolidation pressure, and its relation is linear in log-log plots.

3) There is a trend that the $D_{p50}$ for both intact and consolidated specimens decreases with a decrease in $e$, although there is a large scatter in this relation.

4) A good correlation exists between the hydraulic conductivity ($k$) and $nD_{50}^2$, where $n$ is the porosity, $e(1+e)$, for both the intact and consolidated specimens. However, $k$ for Bothkennar and Bangkok clays is relatively small in spite of their large $nD_{50}^2$. 

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**Fig. 12** Relation between void ratio and hydraulic conductivity for artificial mixture soils

**Fig. 13** Relation between $nD_{50}$ and hydraulic conductivity for artificial mixture and natural soils
5) Using artificially mixed soils, the influence of grain size component on the PSD was studied. From these two mixtures, completely opposite trend has been obtained; i.e., for the sand mixture, the $e$ decreases with the increase in sand content, while for diatomite mixture, the $e$ increases with the increase in diatomite content, because of the existence of large pore in particles of the diatomite. For both mixture of diatomite and Toyoura sand, however, $D_{50}$ increases with the increase in their contents. It is considered that the content of silt or sand particles has an important effect on the PSD.

6) The value of $k$ for both the sand and the diatomite mixtures increases with the increase in their content, even though the their $e$ and $k$ relations are completely different.

7) Ever for the artificial mixture, a clear relation exists between $nD_{50}$ and $k$. This relation is close to that of Bothkennar and Bangkok clays.

REFERENCES


