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学 位 論 文 題 目	TEMPERATURE EFFECTS ON GEOTECHNICAL PROPERTIES AND SOLUTE TRANSPORT PROCESSES IN CLAY （粘土の地盤工学的特性と溶質移動に温度が与える影響）
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論文の内容の要旨

The general structure of this study is illustrated in Fig. 1. This dissertation is organized by six chapters mainly consisting of laboratory measurements and analysis of geotechnical properties (consolidation characteristics, shear stiffness, and permeability), solute diffusion, and sorption in kaolin clay at different temperatures ranging from 5˚C to 40˚C.

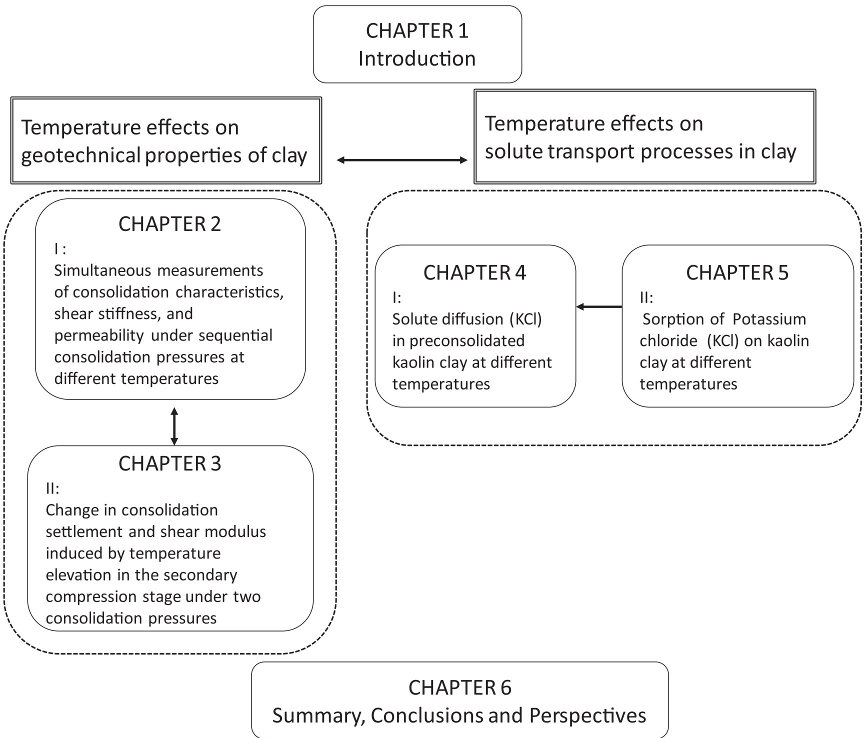


Figure 1 Structure of this study

In Chapter 1, it introduced that thermal disturbance released from the underground structures such as ground source heat pump (GSHP) systems in geoenvironment is leading to ground temperature change and may impact properties of sediments and biological and chemical processes in soil-water system. A thorough literature review reported the influence of temperature on geotechnical properties of clay sediments which are important for design consideration of any projects and the effects of temperature on solute transport processes in clay sediments which are essential for risk assessments of soil and groundwater pollution. Then, the present study was proposed to meet the lack of information and measurements on geotechnical properties and solute transport processes focusing on temperature variation range caused by operation of GSHP systems (±10˚C of local ground temperature).

In chapter 2, the simultaneous measurements of consolidation characteristics, shear stiffness, and permeability of preconsolidated kaolin clay under sequential consolidation pressures were conducted at different temperatures (5 ˚C, 15 ˚C, and 40˚C) by using modified oedometer apparatuses developed by our group. For both H 10 cm and the standard H 2 cm specimens, temperature influence on consolidation parameters such as coefficient of consolidation (c_v) and coefficient of volume compressibility (m_v) was observed as stress history dependent parameters showing higher c_v and lower m_v at higher temperature under overconsolidated (OC) state whereas constant c_v and m_v for three temperature conditions under normally consolidated (NC) state. Since temperature effects on c_v and m_v for both specimens were very similar, it can be suggested that one dimensional Terzaghi's theory was applicable for H 10 cm specimens used in the modified oedometer tests. For both specimens with high e_0 ($1.5<e_0<1.65$), the apparent preconsolidation pressure (P_{ac}) markedly increased with increasing temperature while compression index (C_c) slightly increased. The shear modulus ($G_{b.e}$) as a function of void ratio resulted from the successive applied consolidation pressures at 40 ˚C was larger than that at 5˚C even at the same void ratio. The trend of the measured $G_{b.e}$ under wide range of consolidation pressures closely followed the two existing empirical formulas for predicting of shear modulus. The increase in $G_{b.e}$ at higher temperature was explained by inter-particle contacts enhanced by inter-particle forces between particles resulted from the decrease in dielectric constant property of pore water at 40 ˚C. While permeability (k) of kaolin clay typically increased with increasing void ratio accordingly with Kozeny-Carman theory, temperature effects on the permeability (k) of the clay as a function of void ratio were not significant within the studied temperature range between 5 °C and 40 °C. The pore size distribution measurements verified this observation by indicating similar pore structure of the tested samples at 5˚C and 40˚C, and then, resulted in similar permeability. Despite the similar permeability of clay as a function of void ratio within the studied temperature range, the saturated hydraulic conductivity (K_{sat}) was basically affected by temperature like larger K_{sat} at higher temperature due to temperature dependent pore water viscosity and water density properties. As a result, it can be considered that temperature effects on consolidation parameter (P_{ac}) and shear stiffness ($G_{b.e}$) property of a unit tested sample were interrelated each other. Because OC condition illustrated in e/e_0-log p curve corresponded to that in $G_{b.e}$ vs e/e_0 relationship showing the variation with temperature. Similar slopes were obtained under different temperature for NC condition in both e/e_0 -log p curve and $G_{b.e}$ vs e/e_0 relationship. Consequently, the combined measurements of consolidation, shear stiffness and permeability and pore structure investigation provided a thorough understanding that for sample with high e_0 (>1.5), the changes in fabric structure (likely caused by the enhanced inter-particle forces between clay particles at higher temperature (>15˚C) would result in the increased shear stiffness and, consequently, higher P_{ac} at the temperature of 40˚C.

In chapter 3, the investigation was extended to certify the consolidation and shear stiffness properties induced by temperature elevation observed in chapter 2. Thus, for preconsolidated kaolin samples with high e_0 ($1.5<e_0<1.65$)

and low e0 (1.4<e_0<1.5), change in consolidation settlement and shear modulus were simultaneously measured under temperature elevation from 15 ˚C to 40 ˚C in the secondary compression stage at two consolidation pressures representing OC state and NC state. For both OC and NC clays, the decrease in normalized void ratio was observed after being heated for 7 days implying that heating reactivated the consolidation settlement in secondary compression stage. It was supported that the lower water viscosity at 40 ˚C accelerated the clay particle arrangements under a substantial stress. The Ref. samples which were continuously kept under constant temperature of 15˚C showed the decrease in void ratio after 7 days probably due to time dependent consolidation behavior. The magnitude of consolidation settlement in TC samples under OC and NC states were completely larger than that in Ref. samples and it was more significant in TC samples with high e under OC state. After temperature elevation for 7 days, the larger shear modulus (G_2) was attained for OC samples with both void ratios while G_2 did not recover back to the value of G_1 for NC samples. For OC samples, the greater shear stiffness was probably contributed not only by the increase in consolidation settlement but also by the closer inter-particle arrangements at 40˚C. For NC sample, the behavior of change in shear modulus was quite similar to the results obtained for saturated sand samples indicating that this behavior was resulted only from the temperature dependence of bender element performance and it is not representing the real shear stiffness response affected by temperature. The magnitude of increase in shear stiffness was significantly larger for samples with high e under OC state as compared to Ref. samples. In summary, it can be proposed that effect of temperature elevation was stress history dependent and consolidation and shear stiffness properties of the samples with high e_0 under OC state was highly depended on temperature elevation. The observation in chapter 2 in which greater stiffness at 40˚C since at 9.8 kPa corresponded to OC state was found for H 10 samples with high e_0 verified this assumption. Therefore, the increase in soil temperature (>15˚C) induced the consolidation settlement but with the increase in shear stiffness property of the clays having void ratio greater than 1.5.

In chapter 4, temperature effects on solute diffusion, one of the major solute transport processes, in preconsolidated kaolin clays were investigated through the tracer solution of potassium chloride (0.1M KCl) at different temperatures (6˚C, 15˚C, 40˚C) using specified volume diffusion (SVD) apparatuses. Both cation (K^+) and anion (Cl^-) diffusion behaviors as a function of temperature were examined for saturated samples with high e_0 (1.65) and low e_0 (1.15) (at two levels of compaction). For both ions, the relative concentration in the source chamber decreased with time while C/C_0 in the receptor chamber increased with time for the kaolin clays with both high e_0 and low e_0 mainly due to the solute diffusion from the source to the receptor through the clay. The solute diffusion coefficients (D_s) of both K^+ and Cl^- were evaluated using two-chamber semi-analytical method. The precise D_s values corresponding to change in concentration in the receptors were deduced by linearization of the measured data with extreme high coefficient of regression for all cases. The positive correlation between solute diffusion coefficient (D_s) and temperature was observed showing larger D_s at higher temperature for both cation and anion diffusions in the samples with high e_0 and low e_0 mainly due to less viscosity of pore fluid and greater atomic motion at higher temperature. The D_s for Cl^- ion were relatively higher than the D_s for K^+ ion mostly due to the retardation of K^+ ion on kaolin clay. The Arrhenius equation for temperature dependence diffusion was quite applicable for the potassium and chloride diffusions in kaolin clay at different bulk densities (void ratios). The activation energy (E_a) was independent of chemical species, density, and temperature in this study varying from 14.6 to 15.3 $kJmol^{-1}$. The solute diffusivity of K^+ and Cl^- ions as a function of water-saturated porosity (correlated to void ratio) was tested against the literature data for both clay minerals and natural soils suggesting a markedly higher liquid phase tortuosity for pure clay minerals. Therefore, this information of temperature effects on D_s as a function of water-saturated porosity need to be taken into account when predicting groundwater pollution associating with seasonal operation of GSHP systems.

In chapter 5, temperature effects on potassium and chloride sorption on kaolin clay were investigated by performing batch kinetic and equilibrium sorption experiments under different concentrations and at three temperatures (6˚C, 15˚C, 40˚C). Only potassium sorption on the clay was predominant while there was no sorption of chloride onto the clay. Potassium kinetic sorption onto the clay showed that Pseudo second order kinetic rate constant at 40˚C was greater than at 6˚C implying that sorption rate was faster at higher temperature. Generally, the equilibrium sorption at three temperatures was fitted with Freundlich isotherm model. The Freundlich adsorption coefficients increased with increasing temperature due to dominant cation exchange mechanism between exchangeable K^+ and positive charge onto the clay surface resulting from change in pH at high temperature. Additionally, retardation factor (R_d) was negatively correlated with equilibrium concentration of potassium showing the lower value of R_d (1.1 to 1.3) at highest concentration of 4000ppm. This was in agreement with the observation of slower potassium diffusion as compared to chloride diffusion with the ratio of 1.1 to 1.2. Therefore, this investigation represents temperature effects on exchangeable ion sorption behaviors onto non-swelling clay.

Based on the measured data and results, this study revealed that solute/contaminants transport in porous media rather than geotechnical properties of clay sediments should be concerned as a critical issue under elevated temperature condition (around + 25˚C to local typical ground temperature). Furthermore, the whole research attributes to the platform knowledge towards a unified understanding of temperature effects on geotechnical properties and solute transport process in clay. The thermal disturbance in geoenvironment influences geotechnical properties and solute transport parameters accordingly with temperature dependence pore fluid and fabric structure properties.

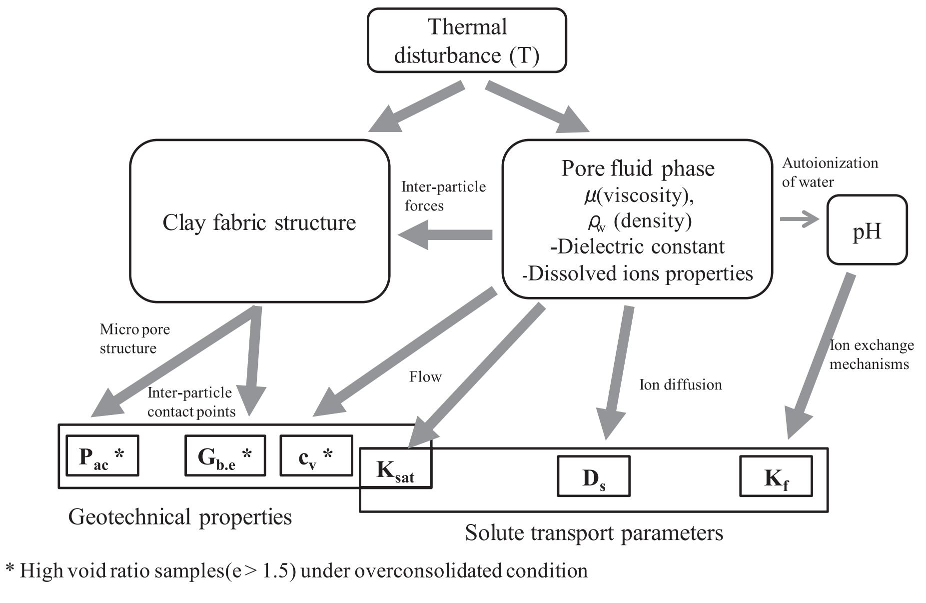


Figure 2 Effect of temperature onto geotechnical properties and solute transport processes.

Following this study, the future studies are required to comprehend the above understanding by conducting similar experiments using undisturbed natural sediments around the heat exchange tube of GSHP systems with other important dissolved chemicals. Then, it might be useful for risk assessments of the issues related to the intensive application of GSHP systems in geoenvironment.

論文の審査結果の要旨

当学位論文審査委員会は，平成 25 年 8 月 5 日に論文発表会を開催し，論文内容の発表に続いて質疑と論文内容の審査を行なった。以下に審査結果を要約する。

近年，省エネや二酸化酸素排出量削減を目的とし，地下水温と外気温との温度差エネルギーを用いた冷暖房システム（地中熱利用ヒートポンプシステムなど）の普及が世界的に広まっている。一方で，地下空間の積極的活用を目的とした深度地下開発も盛んに行われている。このようなシステムや深度地下開発は，年間を通して水温がほぼ一定に保たれている地下環境での熱交換（排熱や吸熱）を引き起こすことから，人為的な熱攪乱を地下環境に与えることとなる。地下熱環境の変化が与える環境影響としては，地盤力学特性（圧密特性など）の変化，重金属類などの溶解・移動，地盤内物質（溶質，水，ガスなど）移動特性の変化，土壌微生物・細菌の増殖などが予想される。しかし，現状では地下熱環境の変化が及ぼす環境影響については，十分な知見が得られているとは言い難く，これらの影響を定量的に評価できる環境アセスメントツール等もこれまで提案されていない。このような背景のもとに，本学位論文では，粘土（カオリン粘土）を対象に，特に熱環境の変化（異なる温度条件下，温度の上昇）が地盤力学特性と地盤内物質移動特性に及ぼす影響を室内実験によって調べ，これらの特性を表現するパラメーラ群（圧密係数・せん断剛性，透水係数，溶質拡散・吸着係数など）の温度依存性に注目することで，熱環境の変化が地盤骨格・間隙水特性に及ぼす影響を整理した。

第 1 章では，研究の背景や目的，関連する既往の研究について記述している。既往の研究については，特に本研究で主要な内容となる，粘土の地盤工学的特性（圧密や透水など），溶質移動パラメータ（土壌溶質拡散係数や吸着係数）に温度が及ぼす影響についてのレヴューを詳細に行っている。

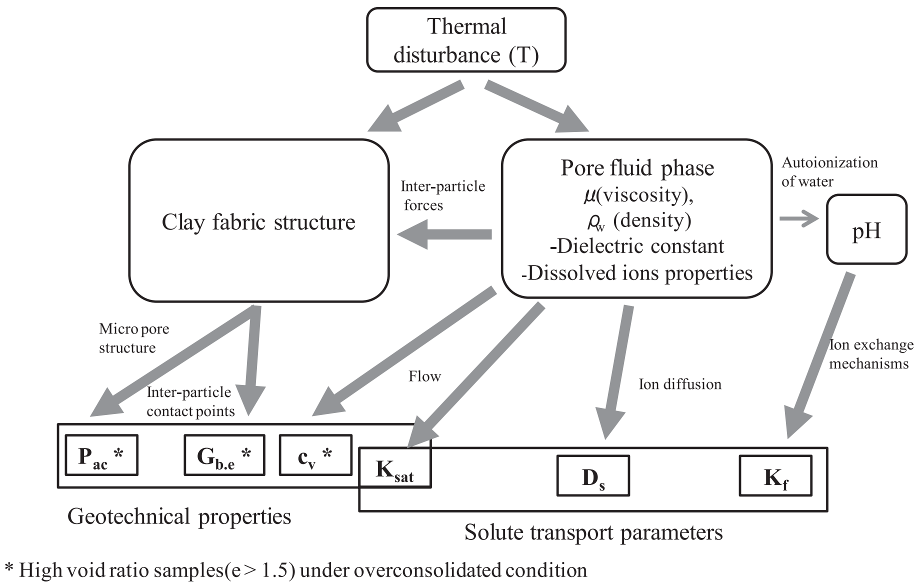
第 2 章では，実験材料としてカオリン粘土を用いて，ベンダーエレメントを装着した温度調節型圧密試験装置を新たに開発し，異なる温度下（5，15，40℃）で圧密試験を実施している。試験では，e-log p 曲線，圧密パラメータ（圧密係数 C_v，体積圧縮係数 m_v，見かけの圧密降伏応力 P_{ac}，透水係数 k），せん断剛性 - 間隙比関係（$G(e)$）を求めた。その結果，C_v・m_v・k には温度の影響があまり見られないものの，P_{ac} と $G(e)$ は高温条件下で大きな値を示すことが明らかになった。間隙径分布に温度影響がほとんど見られなかったことから, 高温下で粘土粒子の接点間力が増したことがせん断剛性 G を大きくした理由として考察を試みている。

第 3 章では，同カオリン粘土の二次圧密下で温度を 15℃から 40℃に上昇させた場合の，圧密沈下量（ひずみ変化量）とせん断剛性 G の変化を調べている。その結果，見かけの圧密降伏応力 P_{ac} よりも低応力条件である過圧密領域で温度上昇を与えた方が，正規圧密領域で温度上昇を与えた場合よりも大きな圧密沈下量を示すこと，それに伴いせん断剛性 G が大きくなるなどの新たな知見が得られている。

第 4 章では，カオリン粘土中の溶質拡散に温度が及ぼす影響を明らかにするため，定体積拡散装置（Specified Volume Diffusion Test）を用いて異なる温度下（6，15，40℃）で溶質拡散係数 D_s を実測している。実験には 0.1M の KCl 溶液を用いて, K^+ 及び Cl^- の D_s をそれぞれ測定している。その結果, Cl^- の D_s は, K^+ の D_s の 1.1 ～ 1.2 倍であること，両イオンの D_s 値は温度との間に良好な線形関係が得られるなどの結果が得られた。さらに, D_s の温度依存性にアレニウス式の適用が可能であること, アレニウス式中の（見かけの）活性化エネルギーの違いが D_s の温度依存性を上手く表現できること，を明らかにしている。

第5章では，実測したカオリン粘土の溶質拡散係数 D_s に遅延（遅延係数 R_d）がどのように影響を及ぼしているのかを考察するために，異なる KCl 溶液濃度及び温度下（6，15，40℃）でバッチ吸着実験を行っている。その結果，Cl^- はカオリン粘土への吸着を示さず，D_s に遅延の効果がないのに対して，K^+ は擬2次反応速度的吸着を示し，高温条件（40℃）の方が低温条件（6℃）よりも大きな吸着速度・係数を示し，遅延効果が認められることを明らかにしている。さらに，前章の溶質拡散実験で用いた濃度における K^+ と Cl^- の R_d 値の比は 1.1 ～ 1.3 倍となり，Cl^- と K^+ の D_s 比が遅延係数 R_d の比からも説明できるといった知見も得られている。

第6章では，第2章から第5章までに得られた知見を，特に，地盤力学特性と地盤内物質移動特性を表現するパラメーラ群（圧密係数・せん断剛性，透水係数，溶質拡散・吸着係数など）の温度依存性に注目することで，熱環境の変化が地盤骨格・間隙水特性に及ぼす影響を整理している（下図）。また，本章は結論と今後の研究展開の方向性も示している。



以上のように，本研究は，熱環境変化が地盤力学特性や地盤内物質移動特性に及ぼす影響を，これらの特性を表現するパラメータ群の温度依存性によって評価・整理したものであり，その成果は今後地下熱環境の変化が及ぼす環境影響を定量的に評価する環境アセスメントツールの開発に効果的に活用されるものである。このことから，当学位論文審査委員会は，本論文が博士（学術）の学位に相応しい内容であると判断した。

なお，本論文の内容は，第2章が J. Geology and Earth Sciences，第3章が国際プロシーディングス論文 Proceedings of 18th Southeast Asian Geotechnical Engineering に掲載済みであり，第4章＆第5章は J. Geotechnical and Geoenvironmental Engineering, ASCE へ投稿原稿準備中である。