

Enhanced performance of bentonite-based materials for preservation of geoenvironment

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ABSTRACT

Vertical cutoff wall and mercury waste landfill are in-situ and ex-situ countermeasures, for contaminations containment for preservation of geoenvironment. Bentonite-based materials (e.g. bentonite, soil-bentonite, soil-bentonite-cement etc.) are extensively utilized as barrier materials in these containment techniques. This study aims to investigate the feasibility and efficacy of these bentonite-based materials through comprehensive laboratory tests.

The mechanical performance of soil-bentonite mixture amended with slag cement was studied based on the results of consolidated-undrained (CU) isotropic triaxial compression tests. the stress-strain characteristics, failure modes, shear strength properties, etc. were evaluated. The results indicated that the addition of slag cement positively affects shear strength (e.g., S_u and S_r), elastic-plastic properties (e.g., E_0 and E_{50}), and effective cohesion coefficient (i.e., c'). However, it has a negative effect on the stress-strain behavior and the final failure mode. The curing effect was similar to the effect of slag cement content, with almost no variation observed in the effective friction angle (ϕ') in this test. The unamended specimens were highly stress-dependent, with consolidation stress (i.e., confining pressure) being the major influencing factor. In contrast, the amended specimens were primarily affected by cementation.

The hydraulic performance of soil-bentonite mixture amended with slag cement was studied based on the results of free swelling test, and hydraulic conductivity test on triaxial apparatus and flexible wall permeator. Results indicate that the swelling performance of bentonite improved with the addition of cement reagents but declined in a high EC (i.e. electrical conductivity) environment. The hydraulic performance of amended specimens in the flexible wall was time-dependent due to continuous hydration, with permeation equilibrium observed only at a cement content of 25 kg/m³. In the triaxial apparatus, cement content and curing time significantly affected the hydraulic conductivity of amended specimens, whereas consolidation stress (i.e., confining pressure) was the major factor affecting unamended specimens, which exhibited volumetric strain exceeding 20%.

The adsorption performance of pure bentonite on gaseous mercury was studied based on the results of constant volume adsorption test. The swelling performance of bentonites in ferrous solution was estimated in normal and anaerobic conditions. The results indicated that dry bentonite has a very high adsorption capacity, whereas the adsorption capacity significantly diminishes in specimens with water content, with the distribution coefficient dropping from thousands to tens of mL/g. Additionally, sodium bentonite demonstrated superior adsorption

performance. The swelling performance of bentonite in ferrous solution was influenced by the ferrous concentration: ion exchange enhanced swelling at low concentrations, while strong ionic strength restricted ion exchange at high concentrations. Additionally, the oxidation of ferrous ions was successfully prevented under anaerobic conditions.

In conclusion, this study provides valuable insights into the feasibility of slag cement amendment for soil-bentonite mixtures in vertical cutoff walls, and the efficacy of pure bentonite as barrier layer in mercury waste landfill. The results from triaxial compression tests, hydraulic conductivity tests, free swelling tests and constant volume adsorption tests etc., contribute to a comprehensive understanding in the mechanical performance, hydraulic performance and adsorption performance of bentonite-based materials.

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Chapter 1

Introduction

1.1 General remarks

Soil pollution in urban areas is mainly caused by landfills and the underground seepage of hazardous substances on factory sites (private property). Soil contamination, which must be prevented and remediated for soil protection, is recognized as a form of significant environmental pollution under Japanese environmental law (Ishimaki et al., 2024). In Japan, soil contamination on agricultural land was addressed with legislation in 1970. However, soil contamination in urban areas was not addressed until 2002, with the enactment of the Soil Contamination Countermeasures Act.

Figure 1.1 shows the changes in the number of investigations for soil contamination cases in Japan (MOEJ 2024). The number of investigations increased around 2002, when the law was promulgated, and increased again in 2010 as a result of the amendment of the law, which aimed to extend the conditions needed to investigate and develop more proper and effective countermeasures. As can be seen in Fig. 1.1, the number of investigations for soil contamination cases decreased after 2020, indicating the continuous social efforts in using countermeasures (i.e. containment techniques) for preservation of geoenvironment.

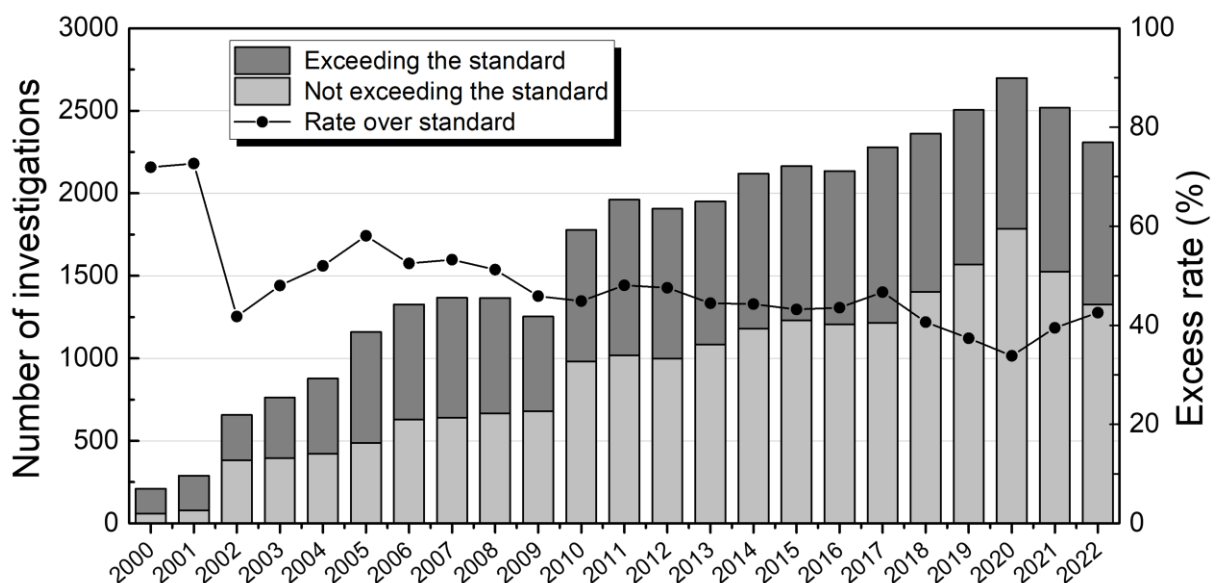


Fig. 1.1 Number of investigations on soil contamination in Japan (MOEJ 2024).

The most distinctive feature of soil contamination is that, unlike air and water pollution, hazardous substances remain in the ground semi-permanently, known as stock pollution, unless they are removed through artificial means (Ishimaki et al., 2024). Countermeasures for soil and groundwater contamination can be divided into in-situ and ex-situ treatment methods.

In-situ treatment methods involve treating contamination directly at the site without removing the soil or groundwater. Techniques such as in-situ bio-remediation, chemical oxidation, stabilization/solidification (S/S), thermal treatment, and phytoremediation are used. These methods are suitable for large or inaccessible sites, aim to minimize site disturbance, and are often more cost-effective for long-term remediation projects. The choice between ex-situ and in-situ methods depends on the type and extent of contamination, site conditions, regulatory requirements, and cost considerations.

Ex-situ treatment involves removing the contaminated material from its original location for treatment elsewhere. This approach includes techniques such as excavation and removal, soil washing, thermal desorption, bioremediation, and pump and treat. These methods are typically used when contamination is localized and accessible, or when immediate risk reduction is required.

In this chapter, an in-situ containment technique and an ex-situ containment technique are introduced, providing the general background for this doctoral research, as bentonite-based materials (e.g. bentonite, soil-bentonite, soil-bentonite-cement) were extensively used. The objectives and structure of this dissertation are also illustrated.

1.2 Bentonite-based materials for preservation of geoenvironment

1.2.1 In-situ containment technique: Vertical cutoff walls

The leakage of heavy metals from heavy industrial bases, tailings dams, and landfill sites can spread through rainfall and groundwater flow, as shown in Fig. 1.2 (Geng et al., 2023). To prevent these pollutants from harming the local biological ecosystem, cutoff walls are typically constructed to block their dispersion. These walls are placed at a certain distance from the pollution source, creating an area known as the transition layer. The region behind the cutoff wall is referred to as the aquifer. Cost-effective vertical cutoff walls, as a valid In-situ containment technique, have been extensively used worldwide in remediation projects for various contaminated sites and landfills (Evans, 1994; Ryan and Day, 2002; Opdyke and Evans, 2005). Such vertical barriers have been constructed to intercept pathways of pollution and to prevent their migration in the aquifer, as shown in Fig. 1.2 (Evans et al., 2008; Soga et al., 2015; Takai et al., 2016).

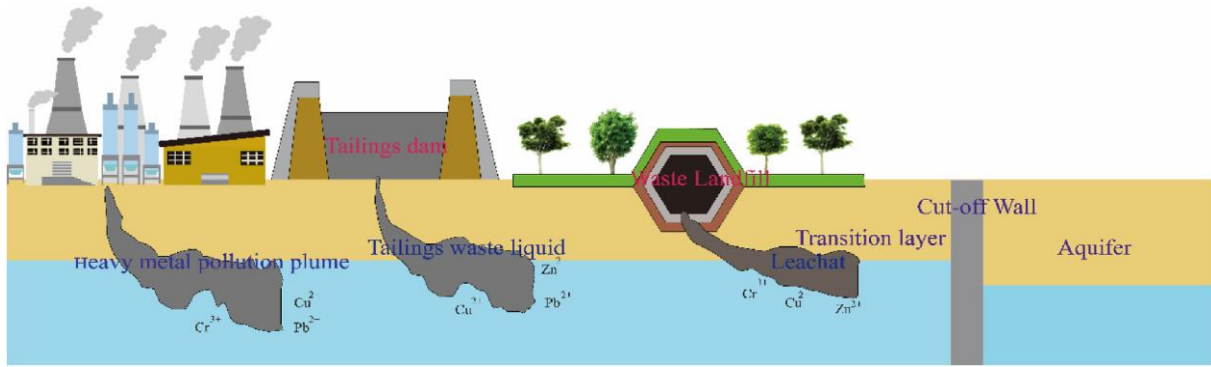


Fig. 1.2 Schematic of pollutants passing through transition layers to cutoff wall (Geng et al., 2023).

Bentonite, labeled with good swelling properties, self-healing performance and extremely low hydraulic conductivity, is extensively utilized in vertical cutoff walls. In accordance with the composition of backfill materials, the vertical cutoff walls using bentonite can be categorized as soil-bentonite (SB), cement-bentonite (CB), and soil-cement-bentonite (SCB) (Opdyke and Evans, 2005; Du et al., 2015).

To install the SCB cut-off walls, the trench cutting and re-mixing deep wall method (TRD) is one of the most technically acclaimed methods (Evans, 2007; Cheng et al., 2022). The construction process of TRD method is executed in two-phase (i.e. trench cutting and re-mixing), as shown in Fig. 1.3 (Katsumi et al., 2008). In the first phase, the trenching machine excavates the soil and mixes with bentonite slurry for homogeneity. Herein, the sodium bentonite with superior swelling capacity and adsorption properties, is extensively applied (Gleason et al., 1997; Kamon et al., 2006; Malusis et al., 2009). In the second phase, additional bentonite powder blends with water and re-mixes with the soil-bentonite mixture. For SCB walls, the cement is blended in the second phase.

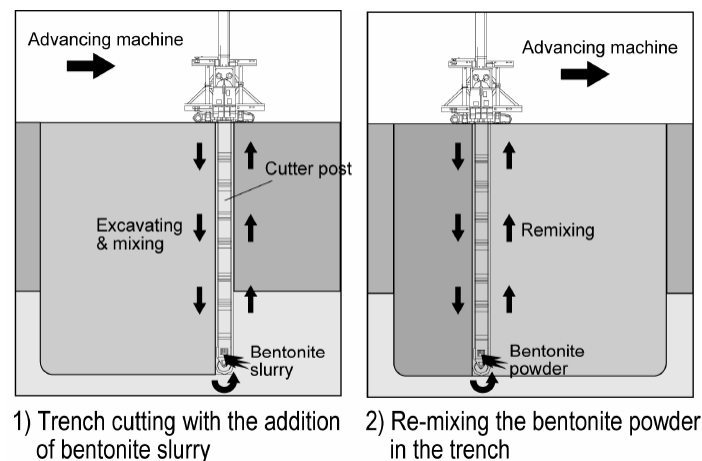


Fig. 1.3 Construction process of SBM vertical cut-off wall with TRD method (Katsumi et al., 2008).

The SB walls, with extremely low hydraulic conductivity and high deformability, have been widely applied. However, SB exhibits relatively low shear strength (i.e. inadequate strength properties), as a result, the effectiveness will be influenced by foundation loads (static loads) or in situations of dynamic loads (Opdyke and Evans, 2005). In order to enhance the mechanical properties of soil-bentonite mixtures, cementitious binders including but not limited to Ordinary Portland Cement (OPC), ground granulated blast-furnace slag (GGBS), ground granulated blast-furnace slag cement and fly ash (FA), have been studied and applied in vertical cutoff walls and other land remediation and ground improvement projects (Shackelford and Jefferis, 2000; Wu et al., 2019; Du et al., 2020).

Previous studies evaluated the strength properties and hydraulic performance of soil-cement-bentonite using OPC through unconfined compression test (UCS test) and hydraulic conductivity test (Ata et al, 2015; Huang et al., 2021). Tian conducted UCS test on the slag cement amended soil-bentonite mixtures using cement content of 50, 75 and 100 kg/m³ after 60, 90 and 180 curing days. The results indicated that the UCS value is dependent on the curing time and is significantly affected by higher cement content (Tian 2021). The UCS test does not yield any information about the stresses carried by the soil mass and water phases, making it of limited use as a fundamental test.

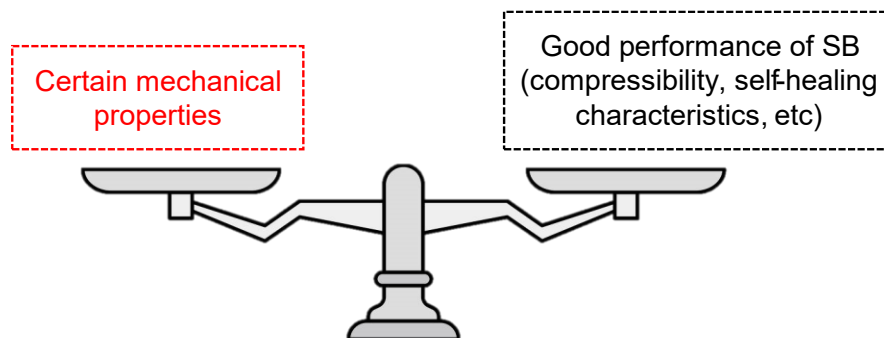


Fig. 1.4 The balance achieved after slag cement amendment.

As shown in Fig. 1.4, it is worthy to investigate the enhanced mechanical performance of SB amend with low content of slag cement, with respect to achieve a balance between certain strength properties and compressibility, as well as self-healing characteristics. Simultaneously, the unpredictable variations of permeability in instances of low slag cement content amendment are also crucial for investigations.

1.2.2 Ex-situ containment technique: Mercury waste landfill

For human reasons, the emissions or releases of mercury may be caused by improper handling, collection, transportation or disposal of mercury wastes. Once mercury is emitted or released into the environment, it persists in the atmosphere, soil and water bodies (Zheng et al., 2021; Zhu et al., 2013). Some of the mercury that is released or emitted into the environment ends up in the food chain, mainly due to the biomagnification of bioavailable mercury, as a result, leading to Mercury Poisoning or even Minamata Disease (Berlin et al., 2007). The discharge of wastewater containing mercury into Minamata Bay, Japan, from 1932 to 1968, the illegal dumping of mercury-contaminated waste in Cambodia in 1998 and the case involving Thor Chemicals in South Africa in 1980s are examples of cases in which wastes containing or contaminated with mercury or mercury compounds were not managed in an environmentally sound manner.

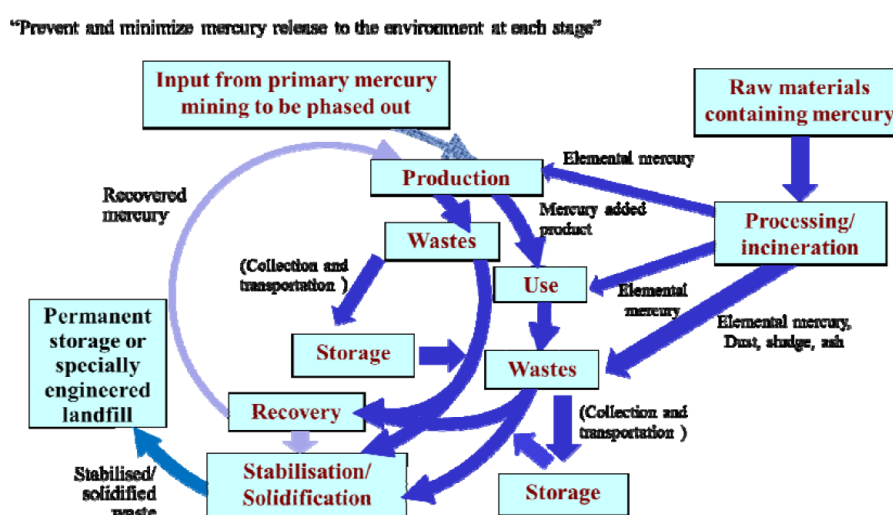
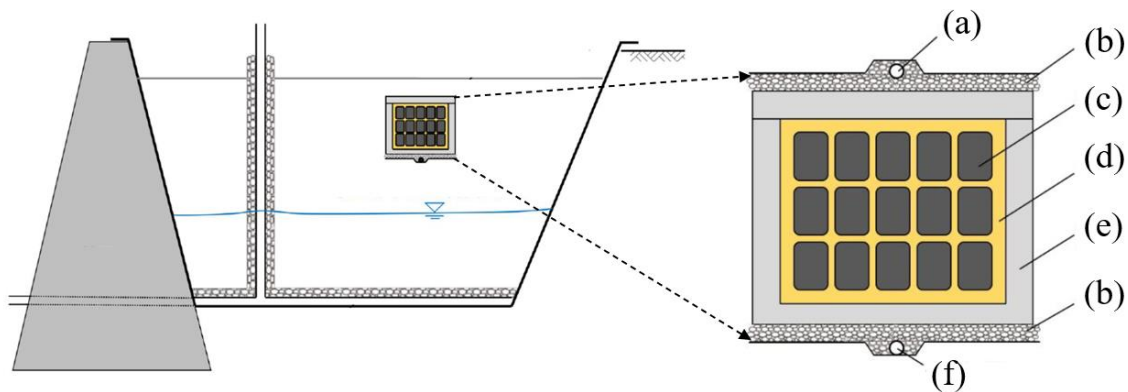


Fig. 1.5 Basic concept of mercury management (UNEP/CHW.12/5/Add.8/Rev.1).

The Minamata Convention recognized mercury, reached in 2013 by more than 100 countries, as a chemical substance of global concern and was intended to protect human health and the environment from anthropogenic emissions and releases of mercury and other substances. Also, with the purpose of ensuring proper treatment of mercury waste by specifically explaining matters to be considered in the new handling, collection, transportation, disposal, etc. In 2015 Basel Convention Conference (COP12) by UNEP (United Nations Environment Program) on the control of Transboundary Movement of Hazardous Waste and Their Disposal, mercury life cycle management was mentioned as the basic concept of mercury management, which is shown in Fig. 1.5. The termination of mercury waste is quite important

for the feasibility and reliability of Stabilization/ Solidification and permanent storage.

According to the ministry of the environment in Japan, the mercury supply has decreased from the peak of 2,500 tons per year in 1964 to less than 5 tons per year after 2016. As the convention limited the use of mercury containing products, safe and reliable methods for the final disposal and control are required. The former used method for the disposal of these wastes is in landfill. However, there exist mercury gas leakage both in the construction procedure and further utilization of bottom ash. As measured around a Hg-polluted traditional landfill in Japan, the Hg concentration through bioaccumulation in soil earthworm near the landfill is 10 to 15 times than common forest soil (He et al., 2018).



* (a) Gas sensor, (b) Crushed stone, (c) Stabilized and solidified mercury waste, (d) Impermeable layer with mercury adsorption capacity, (e) Concrete structure, (f) Drainage pipe.

Fig. 1.6 Schematic of specially constructed mercury waste landfill (MOEJ 2021).

Based on these circumstances, the Ministry of Environment in Japan (MOEJ) put forward the law, that mercury waste should be disposed after stabilized and solidified in an environmentally sound manner (MOEJ 2021). After the process of stabilization/ solidification, the treated waste should be permanently stored in specially constructed landfill, as shown in Fig. 1.6. This special structure is aimed to prevent water intrusion in situations of changeable water level caused by heavy rainfall. And the impermeable layer also functions as potential adsorbent in case of unavoidable mercury gas emission from the treated solid waste.

Bentonite, as a widely used buffering material with high swelling performance and low hydraulic conductivity, also recommended for usage as impermeable layer (i.e. compacted bentonite layer) in this specially constructed mercury landfill. Basically, bentonites are classified as sodium bentonite, calcium bentonite and sodium/calcium-bentonite. In previous studies, sodium bentonite presents better impermeability because of tighter connection between

diffuse electrical double layer, while calcium bentonite present better adsorption capacity for heavy metals due to higher specific surface area for adsorption reaction on the interstitial surface (Komine 2004; Lin et al., 2018).

After stabilization and solidification (S/S) process, the mercury waste is stored in the landfill in Fig. 1.6. However, there is still possibility of gaseous mercury leaching (evaporation). Previous study investigated the leaching concentration of mercury waste after S/S in different types of aqueous environments (i.e. acidic, alkaline, oxidation, reduction, neutral). The results indicate that the leaching concentration of gaseous mercury was lower than 0.0001 ng/L in reduction environment and around 0.1 ng/L in alkaline and oxidation environment (Egawa et al., 2021). Therefore, it is worthy of investigating the adsorption capacity of bentonite on the gaseous mercury in situations of low content of evaporation. At the same time, the swelling performance of bentonite in anaerobic condition is also unknown.

1.3 Objectives

The main objective of this study is to evaluate the feasibility of using low content of slag cement as alternative cementitious binder for the amendment of mechanical performance of soil-bentonite mixtures and maintain its deformability and unpredictable long-term hydraulic performance as well.

SB cutoff walls have extremely low hydraulic conductivity and high deformability. However, the wall is potentially compressed under high static loads (i.e. overburden pressure) or dynamic loads, even with arching effect (Evans et al., 1995; Ruffing et al., 2010). The slag cement, as eco-friendly binder comparing with OPC, is innovative to be studied in the amendment of soil-bentonite mixtures to obtain certain mechanical performance and maintain hydraulic performance as well. High cement content diminishes the swelling and self-healing properties of bentonite, whereas extremely low cement content is not practical. Hence, the effect of cementation and consolidation, considering the real stress conditions, is crucial to be discussed. As well as the long-term hydraulic performance, which affected by the cement hydration throughout the permeation even takes over 90 days (Opdyke and Evans., 2005).

The other issue is to investigate the influences of ferrous and ferric environments on the swelling performance of bentonites, and the adsorption performance of bentonites on elemental gaseous mercury in situations of potential evaporation of gaseous mercury from the stabilized and solidified (S/S) mercury waste.

The adsorption performance of pure bentonite is extensively studied in reagents with various kinds of cations. However, the adsorption capacity on elemental mercury in gaseous

phase is unknown. In the mercury waste landfill, the swelling performance is potentially affected by the ferrous environment. The effect of ferrous concentration, especially under anaerobic condition, is important to be estimated.

1.4 Structure of the dissertation

This thesis consists of 7 chapters. The constitution of the thesis is shown in Figure 1.6, and the experimental methodologies applied for this study are shown in Table 1.1.

In this chapter (Chapter 1), the objectives and the contents of the dissertation are clarified. General background information related to vertical cutoff wall and mercury waste landfill in Japan is presented. Also, fundamental information of bentonite-based materials (e.g. bentonite, soil-bentonite and soil-bentonite-cement etc.) as barrier materials are described.

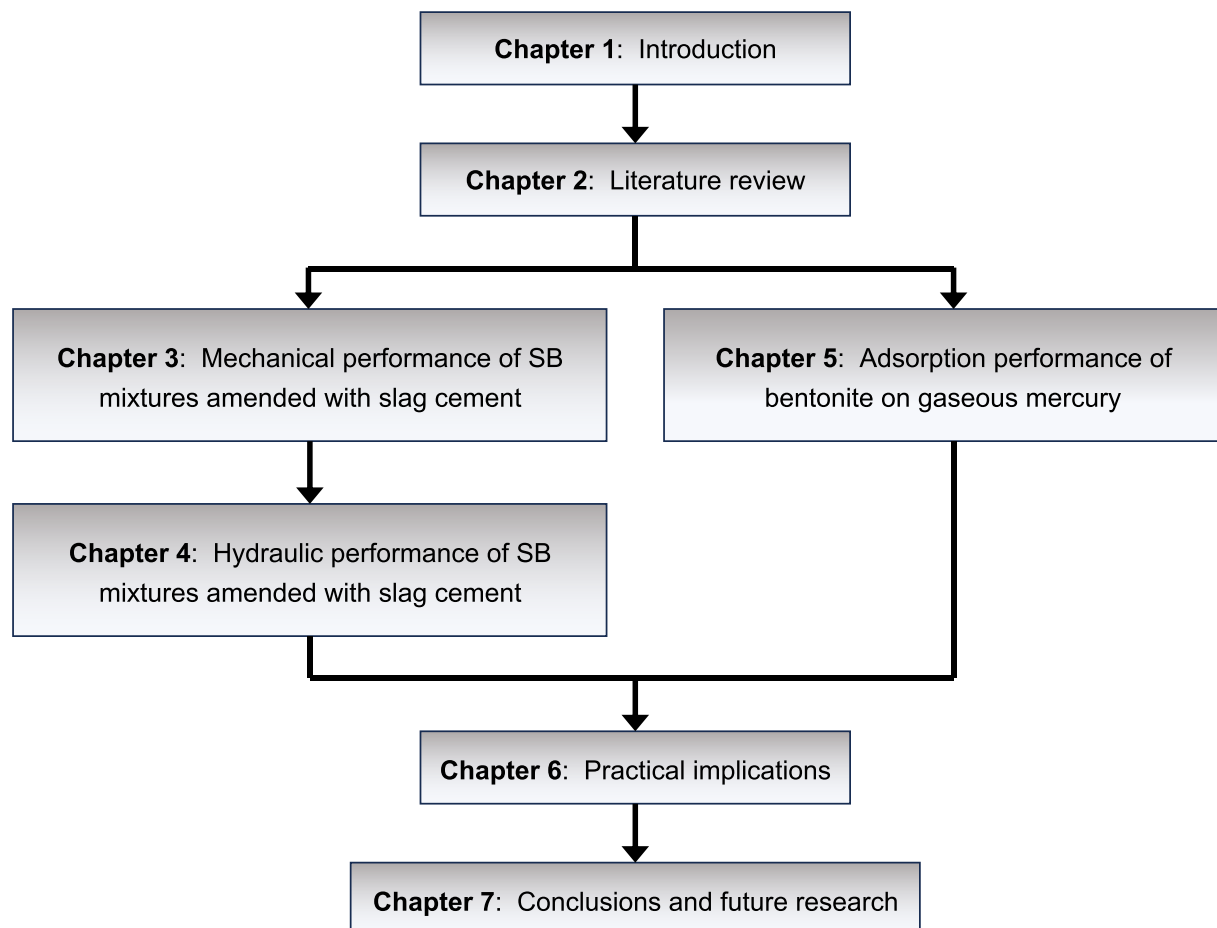


Fig. 1.7 Structure of the dissertation.

In Chapter 2, literature review of the basic mechanism of bentonite-based materials working as barrier materials is presented. Current studies on the mechanical and hydraulic

performance of vertical cutoff walls are discussed, as well as new cementitious binders for enhancement. Adsorbents for the gaseous mercury are reviewed and the utilization of bentonite in other gas or vapor adsorption are presented.

In Chapter 3, mechanical performance of soil-bentonite mixtures amended with slag cement is studied. The basic mechanical properties (e.g. undrained shear strength, undrained strength parameters (c' and ϕ') etc.) from consolidated undrained triaxial compression test are presented. And the effect of cement content, curing time are discussed, as well as the effect of confining pressure, which affect the consolidation process (i.e. void ratio variations).

In Chapter 4, the swelling performance of bentonite in cement reagents is studied. The hydraulic performance of soil-bentonite mixtures amended with slag cement is estimated. In the flexible wall permeator, long-term hydraulic performance combined with cement hydration is evaluated. In the triaxial apparatus, the effect of cementation and the volumetric strain on the hydraulic conductivity are evaluated. The test results in this two apparatus are compared and potential influencing factors are discussed.

Table 1.1 Experimental methodologies in this study.

Chapter	Methodologies	Evaluation approach
3	Triaxial compression test	Stress-strain characteristics
		Failure modes
		Shear strength properties
		Effective stress path and strength envelope
		Excess pore pressure characteristics
		Young's modulus E_0 and secant modulus E_{50}
4	Free swelling test	Effect of cement concentration
	Hydraulic conductivity test #1	Effect of cement addition
		Long-term hydraulic performance
	Hydraulic conductivity test #2	Effect of cement addition
		Effect of consolidation stress
		Effect of curing time
5	Free swelling test	Effect of initial Fe^{2+} concentration
		Comparison of normal and anaerobic conditions
	Gaseous Hg adsorption test	Effect of water content
		Effect of bentonite type

* #1 Flexible wall permeator, #2 Triaxial apparatus.

In Chapter 5, the swelling performance of bentonite in ferrous and ferrous/ferric solutions is studied. The effect of initial Fe^{2+} concentration and the mechanism are evaluated. The adsorption capacity is estimated and compared in sodium and calcium bentonites. The effect of water condition is also discussed.

In Chapter 6, practical implications are described based on experimental results obtained in each chapter in terms of mechanical performance, long-term hydraulic performance, swelling properties and adsorption capacity.

In Chapter 7, conclusions and future research are presented.

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Chapter 2

Literature review

2.1 General remarks

Bentonite is one of the oldest clays that humankind has been using from ancient times as traditional. In recent years researchers have found many applications of bentonite clay due to its various physio-chemical properties (surface area, adsorption, swelling properties, cation exchange properties, etc.) (Borah et al., 2022). Bentonite is primarily composed of the mineral montmorillonite, and significant characteristics is the formation of a double electrical layer when contact with water. This double layer consists of a tightly bound negatively charged layer on the montmorillonite surface and a diffused layer of positively charged ions (cations) in the surrounding water, as shown in Fig. 2.1 (Komine et al., 2009; Wang et al., 2020).

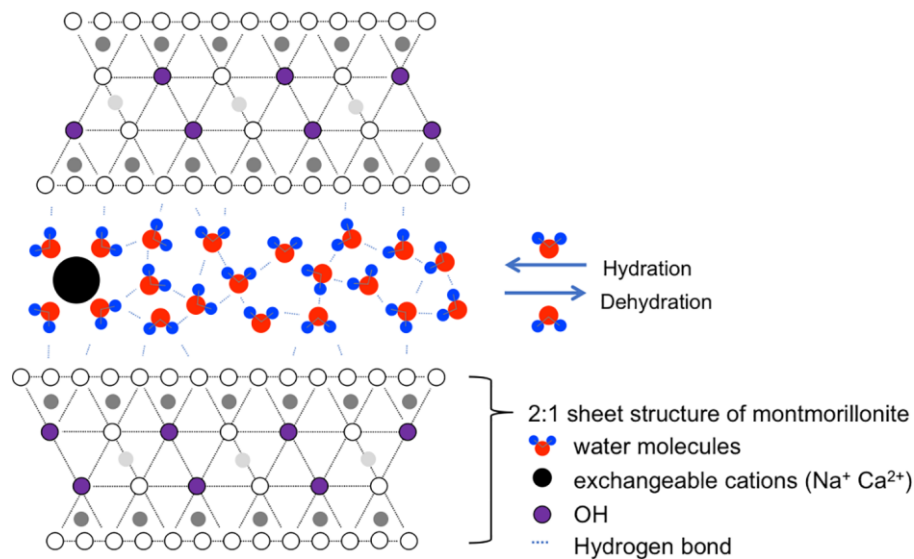


Fig. 2.1 Models of the hydration and dehydration of water in montmorillonite (Wang et al., 2020).

This feature underpins high cation exchange capacity (CEC), which typically ranges from 60 to 120 meq/100g, depending on the specific mineral composition and solution conditions (Ito and Komine et al., 2008; Dohrmann et al., 2012; Choo and Bai., 2016; Ruan et al., 2022). The high CEC allows bentonite to adsorb and immobilize a wide range of cations, including heavy metals and radionuclides, making it an effective material for environmental protection applications.

Additionally, bentonite exhibits significant swelling properties when it absorbs water. This swelling occurs because water molecules infiltrate the interlayer spaces of montmorillonite, causing the layers to expand, as shown in Fig. 2.2 (Komine and Ogata 2003). The swelling performance upon hydration contributes to extremely low permeability, thus it is normally utilized in impermeable barriers. Studies have shown that bentonite barriers can achieve permeabilities as low as 10^{-11} m/s, providing a highly effective barrier against fluid migration (Cho et al., 1999; Shackelford and Sample 2014; Ören and Akar 2017).

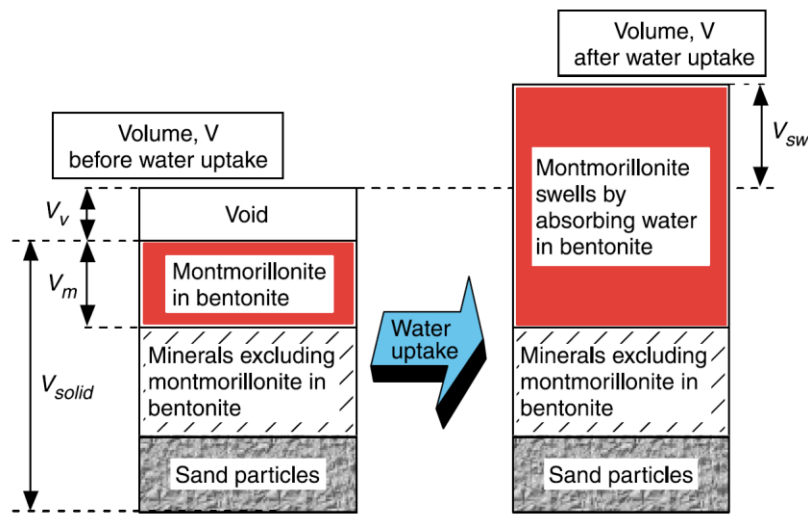


Fig. 2.2 Composition of sand-bentonite mixture before and after contacting water (Komine et al., 2008).

The primary applications are in in-situ containment technique as liners for landfills and ex-situ containment technique for hazardous waste containments. The low permeability and high adsorption capacity are crucial for preventing the migration of contaminants into surrounding soil and groundwater. For instance, in municipal solid waste landfills, a compacted bentonite layer is often used with geomembranes to form a composite liner system that effectively minimizes the breakthrough of leachate (Lee and Shackelford 2005). Furthermore, bentonite is utilized in the isolation of high-level radioactive waste (Pusch 1992). The swelling property of bentonite ensures that any cracks or gaps that might form in the barrier material due to ground movements are self-sealed (i.e. self-healing properties), maintaining the integrity of the containment system (Villar 2021).

While bentonite offers many advantages as a barrier material, its performance can be affected by several factors that must be carefully considered in design and implementation. Furthermore, the chemical environment in which bentonite is used can impact its effectiveness. Exposure to highly saline conditions can lead to ion exchange reactions that reduce its swelling

and adsorption capacities. Comprehensive assessments and site-specific studies are necessary to tailor bentonite-based barrier systems to the particular conditions they will encounter. Despite these challenges, bentonite remains a versatile and essential material in geoenvironmental applications, providing reliable long-term performance in the containment and isolation of hazardous waste.

In this chapter, the current studies on the mechanical performance of cutoff wall using bentonite-based materials are discussed. As well as the hydraulic performance and the influence of bentonite cement interactions. Given that few studies focused on the adsorption capacity of bentonite on gaseous mercury, the literature review summarized the normally-used adsorbents for mercury, and gaseous adsorbates for bentonite.

2.3 Enhancement on mechanical performance of SB

In efforts to enhance certain mechanical properties (e.g., strength and stiffness) of SB, cementitious binders have been utilized, such as Ordinary Portland Cement (OPC), which is the primary and dominant cement used in CB and SCB walls. To install the SCB cut-off walls, the trench cutting and re-mixing deep wall method (TRD) is one of the most technically acclaimed methods (Evans, 2007; Cheng et al., 2022). The construction process of TRD method is executed in two-phase (i.e. trench cutting and re-mixing) (Katsumi et al., 2008). In the first phase, the trenching machine excavates the soil and mixes with bentonite slurry for homogeneity. Herein, the sodium bentonite with superior swelling capacity and adsorption properties, is extensively applied (Gleason et al., 1997; Kamon et al., 2006; Malusis et al., 2009). In the second phase, the cementitious binder (OPC) blends with water and re-mixes with the soil-bentonite mixture.

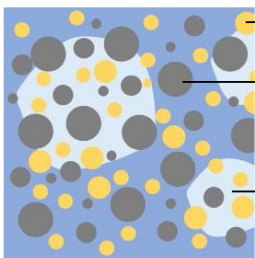
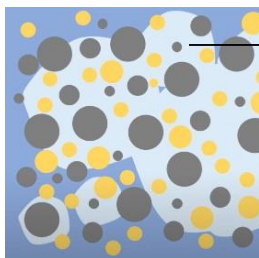
Primary hydration reaction	Secondary cementation reaction
$xCaO + ySiO_2 + zH_2O \rightarrow \frac{C_xS_yH_z}{C-S-H}$  → Bentonite → Cement → C-S-H	$2CaO + Al_2O_3 + 10H_2O \rightarrow \frac{3CaO \cdot Al_2O_3 \cdot 10H_2O}{C-A-H}$  → C-S-H & C-A-H ↓ Alkaline environment Fewer pore space

Fig. 2.3 The chemical interaction between cement and bentonite.

When the hydrated OPC is blended with hydrated bentonite, the calcium ions (Ca^{2+}) released from the OPC will replace the monovalent cations (e.g., sodium ions Na^+), leading to the flocculation of bentonite, as shown in Fig. 2.3 (Koch, 2022; Gaucher and Blanc 2006; Cuisinier et al., 2008). Furthermore, the silicate and aluminate sheets of the bentonite will be dissolute in the high-pH environment (OPC slurry: pH~12). As a result, the swelling capacity and water retention performance will be significantly undermined (Koch et al., 2002; Ahmed and El Naggar, 2016; Wu et al., 2019; Wei and Gencturk, 2019; Kaufhold et al., 2020).

The OPC, produces approximately 0.95 t/t OPC CO_2 with intensive consumption of raw material, accounts for the 7% of the total CO_2 emissions into the earth's atmosphere (Malhotra, 2002; Benhelal et al., 2013). Owing to the negative engineering and environmental impacts of OPC, supplementary cementitious materials (SCMs), including but not limited to ground granulated blast-furnace slag (GGBS) and fly ash (FA) have been studied and applied in land remediation and ground improvement projects (Shackelford and Jefferis, 2000; Wu et al., 2019; Du et al., 2020). However, alkaline activators (e.g. CaO , $\text{Ca}(\text{OH})_2$ and MgO) were necessary for the GGBS in the hydration process. Comparing with OPC, the use of GGBS combining with alkaline activators is not easy implemented in the two-phase TRD method. Moreover, the use of CaO and $\text{Ca}(\text{OH})_2$ will possibly alter the properties of bentonite, while MgO is still under studies considering the economic costs.

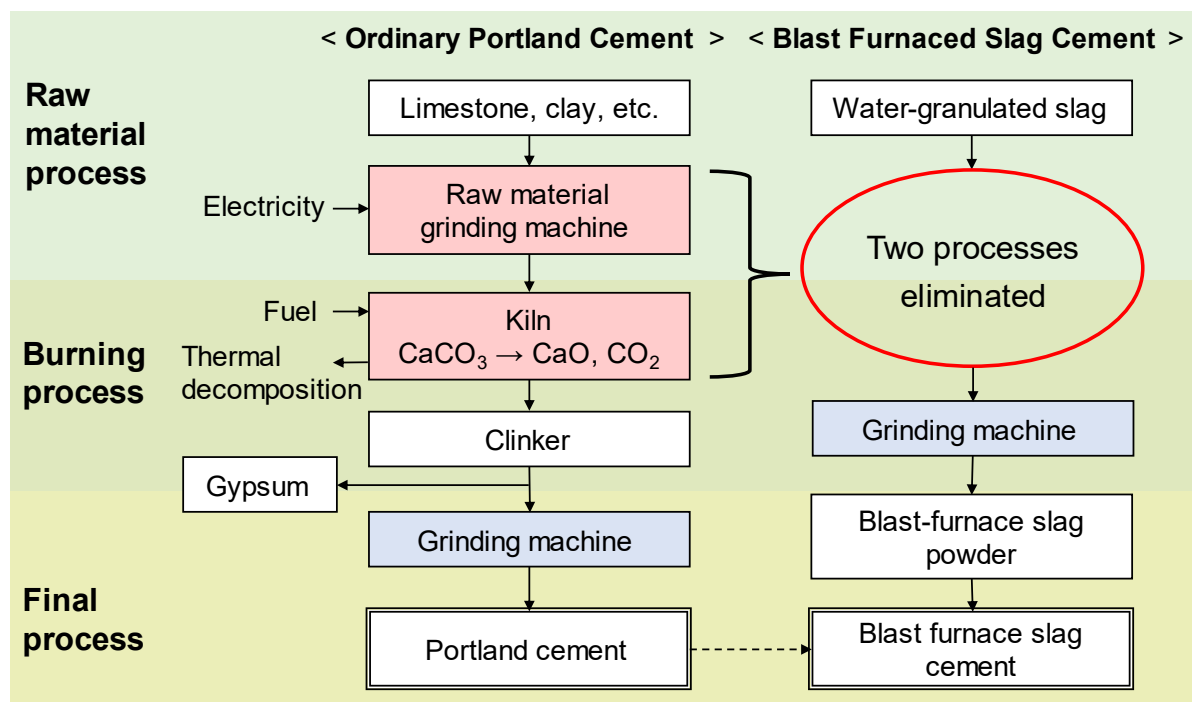


Fig. 2.4 Reduction of CO_2 emission by using slag cement instead of OPC.

Portland Blast-Furnace Slag Cement, as one type of eco-friendly material, manufactured through OPC blended with SCMs (GGBS), is often applied to improve the engineering properties of soft soils, as eco-friendly binder for reduction of CO₂ emission, as shown in Fig. 2.4. The GGBS utilized in slag cement, is capable of reducing CO₂ emissions by 22%, and the raw material and electricity has been conserved as well (Flower and Sanjayan, 2007). Also, based on the pH of the hydrated MgO-activated GGBS system ranging from 10.3 to 10.8, the slag cement blended in the second phase of TRD method will potentially decrease the pH environment, which benefit for properties retention of sodium bentonite (Wu et al., 2021). In order to maintain the workability of the mixtures in the second phase in TRD method, slag cement should be blended in low content to diminish the chemical interactions of flocculation and dissolution of sodium bentonite (Cheng et al., 2022).

Previous studies evaluated the strength properties and hydraulic performance of soil-cement-bentonite using OPC through unconfined compression test (UCS test) and hydraulic conductivity test (Ata et al, 2015; Huang et al., 2021). It is worthy to investigate the enhanced mechanical performance, the variations in deformability and the permeability in instances of relatively low content of slag cement blended in the soil-cement-bentonite mixture. High cement content, for instance, can significantly diminish the swelling and self-healing properties of bentonite. Although adding cement can improve the mechanical strength of the barrier, excessive amounts can lead to a denser material with reduced permeability and compromised swelling capacity. Optimal formulations need to strike a balance between sufficient mechanical strength and the preservation of bentonite properties.

2.4 Hydraulic performance of amended SB

Soil-bentonite mixtures exhibit excellent hydraulic performance due to their low permeability, essential for preventing contaminant migration in waste containment applications. Numerous studies have explored factors affecting the hydraulic conductivity k of soil-bentonite barriers. The hydration and swelling of bentonite, which benefit the hydraulic performance, has been shown in Fig. 2.5. The bentonite content promotes the hydration and swelling that significant reduction in hydraulic conductivity with increasing bentonite content and compaction energy in soil-bentonite slurry walls (Lunardini and Daniel 1993; Germaine and Kavazanjian 1995). However, the addition of binders like cement can significantly affect the hydraulic performance. For SCB backfill, the hydraulic conductivity commonly used in contaminant containment systems is suggested to be less than 1×10^{-8} m/s (Ryan and Day 2002). The hydraulic conductivity with increasing cement content, attributed to reduced pore size as cement hydrates

and fills voids within the mixture (Ghazanfari and Sheikhzadeh 2016). Despite enhancing mechanical properties, binders can impact self-healing capabilities. The self-healing properties in cement-amended mixtures will be improved due to the formation of cementitious bonds (Puppala et al., 2003). However, excessive cement hydration may lead to shrinkage or cracking, compromising self-healing. These findings highlight the intricate relationship between binder addition, hydraulic performance, and self-healing in soil-bentonite mixtures, necessitating careful consideration in engineering design and application.

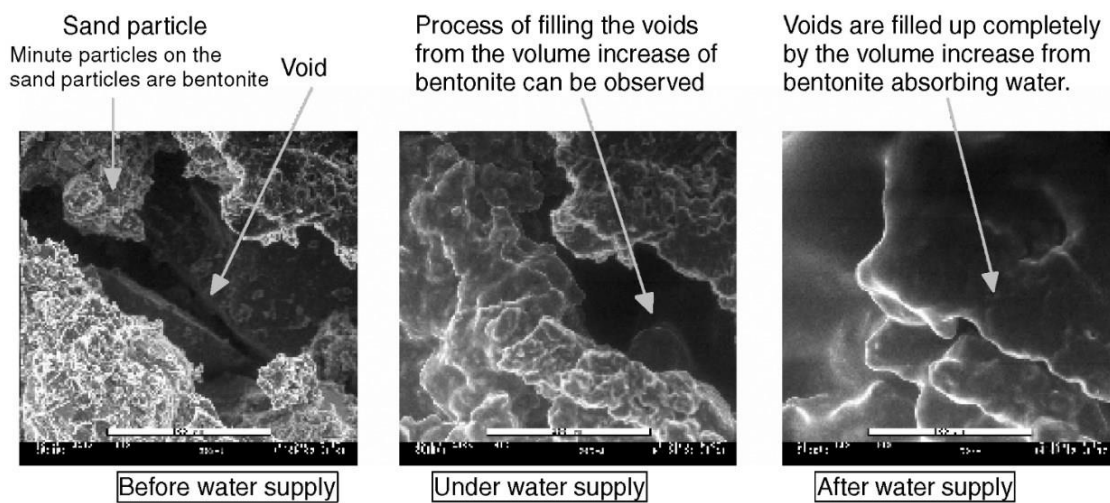


Fig. 2.5 Swelling behavior of bentonite in sand-bentonite (Komine and Ogata, 2003).

The addition of binders like fly ash, cement, and slag cement aims to enhance the mechanical properties of soil-bentonite mixtures. Recent studies have explored their effects on hydraulic performance and self-healing. It is investigated that the impact of cement on soil-bentonite hydraulic conductivity presented a decrease with increasing cement content (Hesami and Nematollahi 2018; Hajiannia and Rashidian 2020). The mechanical and hydraulic behavior of soil-bentonite-cement mixtures, emphasizing the importance of optimizing binder content for desired performance.

While binders improve mechanical strength, the influence of bentonite or other clay minerals on self-healing is crucial. It is noted that the use of metakaolin and calcined clays as pozzolans for concrete highlight the enhancement of self-healing capabilities (Sabir et al., 2001). However, the effectiveness of self-healing may be compromised if binder hydration leads to shrinkage or cracking (Edil et al., 1999). Therefore, comprehensive evaluation of binder effects on soil-bentonite performance, considering both mechanical and hydraulic aspects is necessary, to ensure the integrity of vertical cutoff walls. Table 2.1 compared the different properties of

SB, CB and slag-SB mixtures (Opdyke and Evans 2005; Wu et al., 2019).

Table 2.1 Different properties of SB, CB and slag-SB mixtures

Barrier composition	SB	CB	Slag-SB
Hydraulic conductivity	1×10^{-9} m/s	1×10^{-9} m/s (90 days)	$<1 \times 10^{-9}$ m/s (90 days)
Solids content	~70%	~20%	~70%
UCS	~0	~100 kPa @ 28days	~200 kPa @ 28days
Strain to failure	Plastic	Brittle	Rely on slag content
Time dependency	Consolidation: Rapidly ~within a few days	Hydration reactions: over 90 days	Unknown
Construction stages	Two phase	One phase	Two phase

2.5 Gaseous mercury adsorption and utilization of bentonite

Mercury and mercury components, as toxics from natural releases and artificial emissions, systematically cycles in the environment and finally ends up in the food chain. Once human bodies poisoned, the mercury target at brain and kidney in priority, and easily impose neurological dysfunction or even immune dysfunction (Park et al., 2012; Berlin et al., 2007). For human reasons, the mercury emitted or released from improper handling, collection, transportation or disposal of mercury containing wastes like household batteries, lamps, mirror coatings and thermometers (Tim et al., 2014; Zhu et al., 2013).

Researchers investigated the capability of gas adsorption in pure gas phase or flue gas composition. Modified bentonites were applied as the adsorbent for CO₂, with the adsorption capacity from 0.5 mmol/g to 3.2 mmol/g (Elkhalifah et al., 2013, 2015). In the test of H₂ and H₂S gas adsorption using cation-exchanged bentonite, the equilibrium adsorption properties range from 0.2 mmol/g to 1.8 mmol/g (Alver et al., 2017; Stepova et al., 2009).

The mercury Hg, as volatile metal at room temperature, presents two forms of gaseous and liquid components (Hg, HgCl₂, HgS, HgO, etc.) (He et al., 2018). Mercuric chloride liquids are the aqueous phase applied in research, and activated carbon is one of the commonly used adsorbents (Asasian et al., 2015; Chiarle et al., 2000). However, the treated mercury solids in specially constructed mercury waste landfill are HgS compounds, thus mercury releases only

be in gaseous phase as elemental mercury Hg(0). In most circumstances, mercury gas represents as by-product or co-existing components. The activated carbon, fly ash, mineral-based sorbents, mineral chalcogenides and nano-ceramic material were utilized for mercury gas adsorption in industrial fuel gas, coal combustion flue gas and nonferrous smelting flue gas (Kang et al., 2020; Zheng et al., 2021). In this research, the adsorption amount ranges from 0.5 to 5 mg/g at over 100°C. Aside from fuel gas, the mechanism of mercury gas adsorption in air at normal temperature has not yet been studied.

The constant volume method is one of three categories for gas adsorption test, while the other two are dynamic adsorption column method and gravimetric method, respectively (Kim et al., 2006). Constant volume method was applied in one special research to investigate the adsorption performance of silica sand, decomposed granite soil, mordenite, and calcium bentonite on gaseous mercury and air mixture at 25.0°C (Ishimori et al., 2020). The adsorption capacity of dry soil specimens was estimated, as presented in Table 2.2. It is indicated that bentonite presents the best adsorption capacity comparing with other types of soils. However, moisture soil specimens (i.e. different water content) have not been studied.

Table 2.2 Distribution coefficients and equilibrium durations for soil specimens (Ishimori et al., 2020).

Unit (mL/g)	Silica sand	Decomposed granite soil	Mordenite	Calcium bentonite
After 24 h	4.40	12.8	7.09	14.9
After 2 days	8.59	26.5	14.4	31.3
After 4 days	16.3	56.1	29.7	68.3
After 8 days	28.9	121	62.5	159
Equilibrium duration	48.7 days	54.3 days	80.9 days	55.3 days

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Chapter 3

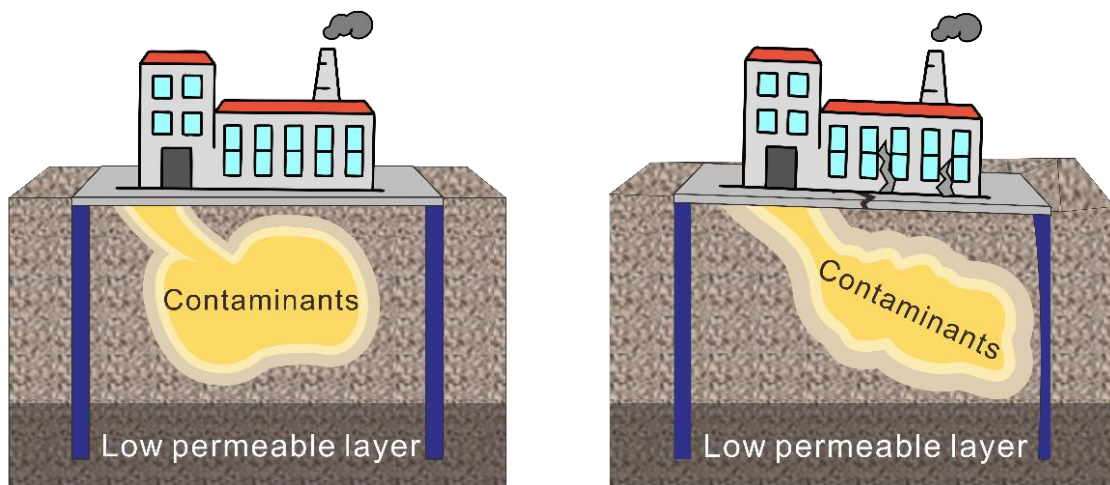
Mechanical performance of SB mixtures amended with slag cement

3.1 General remarks

SB mixtures exhibit relatively low shear strength (i.e. inadequate strength properties), as a result, the effectiveness of SB vertical cutoff wall will be influenced by foundation loads (static loads) or in situations of dynamic loads, as shown in Fig. 3.1 (Opdyke and Evans, 2005).

In efforts to enhance certain mechanical properties (e.g., strength and stiffness) of SB, cementitious binders have been utilized. Normally, the strength and stiffness properties are obtained from unconfined compression strength test, direct shear test, or triaxial compression test (CD/CU/UU), depending on the soil types, stress conditions of the soil and application scenario. Supplementary cementitious materials (SCMs), including but not limited to Ordinary Portland Cement (OPC), ground granulated blast-furnace slag (GGBS), Slag cement, fly ash (FA) and lime, etc.

From a sustainability perspective, slag cement is designated as the blinder. Meanwhile, in order to maintain the workability of the mixtures and decrease the pH in the second phase in TRD method, slag cement usage in low content is implemented, also diminish the chemical interactions between sodium bentonite and slag cement (Cheng et al., 2022). For the emulation of various realistic stress scenarios, the triaxial compression test is prioritized in this research.



(a) Vertical cutoff wall.

(b) Inadequate strength properties.

Fig. 3.1 Schematic view of vertical cutoff wall in contaminated site.

In this chapter, a systematic blending procedure of SB and SB-C is implemented, and a series of Consolidated-Undrained (CU) triaxial compression tests on SB and SB-C are conducted to investigate the variations in the mechanical performance (e.g. shear strength properties, typical failure modes, effective stress path and strength envelope, Young's modulus E_0 and Secant modulus E_{50}), aiming to explore the effect of slag cement amendment on the mechanical performance and the applicability of slag cement in low content for the construction of SCB walls using two-phase TRD method.

3.2 Materials and methodology

3.2.1 Properties of the soil and binders

In this study, the base soil was decomposed granite soil with particle dry density of 2.73 g/cm^3 , which is widely distributed in Japan. The grain size distribution curve of the decomposed granite soil by JIS A1204 (2020) has been presented in Fig. 3.2. Powder bentonite and slag cement are two binders in this chapter.

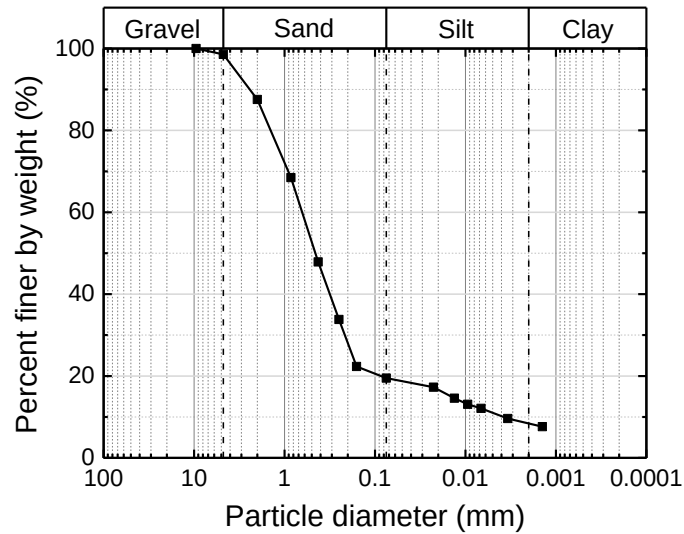


Fig. 3.2 Grain size distribution curve of decomposed granite soil.

The powder bentonite used in this study was commercial sodium bentonite (Kunigel-V1) from Kunimine Industries Co., Ltd., Japan. The free swelling index of this bentonite was $\sim 23 \text{ mL/2g}$, and the passing percent for grain size $> 0.075 \text{ mm}$ was 90%. The fundamental properties of this sodium bentonite were evaluated on the same batch of Kunigel-V1 and summarized in Table 3.1, although the montmorillonite content was reported by a previous study (Wang et al., 2022). Additionally, the main accessory minerals of this bentonite are Quartz, Feldspar,

Plagioclase and Calcite (Wang et al., 2020).

Table 3.1 Fundamental properties of sodium-type commercial bentonite (Kunigel-V1).

Index	Values	Testing method
Particle density (Mg/m ³)	2.76	JIS A1202 (2020)
Liquid limit (%)	485.7	JIS A1205 (2020)
Plastic limit (%)	37.2	JIS A1205 (2020)
Plasticity index	448.5	—
Passing percent for grain size > 0.075 mm (%)	90	JIS A1204 (2020)
Montmorillonite content (%)*	53	JIS Z2451 (2019)
Cation exchange capacity (meq/100g)	98.72	JGS 0261 (2009)
Na ⁺ exchange capacity (meq/100g)	62.38	—
Ca ²⁺ exchange capacity (meq/100g)	33.34	—
K ⁺ exchange capacity (meq/100g)	0.45	—
Mg ²⁺ exchange capacity (meq/100g)	2.55	—

* The Montmorillonite content was reported by Wang et al. (2022) as according to JIS Z2451 (2019) using methylene blue adsorption on bentonite.

The slag cement is commercially manufactured Portland Blast-Furnace Slag Cement (Type B, Sumitomo Osaka Cement Co., Ltd.) with the particle density of 3.04 g/cm³ and specific surface area of 3.900 cm²/g. The Ig. loss of the slag cement is 1.31%, and the chemical composition of SiO₂, CaO, MgO and SO₃ are 25.37%, 54.92%, 3.38% and 1.88%, respectively. The chemical composition of the sodium bentonite (Kunigel-V1) is provided from the Kunimine Industries Co., Ltd. The chemical compositions of these two blinders have been summarized in Table 3.2. As a comparison, the chemical compositions of OPC and lime (e.g. Limestone powder) are also illustrated (Kameshima et al., 2012).

Table 3.2 Chemical compositions of sodium bentonite (Kunigel-V1) and slag cement.

Unit (%)	ig.loss	SiO ₂	CaO	MgO	SO ₃	Al ₂ O ₃	Fe ₂ O ₃	Na ₂ O	K ₂ O	TiO ₂
Bentonite	5.3	69.4	2.1	2.2	-	15.6	2.0	2.0	0.3	0.1
Cement	1.3	25.4	54.9	3.4	1.9	-	-	-	-	-
OPC	2.6	20.0	64.3	1.6	2.0	-	-	-	-	-
Lime	41.5	-	54.6	1.0	0.0	-	-	-	-	-

3.2.2 Specimen preparation procedure

The flow of SB and SB-C mixture preparation have been illustrated in Fig. 3.3. First, the bentonite slurry (5% bentonite by weight basis) was prepared by mixing powder bentonite with distilled water for 30 mins using a slurry mixer and hydrated at a constant temperature of 20 °C for 24 h. This slurry content was determined to achieve 150 mm flow value of soil-slurry mixture by JIS R5201 (2015). Hereafter, the base soil (decomposed granite soil) was sieved through a 4.75 mm sieve and blended with distilled water and slurry using a soil mixer for 5 mins to homogenize the mixture, which resemble the condition with the first construction process of the TRD method (Katsumi et al., 2008; Takai et al., 2016). Then, the bentonite powder (with proportions of 97.5 kg/m³) was added in two batches and blended using soil mixer for 5 mins respectively. At this step, the SB mixture has been prepared. To achieve amended SB mixtures (i.e., SB-C), additional slag cement (with proportions of 25, 50, 75 or 100 kg/m³) was added in one batch and blended for 10 mins.

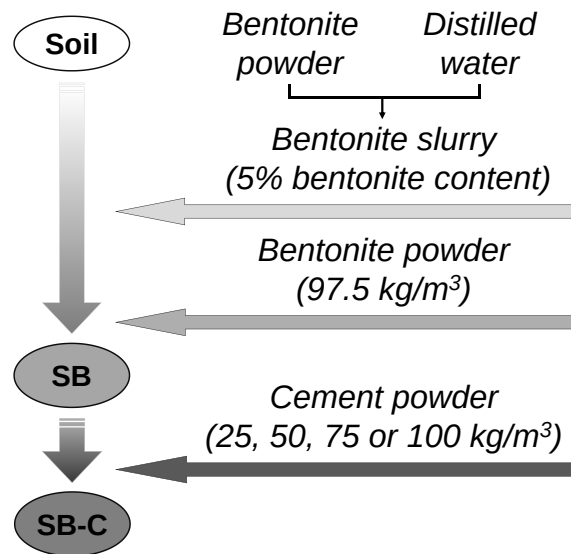


Fig. 3.3 Flow of SB and SB-C mixture preparation.

To ensure the homogeneity, the specimens for triaxial compression test were prepared with the target dry density of 1.3 g/cm³. After blending process, the SB-C mixtures were scooped into plastic cylinder in layers (with a height of 100 mm and a diameter of 50 mm) and tapped against a hard surface to extract air bubbles (Luis et al., 2019). The specimens were then sealed immediately with plastic wrap and aluminum foil to minimize possible moisture change and cured at 20 °C for 7 D and 28 D, respectively. The schematic view of the sealed specimen has been illustrated in Fig. 3.4.

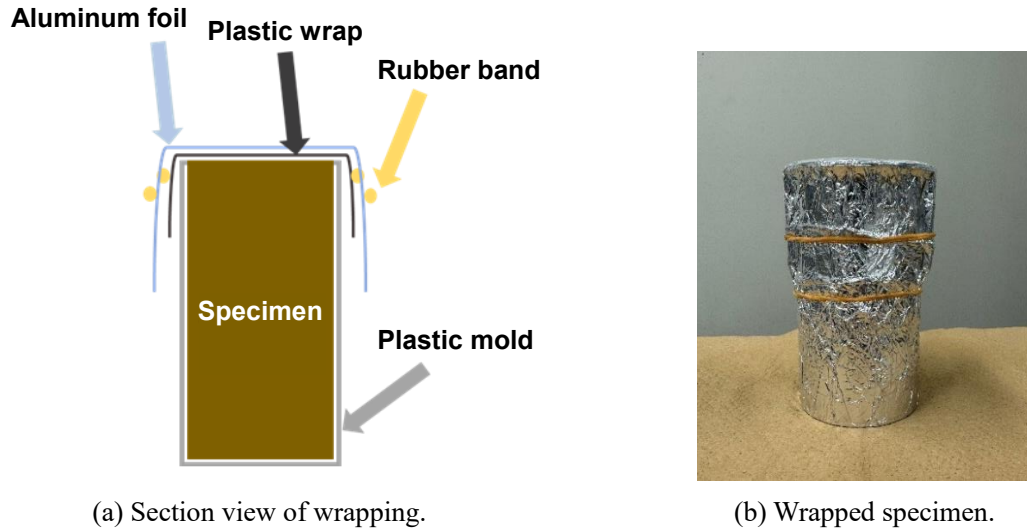


Fig. 3.4 Wrapping structure of the specimen.

The testing scheme in triaxial compression test has been summarized in Table 3.3. Herein, the specimens are named as SB-C(cement concentration)-(curing time). For example, the SB-C25-7, means the soil bentonite mixture amended with 25 kg/m³ slag cement and the curing time is 7 days.

Table 3.3 Testing scheme of triaxial compression test.

	Bentonite slurry (L/m ³)	Bentonite powder (kg/m ³)	Slag cement (kg/m ³)	Curing time (D)	Confining pressure (kPa)
SB-C0-0 (SB)			0	0	
SB-C25-7			25	7	
SB-C25-28				28	
SB-C50-7	350	97.5	50	7	40, 80, 160
SB-C50-28				28	
SB-C75-7			75	7	
SB-C75-28				28	

To confirm the dry density of each specimen, the weight of each specimen was measured before wrapping. Also, the residual of the mixtures was separated into three batches to measure the water content, as according to JIS A1203 (2020). Through calculating in this way, the dry density of each specimen before curing could be confirmed. The calculated dry density of specimens before curing has been summarized in Table 3.4.

Table 3.4 The dry density of specimens before curing.

SB-C0-0						
Confining pressure (kPa)	40	80	160			
Dry density (g/cm ³)	1.28	1.28	1.30			
SB-C25-7			SB-C25-28			
Confining pressure (kPa)	40	80	160	40	80	160
Dry density (g/cm ³)	1.29	1.30	1.29	1.30	1.31	1.29
SB-C50-7			SB-C50-28			
Confining pressure (kPa)	40	80	160	40	80	160
Dry density (g/cm ³)	1.31	1.30	1.31	1.31	1.30	1.31
SB-C75-7			SB-C75-28			
Confining pressure (kPa)	40	80	160	40	80	160
Dry density (g/cm ³)	1.31	1.30	1.29	1.31	1.30	1.30

3.2.3 Triaxial compression test

To evaluate the mechanical performance of SB and SB-C specimens, a series of isotropic consolidated-undrained (CU) triaxial compression test were conducted as per JGS 0522 (2020), and the test procedure was aligned with ASTM D4767-11 (2020).

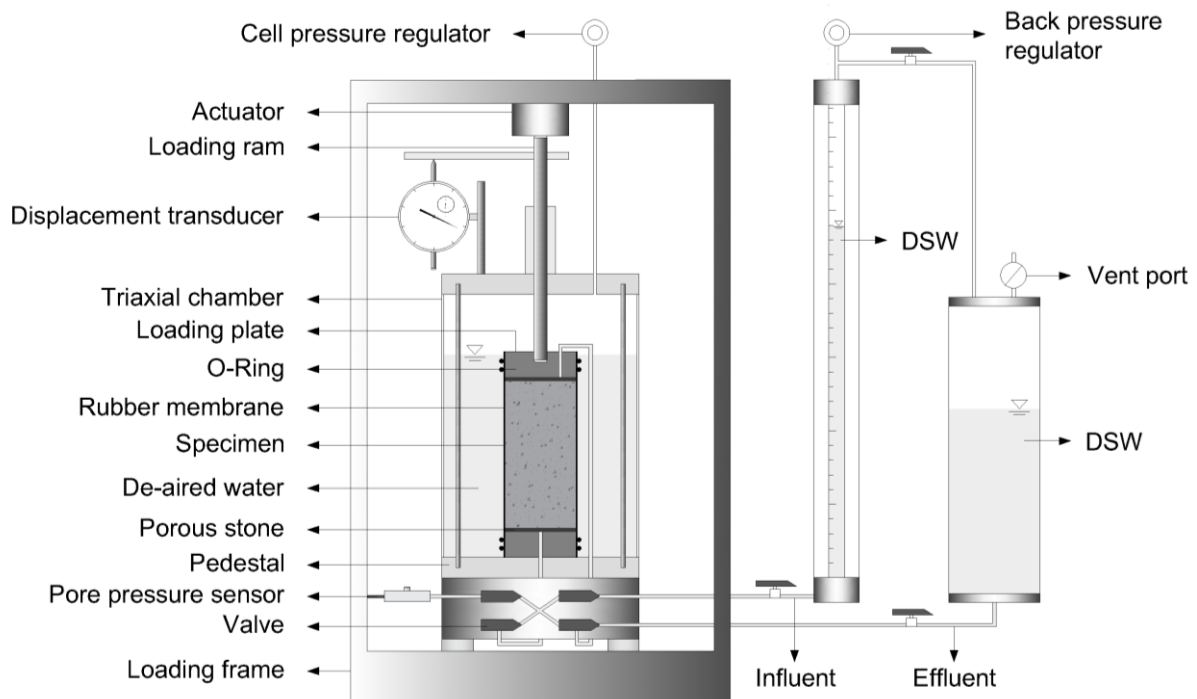
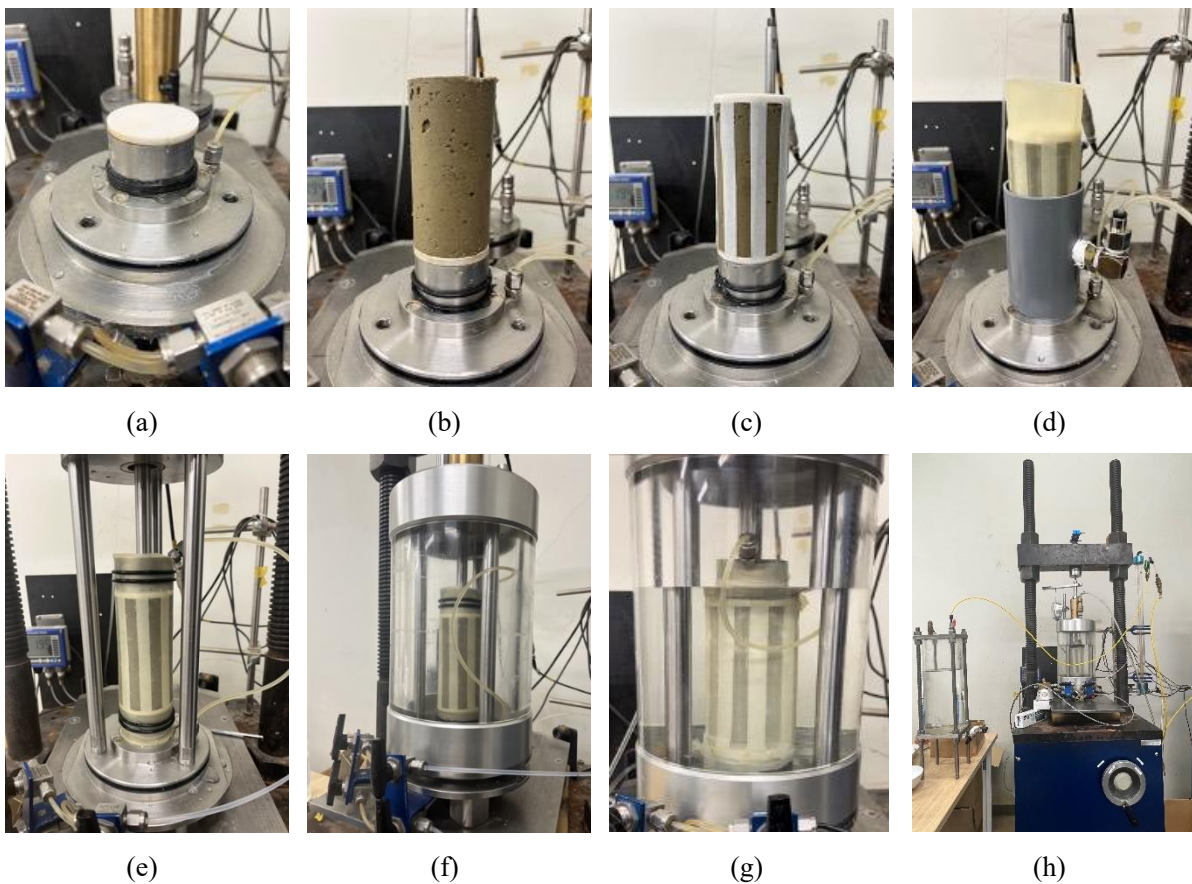


Fig. 3.5 Schematic diagram of triaxial compression test apparatus.

A schematic diagram of the triaxial compression test apparatus is presented in Fig. 3.5. In this triaxial compression test apparatus, the main components include a loading frame, a triaxial chamber, cell pressure and back pressure regulators, and influent/effluent vertical standpipe.

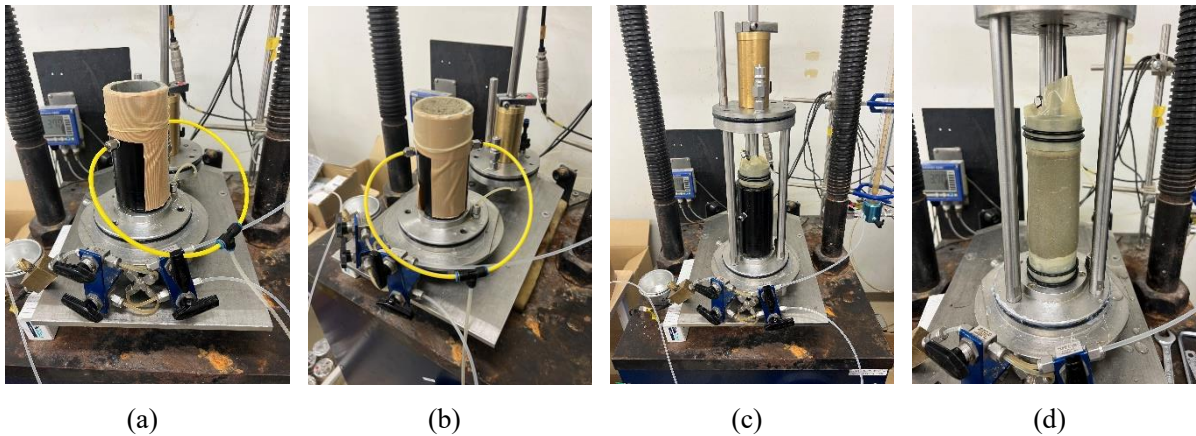
The triaxial compression test commenced with the installation of the specimen in the triaxial chamber (triaxial cell). Herein, the installation process of SB and SB-C specimens were different. For SB-C specimens, after curing for 7 D and 28 D, the specimen was disassembled from the plastic cylinder, and the weight and height were measured for the initial state estimation. After the measurement, the specimen was sealed in water-tight, rubber membrane with O-ring. The detailed procedure has been illustrated in Fig. 3.6.



- * (a) Pre-saturation of influent and effluent pipes and installation of porous stone.
- (b) Installation of saturated filter paper and disassembled specimen.
- (c) Installation of saturated filter paper for optimization of consolidation process.
- (d) Installation of latex membrane through air extraction.
- (e) Installation of O-Ring from two sides on the loading plate.
- (f) Installation of the triaxial chamber and displacement transducer.
- (g) Injection of the de-aired tap water into the triaxial chamber.
- (h) Connection of air pipes with cell pressure and back pressure regulator.

Fig. 3.6 Installation procedure of SB-C specimens.

For SB specimens, the detailed procedure of SB specimen installation has been shown in Fig. 3.7. After the meticulous blending process, the soil-bentonite mixture was carefully scooped into a specifically crafted metal cylinder (with an inner height of 100 mm and a diameter of 50 mm), which was affixed onto the bottom loading plate. Inside the metal cylinder, a latex membrane was initially enveloped, and the saturated porous stone was also installed onto the bottom loading plate. During the process of SB mixture filling into the metal cylinder, the metal cylinder was connected with air pipe from two side with continuous air pumping to maintain the latex membrane tightly contacting with the inner surface. After filling in the SB mixture, the metal cylinder was then disassembled into two parts from opposing sides, and the SB specimen was sealed with O-Ring and equipped as the procedure employed for the SB-C specimens, following Fig. 3.6 (f) to Fig. 3.6(h).



- * (a) Installation of saturated porous stone and installation of specifically crafted metal cylinder onto the loading plate, with latex membrane was initially enveloped.
- (b) Pouring the SB mixture into the container with designated weight.
- (c) Installation of O-Ring on the top loading plate.
- (d) Disassembling of the metal cylinder and installation of O-Ring on the bottom loading plate.

Fig. 3.7 Installation procedure of SB specimens.

The filter paper for optimization of consolidation process was exclusively used for SB-C specimens, and also for the equalization of effective stresses within the specimen during compression process. As show in Fig. 3.6(e), the filter paper was attached to the surround surface on the specimen. As expectation to shorten the consolidation process, the efficiency is strongly dependent on the coverage area ratio of the filter paper on the circumferential surface of the specimen and the consolidation state (Oda and Mitachi 1990; Leroueil 1988). In this chapter, the coverage area ratio of the filter paper on the circumferential surface of the specimen was 50%, as illustrated in Fig. 3.8. In normally consolidated range, the effect of the filter drains

on shear strength is negligible (Leroueil 1988). Thus, in this chapter, an assumption could be reached that no influence of this filter paper on the strength properties within normal consolidation state.

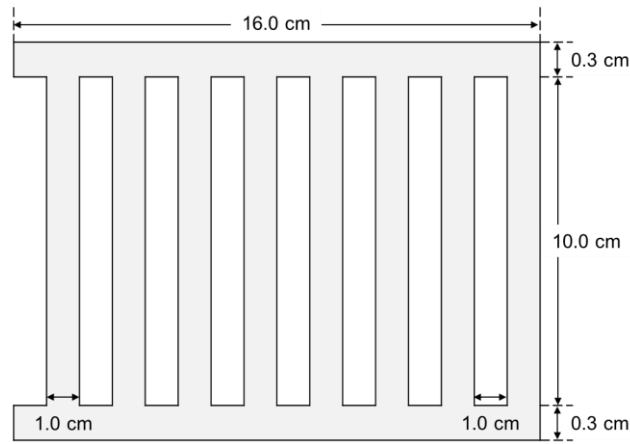


Fig. 3.8 The filter paper for optimization of consolidation process.

Following the installation of the specimens, the cell was filled with de-aired tap water, as shown in Fig. 3.6(g). To ensure the full saturation during the triaxial test, 300 kPa back pressure was maintained through the saturation, consolidation and compression process. In the saturation process, the specimens were subjected to a confining pressure of 10 kPa (i.e., a difference of 10 kPa between cell pressure and back pressure), so that unintended swelling of the specimen resulting from a high back pressure or consolidation of the specimen due to high cell pressure can be prevented (Kayadelen et al., 2007).

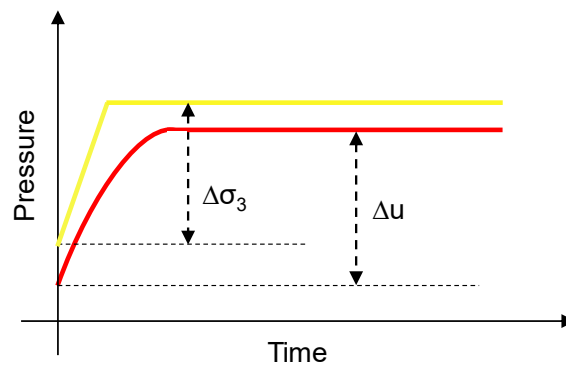


Fig. 3.9 The variation of pore pressure and confining pressure in B value verification.

The total saturation time ranging between 12 to 24 h, which depending on the level of saturation estimated through the pore pressure coefficient B value, which is a coefficient for the

verification of saturation depending on the relationship between pore pressure and confining pressure. Herein, the B value is defined in Equation (3-1),

$$B = \frac{\Delta u}{\Delta \sigma_c'} \quad (3-1)$$

where Δu is the increase in the pore pressure due to the increment in the confining pressure $\Delta \sigma_c'$ of the specimen.

Normally, the B value for cement stabilized or treated soil mass have a B value ranged from 0.7 to 0.9, while for clayey soil B value higher than 0.95 implies saturation (Amini and Hamidi., 2014; Quiroga et al., 2017; Luis et al., 2019; Lu et al., 2022). In this paper, the B value was exceeding 0.95. Occasionally the B value was between 0.90 to 0.95, especially for SB-C50 and SB-C75 specimens under high confining pressure (i.e. 160 kPa) and longer curing time (i.e. 28 days), which was attributed to the relatively high rigidity of the skeleton rather than the presence of air within the system (Wissa and Ladd., 1964). As estimated, the B value of SB and SB-C specimens meet the required conditions, and all of the B value are summarized in Table 3.5.

Table 3.5 B value of SB and SB-C specimens.

SB-C0-0						
Confining pressure (kPa)	40	80	160			
B value (Up)	1.10	1.04	0.99			
B value (Down)	1.09	1.02	0.98			
SB-C25-7			SB-C25-28			
Confining pressure (kPa)	40	80	160	40	80	160
B value (Up)	0.95	0.95	0.96	0.95	0.92	0.94
B value (Down)	0.95	0.95	0.98	0.95	0.90	0.91
SB-C50-7			SB-C50-28			
Confining pressure (kPa)	40	80	160	40	80	160
B value (Up)	0.95	0.94	0.90	0.90	0.90	0.91
B value (Down)	0.93	0.93	0.91	0.91	0.90	0.94
SB-C75-7			SB-C75-28			
Confining pressure (kPa)	40	80	160	40	80	160
B value (Up)	0.94	0.95	0.92	0.90	0.91	0.90
B value (Down)	0.93	0.96	0.94	0.91	0.92	0.92

Subsequent to the completion of saturation, the saturated specimens were consolidated under different isotropic confining pressures (i.e., $\sigma_c = 40, 80$ and 160 kPa), which considered the stress distribution in SB wall through the depth under overburden pressure (i.e. static loads). As shown in Fig. 3.10(a), the total stress in the SB wall was proved to be lower than the overburden pressures from depth of 0 m to 14 m. As a matter of fact, the effective stress distribution through the depth is also lower than the overburden pressure, which can be attributed to the arching effect (Evans et al. 1995). In this chapter, the confining pressure of $40, 80$ and 160 kPa were implemented as a factor on the mechanical performance of SB and SB-C specimens to simulate the different stress conditions through the depth of the cutoff wall, as well to evaluate the effective strength parameters (c' and ϕ').

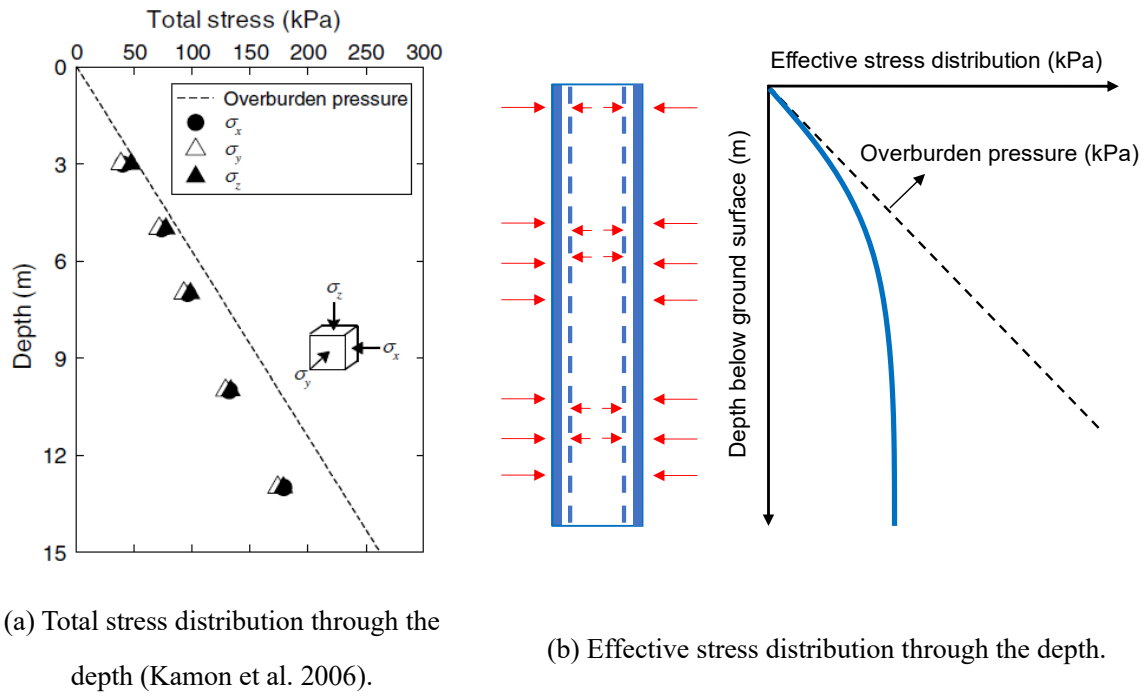


Fig. 3.10 The stress distribution in vertical cutoff wall vs overburden pressure.

The designated confining pressure were accomplished by increasing the cell pressure at a constant rate to the desired value with the back pressure being constant, and the confining pressure conditions in the consolidation and compression process were sustained consistency. After the specimens have been saturated, a series of isotropic consolidation were performed by referencing the JGS 0522 (2020).

The termination of the consolidation process (i.e. consolidation time) was determined by the 3t-method. As illustrated in Fig. 3.11, the ΔV_t - $\log t$ curve was drawn automatically from the computer. The steepest tangent of the ΔV_t - $\log t$ curve was drawn, and a straight line parallel to

the steepest tangent of the curve was drawn at the $3t$ point in time (i.e. the “ $3t$ line”). Herein, the point at which the ΔV_t - $\log t$ curve touches the $3t$ line is defined as the point at which consolidation terminates. As estimated, the consolidation process took from hours to a few days. After consolidation, the hydraulic conductivity of each specimen was evaluated as according to ASTM D5084-03 (2011).

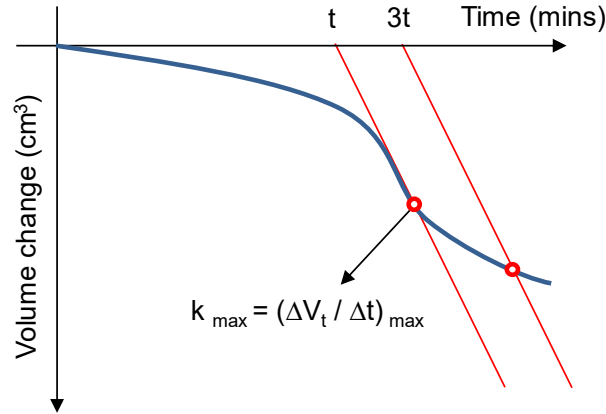


Fig. 3.11 The criterion for termination of consolidation process ($3t$ -method).

The hydraulic conductivity of the specimens was estimated after consolidation. Finally, the specimens were subjected to axial compression at an axial strain rate of 0.05 mm/min (i.e., 3% strain /h) until reaching approximately 15% axial strain, as long as pore water was not allowed to flow in or out of the specimen. In the compression process, the deviator stress, axial strain, pore pressure and cell pressure were recorded through the computer automatically.

3.3 Results and discussion

The Cam clay model describes the soil behavior in elastic-plastic model, which goes as least some way towards incorporating a rather more realistic description of the effective stress changes that occur in constant volume compression. Given that the triaxial compression test is based on axially symmetric stress conditions, some assumptions based on isotropic CU triaxial compression test are necessary. Herein, the response of the soil specimen depends on the mean effective stress p' and a general deviator stress q . The q and p' are determined using equation (3-2) and (3-3) (Wood 1990),

$$q = \frac{1}{\sqrt{2}} \sqrt{(\sigma_1' - \sigma_2')^2 + (\sigma_2' - \sigma_3')^2 + (\sigma_1' - \sigma_3')^2} \quad (3-2)$$

$$p' = \frac{\sigma_1' + \sigma_2' + \sigma_3'}{3} \quad (3-3)$$

where σ_1' is the effective major stress, while σ_2' and σ_3' are the effective minor principal stress, provided from the cell pressure, where $\sigma_2' = \sigma_3'$. Therefore, in this chapter, the equation (3-2) and (3-3) can be rewritten in terms of $\sigma_2' = \sigma_3'$ to (3-4) and (3-5),

$$q = \sigma_1' - \sigma_3' = \sigma_1 - \sigma_3 \quad (3-4)$$

$$p' = \frac{\sigma_1' + 2\sigma_3'}{3} \quad (3-5)$$

3.3.1 Stress-strain characteristics

As referring to JGS 0522 (2020) and ASTM D4767-11 (2020), the failure of specimen is corresponding to the maximum deviator stress ($q_{\max}$) or deviator stress at 15% axial strain, whichever is the first obtained during the test. The stress-strain relationship of SB and SB-C specimens can be classified as strain-softening curves (a line in Fig. 3.12) and strain-hardening curves (b line in Fig. 3.12).

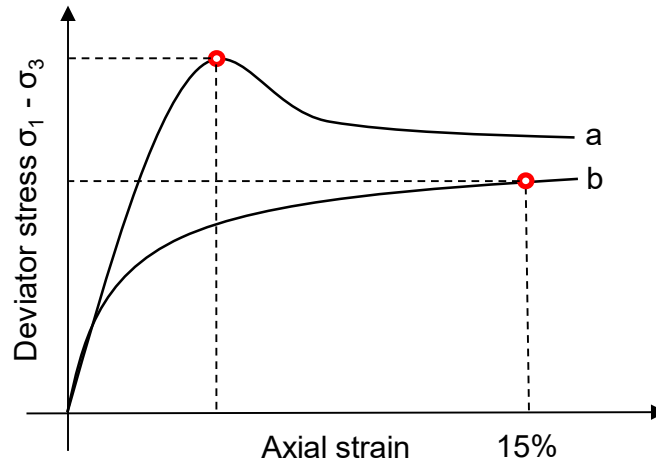


Fig. 3.12 Classification of deviator stress and axial strain relationship.

Strain-softening occurs when a soil undergoes increasing deformation, causing a reduction in its shear strength and stiffness. This phenomenon is often observed in cohesive soils such as clays, where continued deformation leads to particle reorientation, dilation, and the breakdown

of inter-particle forces, resulting in a decrease in strength. On the contrary, strain-hardening usually describes the behaviour of certain granular soils, with the retention of strength and stiffness, which is attributed to particle rearrangement, leading to an increase in inter-particle friction and effective stress transfer. This phenomenon is crucial in understanding the behaviour of soil under dynamic loading, such as during earthquakes or construction activities. The stress-strain relationship of SB and SB-C under different confining pressure (i.e. 40, 80, 160 kPa) has been presented in Fig. 3.13 and Fig. 3.14, respectively.

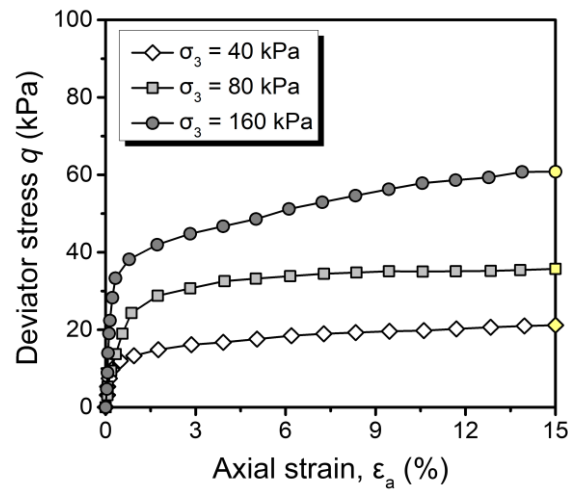
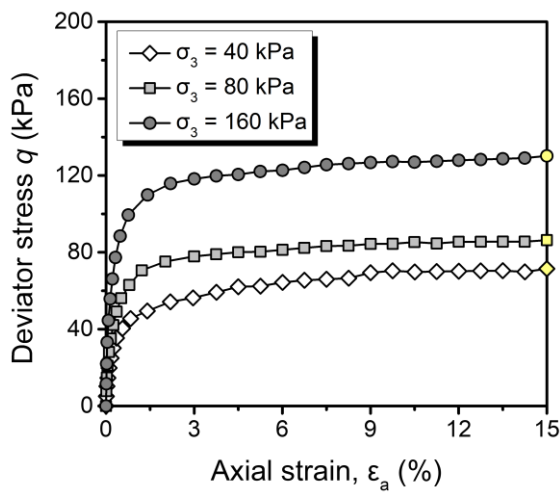
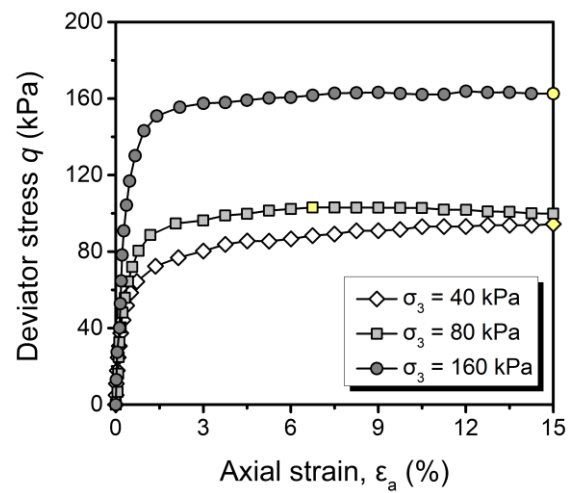


Fig. 3.13 Variation of deviator stress with axial strain for SB-C0-0.



(a) SB-C25-7



(b) SB-C25-28

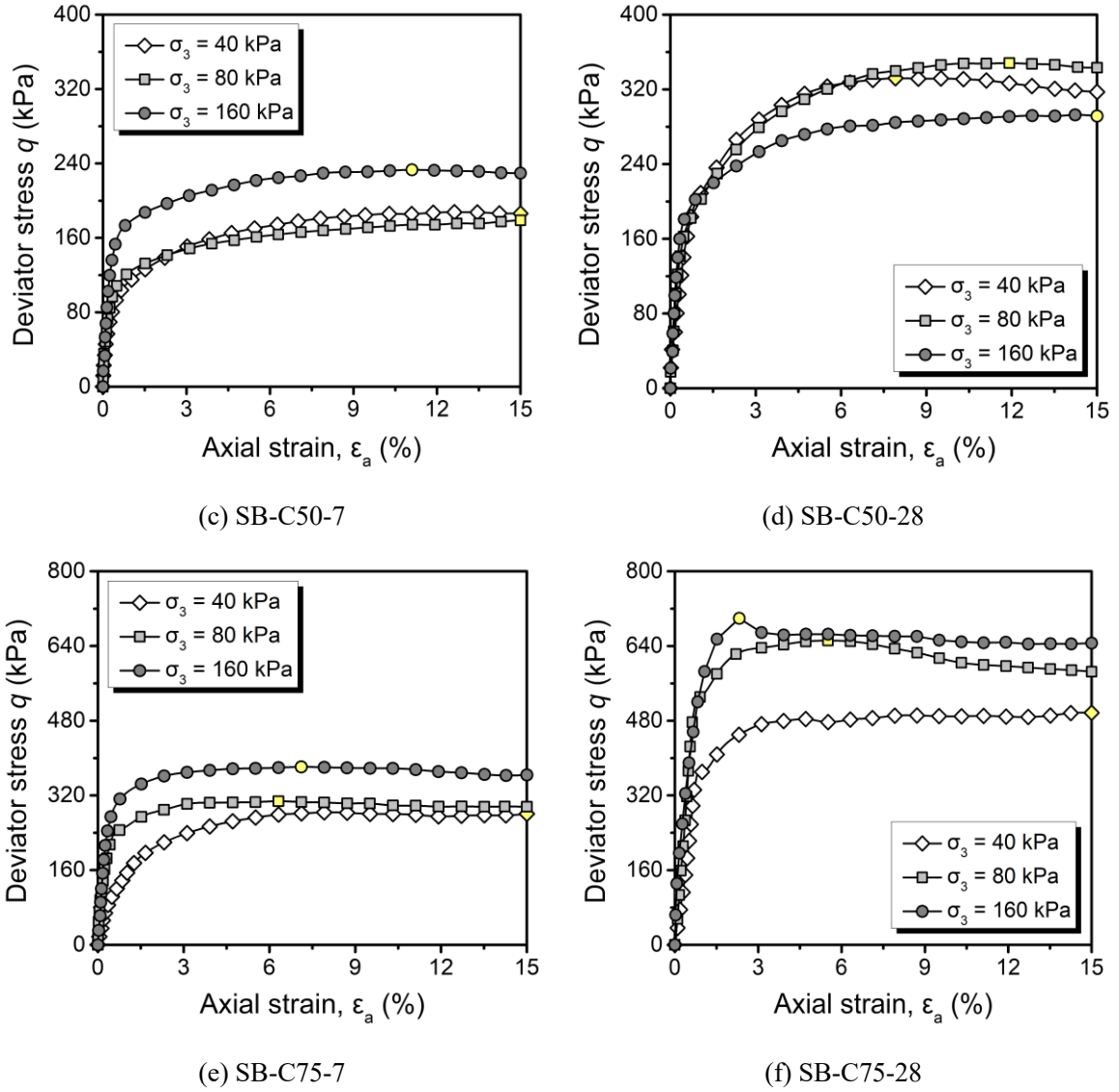


Fig. 3.14 Variation of deviator stress with axial strain for SB.

In general, the stress-strain relationship (q - ϵ_a curve) at higher confining pressure lies above on that at lower confining pressure attributed to the effect of consolidation, except for the SB-C50 specimens. Moreover, owing to the influence of the hydration and cementation of slag cement, the q - ϵ_a curve of 28 D specimen lies above on 7 D specimen in each mix proportion. In Fig. 3.13, the SB shows strain-hardening behavior, especially under $\sigma_3 = 160$ kPa, with the highest deviator stress at 15% axial strain. The SB-C25 shows similar strain-hardening behavior, while only SB-C25-28 ($\sigma_3 = 80$ kPa) presents slight strain-softening behavior. For SB-C75 specimens, the specimens under $\sigma_3 = 40$ kPa presents strain-hardening behavior, while it shows strain-softening behavior under higher confining pressure. However, under 80 and 160 kPa confining pressure, the deviator stress for SB-C75-7 did not significantly decrease from the peak to residual q but did decrease significantly for SB-C75-28. The most typical stress-strain

characteristics are in SB-C50, as the stress-strain relationship represents the characteristics of both strain-hardening and strain-softening behavior, as a balance between SB-C25 and SB-C75 specimens.

3.3.2 Failure modes

To further estimate the mechanical performance of specimens, the failure characteristics of the specimens (i.e. rupture planes within the specimen) after triaxial compression test have been summarized in Table 3.6. And four typical failure modes (i.e. Ductile failure, Slight planar failure, Planar failure, Split failure) of SB and SB-C specimens, have been illustrated in Fig. 3.15. Fig. 3.15(a) shows a ductile failure mode for all SB-C0-0 and SB-C25-7 specimens, and SB-C25-28 under $\sigma_3 = 40$ kPa, which is comparable to the strain hardening stress-strain curves in Fig. 3.13, Fig. 3.14(a) and Fig. 3.14(b). The slight planar failure is the integrate of ductile failure and planar failure, characterized as high deformability with no visible shearing planar. Specimens of SB-C25-28 ($\sigma_3 = 80$ and 160 kPa) and SB-C50-7 ($\sigma_3 = 40$ kPa) were captured as slight planar failure, as shown in Fig. 3.15(b).

Table 3.6 Failure modes of SB and SB-C specimens.

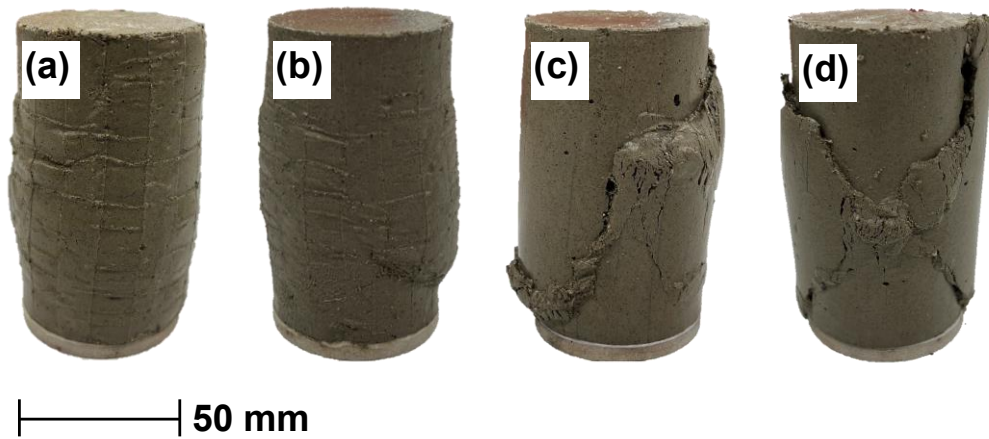
σ_3 (kPa)	SB-C0-0	SB-C25-7	SB-C25-28	SB-C50-7	SB-C50-28	SB-C75-7	SB-C75-28
40	a	a	a	b	c	d	d
80	a	a	b	c	c	d	d
160	a	a	b	d	d	d	d

* a. Ductile failure, b. Slight planar failure, c. Planar failure, d. Split failure.

Another important finding is that all of the SB-C75 was in split failure with obvious intersecting rupture planes, which can be explained by the hydration and pozzolanic reactions from high cement content, which forming calcium silicate hydrate gel and filling voids and fissures, resulting in the rigidity of specimens (Ouhadi et al., 2014; Ho et al., 2017; Luis et al., 2019). The split failure can also be witnessed in the SB-C50 under $\sigma_3 = 160$ kPa, which can be attributed to the combined effect of cementation and soil consolidation. The rest of the SB-50 specimens were in planar failure, as shown in Fig. 3.15(c), with one obvious failure planar cross the specimen.

To summarize the stress-strain and failure characteristics, as the cement content increases from 0 to 75 kg/m³, the stress-strain relationship changes from strain-hardening to strain-softening behavior, and the failure characteristics transformed from ductile failure to slight

planner failure to planner failure to split failure. In vertical cutoff walls, to maintain high deformability and good hydraulic performance, cracks (i.e. planar failure and split failure) should be rigorously prevented. The cement content from 25 kg/m³ to 50 kg/m³ is turning point, which represents the failure mode from ideal ductile failure to subpar planner failure, while 50 kg/m³ to 75 kg/m³ representing another crucial turning point that the stress-strain relationship converts from strain-hardening to strain-softening behavior, as the corresponding failure mode changes from subpar planner failure to undesirable split failure.



(a) Ductile failure, (b) Slight planar failure, (c) Planar failure, (d) Split failure.

Fig. 3.15 Typical failure modes of specimens.

3.3.3 Shear strength properties

After triaxial compression test, the specimens were in different failure modes with rupture planes within the specimen, which can be connected with different stress conditions in Mohr-Coulomb failure. The Mohr-Coulomb failure criterion defines that the failure of specimen (i.e. soil mass) will occur if the resolved shear stress τ on any planes in the specimen reaches a critical value, which can be written as (3-6),

$$\tau = \pm (c' + \sigma' \tan \varphi') \quad (3-6)$$

where c' is the intercept on the shear stress axis, as shown in Fig. 3.16(a). σ' represents the effective normal stress and φ' is the friction angle.

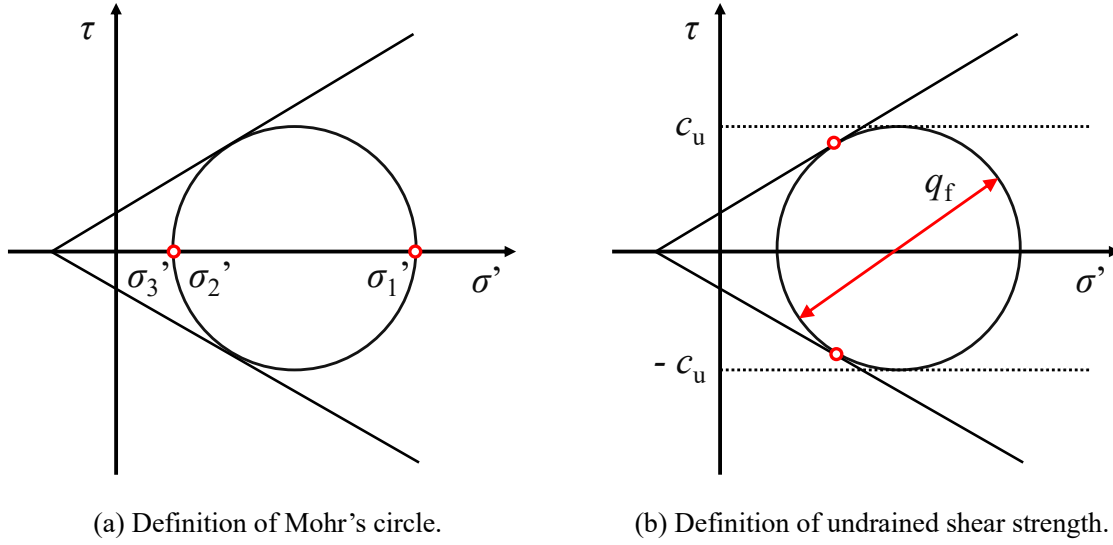


Fig. 3.16 Mohr-Coulomb failure circles.

In terms of $\sigma_2' = \sigma_3'$ in the isotropic CU triaxial compression test, where the cell pressure provides the minor (and equal intermediate) principal stress, the Mohr-Coulomb failure circle and pair of straight lines (i.e. failure lines) can be drawn as Fig. 3.16(a). In Fig. 3.16(a), the failure of the specimen occurs when the Mohr's circle of effective stress touches these pair of straight lines. Thus, in Fig. 3.16(b), the undrained shear strength c_u is the radius of the Mohr's circle, in other words, half of the ultimate deviator stress at failure q_f . The coefficient c is often called cohesion but is more usefully regarded merely as an intercept on the shear stress axis which defines the position of the Mohr-Coulomb strength line. Herein, in order to escape from the physical associations of the cohesion intercept, the symbol of c has been substituted as S (Wroth and Houlsby 1985). Therefore, the definition of undrained shear strength c_u based on Fig. 3.16(b) can be written as (3-7),

$$c_u = S_u = \frac{q_f}{2} = \frac{q_{\text{peak}}}{2} \quad (3-7)$$

where S_u represents the undrained shear strength in this chapter. The results of the undrained shear strength (S_u) and failure axial strain (ϵ_u) obtained from the triaxial CU test as a function of cement amendment under different confining pressure are summarized from the yellow points (i.e. peak deviator stress q_{peak}) in Fig. 3.13 and Fig. 3.14. The undrained shear strength (S_u) and failure axial strain (ϵ_u) are presented in Table 3.7 and drawn in Fig. 3.17.

Table 3.7 Undrained shear strength S_u and failure axial strain ε_u .

	40 kPa		80 kPa		160 kPa	
	S_u (kPa)	ε_u (%)	S_u (kPa)	ε_u (%)	S_u (kPa)	ε_u (%)
SB-C0-0 (SB)	10.6	15.0	17.9	15.0	30.4	15.0
SB-C25-7	35.7	15.0	43.2	15.0	65.1	15.0
SB-C25-28	47.2	15.0	51.5	6.7	81.3	15.0
SB-C50-7	93.1	15.0	89.5	15.0	116.6	11.1
SB-C50-28	166.0	7.9	174.1	11.9	145.7	15.0
SB-C75-7	141.8	7.9	153.9	6.3	190.5	7.1
SB-C75-28	248.5	15.0	325.9	5.5	334.7	2.3

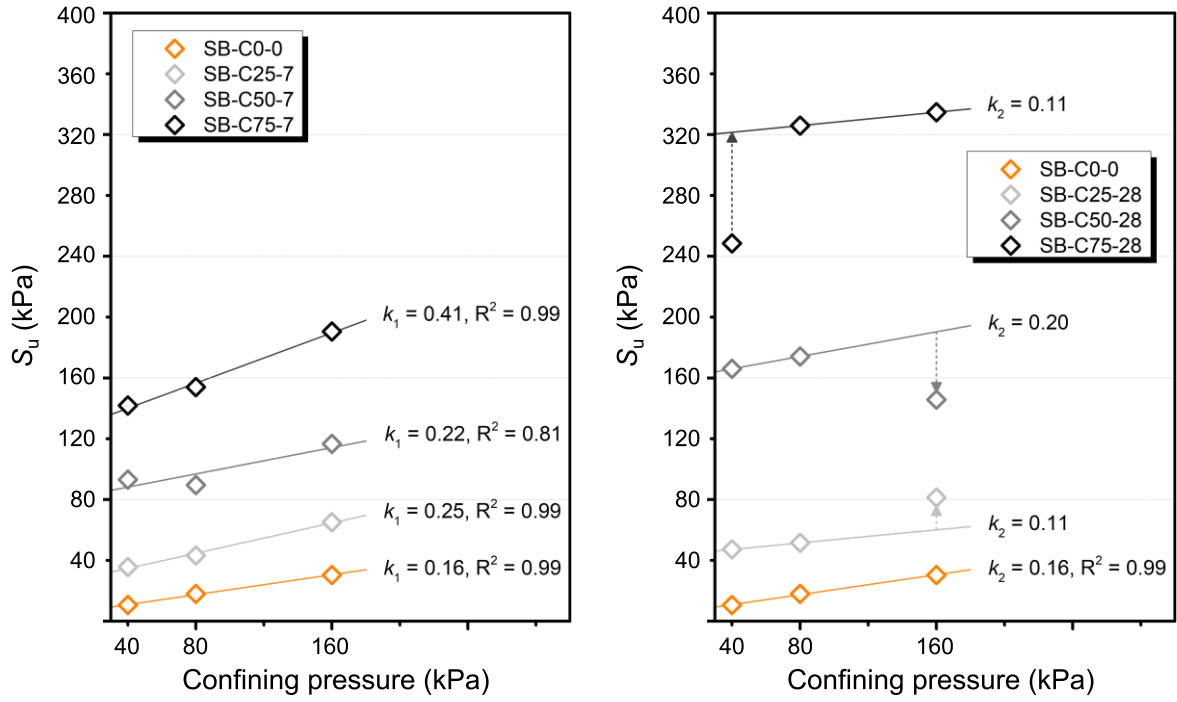


Fig. 3.17 Variation of undrained shear strength under different confining pressure.

At a certain confining pressure, the value of S_u increases as the cement amendment augments, but the ε_u was different. In SB-C0 and SB-C25, the ε_u was 15%, with one exception for the SB-C25 ($\sigma_3 = 80$ kPa) that the ε_u was 6.7% but the reduction percentage in q from peak to residual was only 3%. The ε_u in SB-C50-7 was 15% in $\sigma_3 = 40$ and 80 kPa cases, while 11.1% in $\sigma_3 = 160$ kPa case. Besides of the SB-C75-28 ($\sigma_3 = 40$ kPa), the ε_u of SB-C75 was relatively low with the range from 2.3% to 7.9%. Therefore, it suggested that the SB-C75 presents better

shear properties with the slag cement amendment. However, as compared with SB-C25 and SB-C50, the deformability of SB-C75 is relatively low due to the significant effect of cementation, as a result, the failures are obtained within 8.0% axial strain in most conditions.

From Fig. 3.17, it is obvious that the cement amendment has positive effect on S_u in either 7 D specimens or 28 D specimens. Furthermore, the favourable effect of curing can be witnessed by comparing Fig. 3.17(a) and (b) in each group of specimens at a certain confining pressure and slag cement content, especially in SB-C75 that the cementation occurs during 28 D significantly increase the S_u in 1.8 to 2.1 times comparing with 7 D. To estimate the effect of confining pressure and slag cement content on S_u , linear fitting on the specimens under $\sigma_3 = 40, 80$ and 160 kPa have been calculated in Fig. 3.17. The impact of confining pressure on S_u can be estimated by the slope of the fitting lines (i.e. k) and the corresponding R^2 . In Fig. 3.17(a), SB-C0-0, SB-C25-7 and SB-C75-7 are in well-fitted linear regression as the R^2 are over 0.99, which indicates that the confining pressure have the major effect on S_u at early curing time (7 D). And the increasing k_1 from 0.16 to 0.41 implies the interacted effect of cementation and consolidation, which jointly enhanced the undrained shear strength.

In Fig. 3.17(b), the amended groups (i.e. SB-C25-28, SB-C50-28, SB-C75-28) are not in well-fitted linear regression. Herein, the points of SB-C25-28 and SB-C50-28 in $\sigma_3 = 40$ and 80 kPa were connected as one straight line respectively, as well as the points of SB-C75-28 in $\sigma_3 = 80$ and 160 kPa, with individual slope k_1 from 0.11 to 0.20. Such low k_1 value indicates the minor impact of confining pressure. In SB-C25-28, the S_u increases sharply and above the straight line from $\sigma_3 = 80$ to 160 kPa, indicating the significant impact of high confining pressure. Through the curing time of SB-C, the primary cementitious products (e.g. calcium silicate hydrate (C-S-H), ettringite, and portlandite [$\text{Ca}(\text{OH})_2$, CH]) and secondary cementitious materials (e.g. C-S-H and calcium aluminate hydrate (C-A-H) enhanced the bonding between solid particles and filling the voids within the soil mass (Ryan and Day 2002; Carreto et al. 2015).

Herein, higher confining pressure (160 kPa) in SB-C25-28 stimulated the consolidation process, as a result, filling the pore space and enhanced the undrained shear strength. In SB-C75-28, the situation is reversed that the sharp increase is from $\sigma_3 = 40$ to 80 kPa, while minor increase is under higher confining pressure. Because of the high cement content, more cementitious materials generated in the specimen, as a result, small increment of confining pressure will significantly promote the consolidation and rearrangement of the soil mass particles. Therefore, higher confining pressure may present negligible effect on the volume change and void filling during the consolidation process, as well as the undrained shear stress.

The SB-C50-28 is the most typical case, which has a substantial decrease in S_u from $\sigma_3 = 80$ to 160 kPa, which can be attributed to the flocculation of bentonite in the interaction of bentonite and slag cement, resulting in the loss of deformability and lower shear resistance (Gaucher and Blanc 2006; Balasubramaniam and Chaudry 1978).

Therefore, after 28 days' curing, S_u is dependent on the intersecting impact between cementation and consolidation, as 50 kg/m³ is the turning point, which featured with two stress-strain relationship (i.e. strain-softening and strain-hardening) and three failure modes (i.e. slight planar, planar and split failure). When the cement content is lower than 50 kg/m³ (i.e. 0 and 25 kg/m³), consolidation is the major effect over cementation. On the contrary, when the cement content is higher than 50 kg/m³ (i.e. 75 kg/m³), the cementitious materials from cementation will attenuate the effect of consolidation. The $R^2 = 0.81$ and relatively lower k_1 can also be explained by this reason.

Another crucial shear strength property is the residual undrained shear strength S_r , which is defined as half of the deviator stress q at 15% axial strain (final displacement). The S_r obtained from Fig. 3.13 and Fig. 3.14 has been summarized in Table 3.8.

Table 3.8 Residual undrained shear strength S_r .

σ_3 (kPa)	SB-C0-0	SB-C25-7	SB-C25-28	SB-C50-7	SB-C50-28	SB-C75-7	SB-C75-28
40	10.5	35.7	44.1	93.1	158.0	140.7	248.0
80	17.5	43.2	49.5	89.5	171.5	148.0	292.5
160	30.4	64.7	81.5	118.4	153.0	182.0	323.0

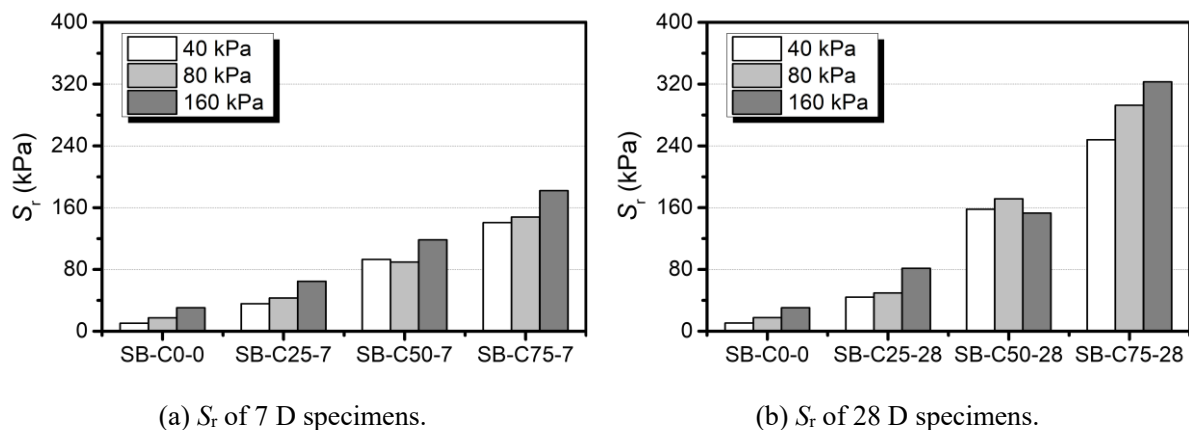


Fig. 3.18 Variation of residual undrained shear strength under different cement content.

Fig. 3.18 shows the variation of residual undrained shear strength (S_r) as the function of slag cement amendment. It can be observed that the S_r increases with the increasing of confining

pressure at a given cement amount except for the SB-C50. Similar upward variation trend of S_r with the augment of cement addition can be found, as the variation trend of S_u in Fig. 3.17. Moreover, when comparing S_r with S_u , for SB and SB-C25, the S_r is about the same with S_u . For SB-C50 and SB-C75, the S_r is exceeding 95% on average the S_u , which points to high strength retention characteristics after reaching peak deviator stress q . Therefore, regardless of the stress-strain characteristics, failure mode and undrained shear strength values, all of SB and SB-C have good strength retention properties.

3.3.4 Effective stress path and strength envelope

As according to (3-6), it is assumed that, for sliding to occur on any plane within the specimen, the shear stress has to overcome a frictional resistance $\sigma' \tan \phi'$, which is dependent on the effective normal stress σ' acting on the plane and on a friction angle ϕ' , and c' , which is independent of σ' . In the elastic-plastic model, the specimen eventually tended towards an ultimate condition in which plastic shearing could continue indefinitely without changes in volume or effective stresses (Wood 1990). This specific condition of perfect plasticity is known as the Critical State Line (CSL), of which the attainment can be expressed as (3-8),

$$\frac{\partial p'}{\partial \varepsilon_q} = \frac{\partial q}{\partial \varepsilon_q} = \frac{\partial v}{\partial \varepsilon_q} = 0 \quad (3-8)$$

In the triaxial compression test, the critical state line is reached with an effective stress ratio, as presented in (3-9),

$$\eta = \frac{q}{p'} = M \quad (3-9)$$

where M is the frictional constant. M is related to the effective stress friction angle, ϕ' . In Fig. 3.16(a), the relationship between the major and minor principal effective stress is (3-10) as considered on (3-6),

$$\frac{\sigma_1' + c' \cot \phi'}{\sigma_3' + c' \cot \phi'} = \frac{1 + \sin \phi'}{1 - \sin \phi'} \quad (3-10)$$

Equation (3-9) can be rewritten in terms of q and p' in (3-4) and (3-5), and becomes (3-11),

$$\frac{q}{p' + c' \cot \phi'} = \frac{6 \sin \phi'}{3 - \sin \phi'} \quad (3-11)$$

Based on (3-11), the relationship between q and p' has been plotted in Fig. 3.19.

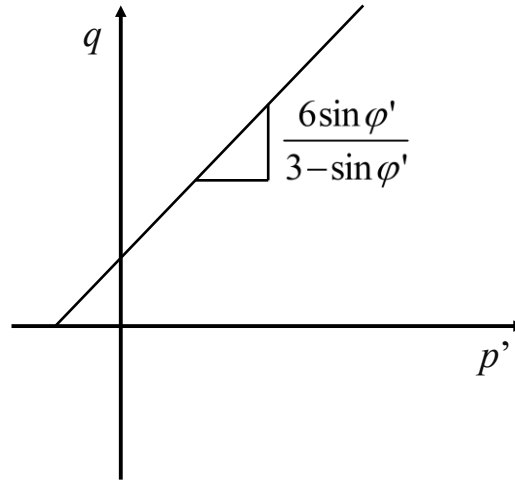


Fig. 3.19 Mohr-Coulomb failure criterion.

Comparison of (3-9) and (3-11) suggests that soils are failing in a purely frictional manner at the critical state (i.e. $c'=0$). The deformations have been so large that the soil has been thoroughly churned up, and any bonding between particles which might have led to some cohesive strength has broken down. In this circumstance, the slope of CSL in Fig. 3.19 can be written as (3-12),

$$M = \frac{6 \sin \phi'}{3 - \sin \phi'} \quad (3-11)$$

In order to evaluate M , Tests should be carried out to large strains to ensure that the effective stress path (i.e. q - p' line) are close to or attaching the CSL. The slope of a best-fitting line of effective stress path will be the frictional constant M . Hereafter, the CSL determined from $\sigma_3 = 40, 80$ and 160 kPa in certain cement content and confining pressure, are named as strength envelope.

In this chapter, the deviator stress q versus mean effective confining stress p' during the triaxial compression test of SB and SB-C 7 D specimens is shown in Fig. 3.20, and that of SB and SB-C 28 D specimens is shown in Fig. 3.21. These curves are strength path with strength envelopes, which was evaluated according to the peak deviator stress and the best linear fitting of these critical state points.

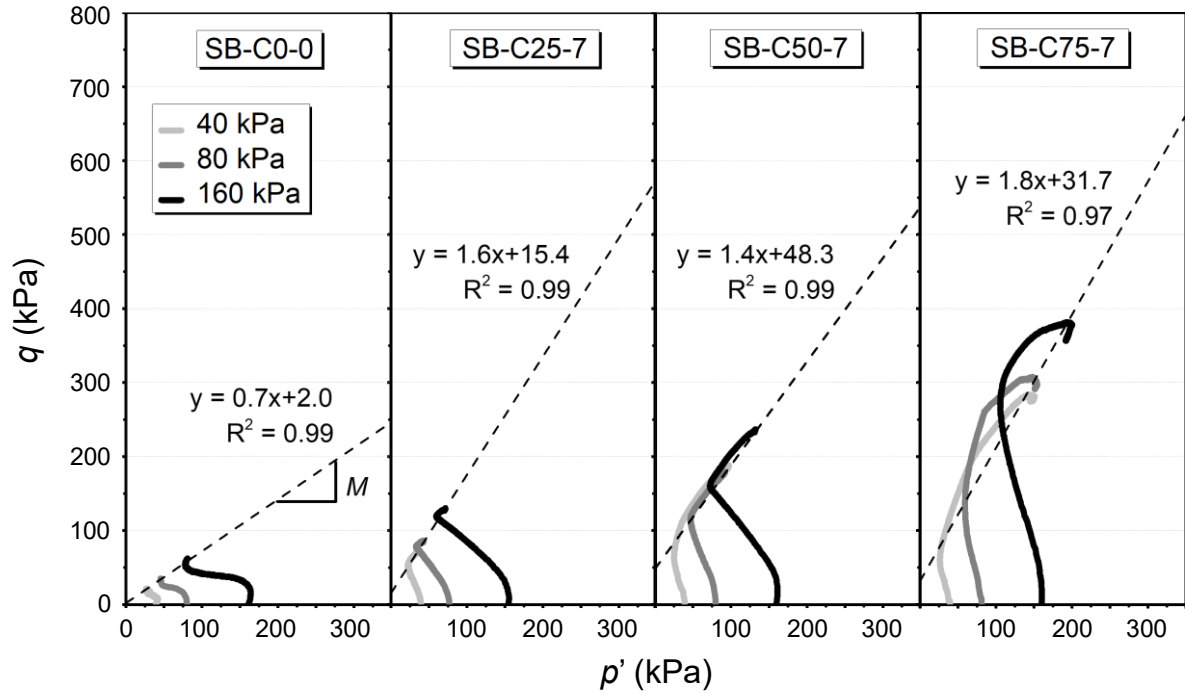


Fig. 3.20 The effective stress path and strength envelope of SB and SB-C 7 D specimens.

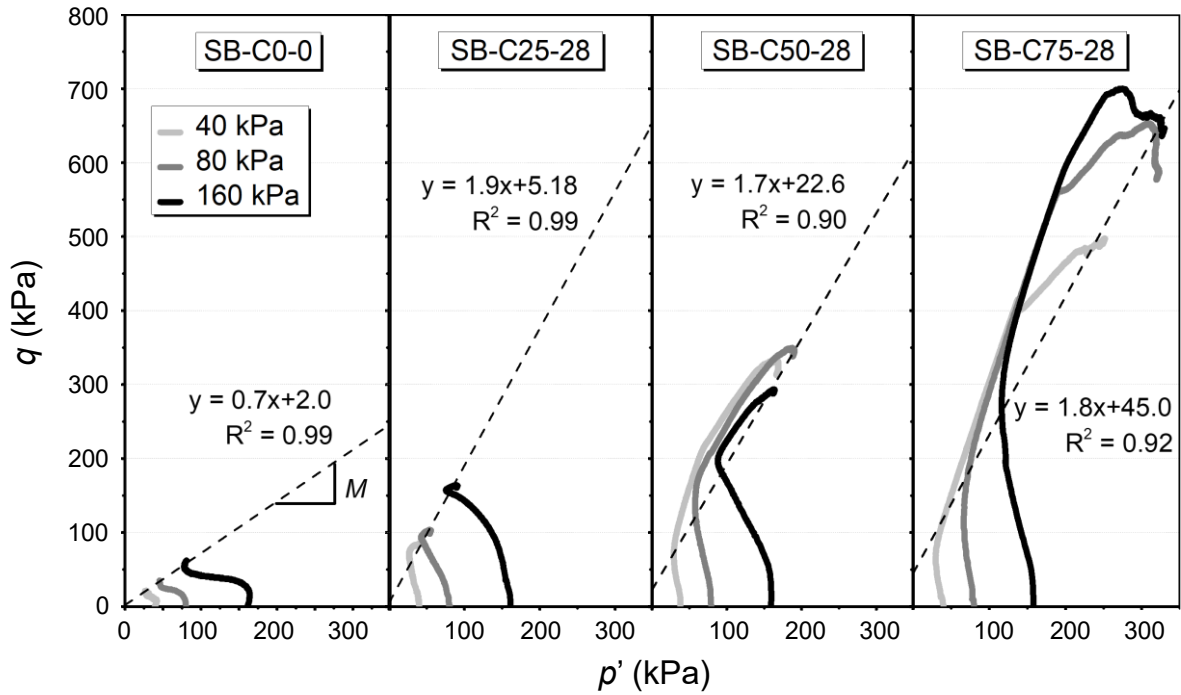


Fig. 3.21 The effective stress path and strength envelope of SB and SB-C 28 D specimens.

In Fig. 3.20 and Fig. 3.21, the slope M of linear strength envelopes for SB and SB-C specimens were determined to estimate the effective strength parameters (c' and ϕ'). The relationship between the internal friction angle ϕ'_{peak} and the parameter M is expressed in

equation (3-12) (Luis et al., 2019). And the effective cohesion coefficient (c') is determined using equation (3-13),

$$M = \frac{6 \sin \varphi'_{\text{peak}}}{3 - \sin \varphi'_{\text{peak}}} \quad (3-12)$$

$$c' = \frac{c}{M} \quad (3-13)$$

where the c is the intercept with the q axis of the linear envelopes.

Table 3.9 Summary of effective cohesion coefficient c' and effective friction angle φ' .

	SB-C0		SB-C25		SB-C50		SB-C75	
Curing time (D)	c' (kPa)	φ'	c' (kPa)	φ'	c' (kPa)	φ'	c' (kPa)	φ'
0	2.0	35.8	-	-	-	-	-	-
7	-	-	15.4	58.2	24.8	59.3	31.7	61.0
28	-	-	5.2	61.6	22.6	59.5	45.0	61.9

The estimated effective cohesion coefficient c' and effective friction angle φ' have been summarized in Table 3.9. The c' and φ' of SB-C0 was 2.0 kPa and 35.8°. The relationship between c' and φ' with the cement content (e.g. 25, 50 and 75 kg/m³) and curing time (e.g. 7 and 28 D) has been illustrated in Fig. 3.22. In a certain curing time (7 D or 28 D), the c' is in best linear fitting ($R^2 = 0.99$) with the cement content, indicating the significant effect of cementation on c' . The c' and φ' of 50 kg/m³ were almost unchanged from 7D to 28 D, which implied 50 kg/m³ is a turning point as found in previous discussion. Moreover, the k_2 of 28 D c' fitting line is higher than that of 7 D. This phenomenon verifies soil bentonite particles are the major effect over cementitious materials at early age and also when the cement content is lower than 50 kg/m³, while the situation undergoes a reversal when the content increases to 75 kg/m³.

For the φ' , the influences of both cement content and curing time are negligible, because minor increase can be seen in φ' , ranging from 58.2° to 61.9° in Fig. 3.22. Owing to the effect of curing, cementitious materials fill the voids between soil particles, reducing particle displacement. However, cement particles themselves make no enhancement on soil particles (i.e. change their shape or size). Therefore, the φ' changed from SB to SB-C, but tiny variation can be witnessed in all the SB-C. On the contrary, the cementation mainly affects soil cohesion c' . Cementitious materials bond soil particles together, forming a strong skeleton structure and

increase soil cohesion c' , as a result, enhances soil strength (e.g. S_u , S_r).

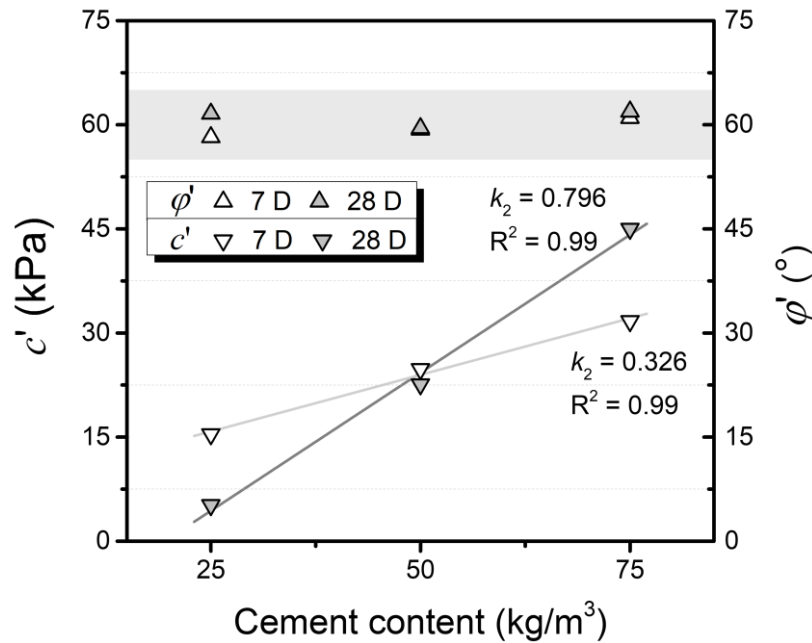


Fig. 3.22 The effect of slag cement content on c' and ϕ' .

The ϕ' is independent on the cement content and curing time, which can be attributed to the similar structure in slag cement amended specimens. As shown in Fig. 3.23, at certain curing time, the cementitious materials will bond soil particles or fill the voids. The slag cement itself contains major strength producing compounds whereas bentonite and soil not. Through the hydration and cementation process, the void within the specimen will decrease from three ways. The first is the cement itself hydrated as bigger particles than bentonite and soil, as a result, fill the voids. The major big particles are tricalcium silicate (C_3S). dry cement contains dicalcium silicate (C_2S), tricalcium aluminate (C_3A), and tetra-calcium Aluminio ferrite (C_4AF), pozzolanic type reaction for additional cementation. The other is the clay portion within the soil mass (i.e. bentonite) very likely also undergoes a pozzolanic type reaction with the cement to produce further cementation.

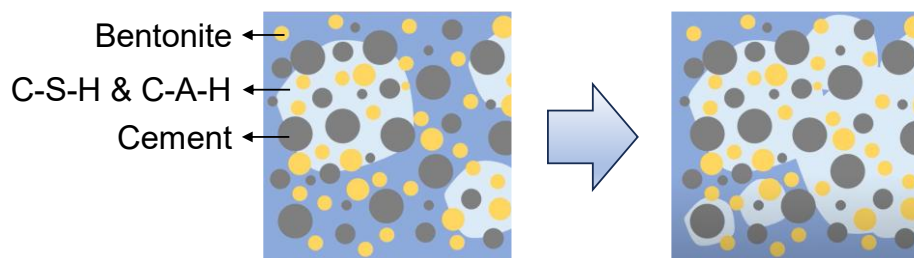


Fig. 3.23 The cementitious particles in the soil mass.

Owing to the cementation reactions, the particles have dramatically changed, correspondingly, the gradation within the specimen also differed. Small amounts of slag cement cause the smaller size particles in a soil to be cemented together to form larger ones and leading to the specimen to dilate during compression thus causing them to behave as over consolidated soils, as shown in Fig. 3.20 and Fig. 3.21. From SB-C0 and SB-C25 to SB-C50 and SC-C75, the failure mechanism varied from particle collapse to dilation, generally the specimen failure modes transformed from ductile to planar and split.

The observed changes in shape of the effective stress paths with increasing curing time and confining pressure in SB-C50 and SB-C75 reflect an increasing tendency for the cemented soil skeleton to dilate during compression or at least to reduce the compression tendency. The small changes in ϕ' can be explained by the similar gradation of the specimens that the big particles interlock with other soil particles to bear the compression. It needs significant more energy to make big particles to overlap from each other, as a result, the particles compressed within themselves first, leading to the increase of the total volume. When the dilation occurred, the specimens tended to totally destroy, which is not consistent with the originality of this research that obtain certain strength properties and maintain deformability and swelling properties.

3.3.5 Excess pore pressure characteristics

Analysis of the mechanical behavior of the specimen (e.g. whether it will fail or how much it will deform) requires an estimate of the excess pore pressure that are likely to develop as a result of the applied changes in total stresses so that the changes in effective stresses may in turn be estimated. Hereafter, the effective stress path and total stress path are shortened as ESP line and TSP line. The principle of effective stress indicates that excess pore pressure u is the difference between the total and effective mean stress as (3-14), which can also be written in an incremental form as (3-15).

$$u = p - p' \quad (3-14)$$

$$\delta u = \delta p - \delta p' \quad (3-15)$$

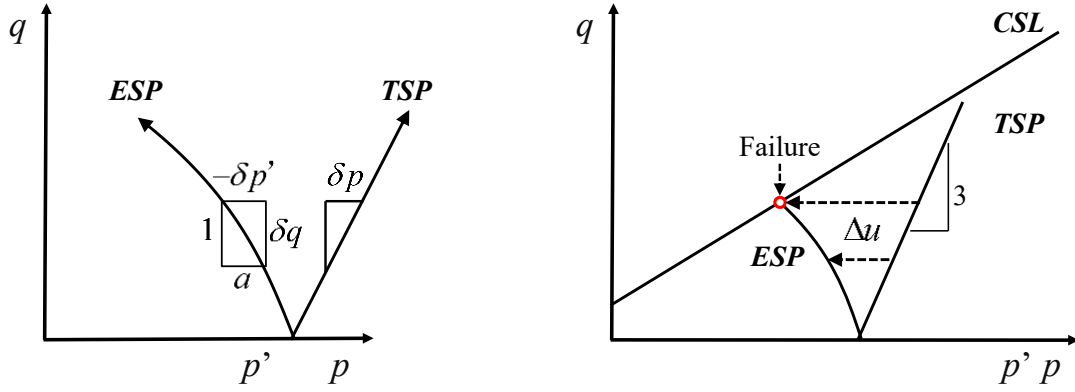
The equation (3-15) can be further illustrated through Fig. 3.24, that the excess pore pressure increase is in part to the increase in total mean stress p and correspondingly in part to the decrease in effective mean stress p' . At a certain stage of the CU triaxial compression test (i.e. the increment on ESP in Fig. 3.24(a)), the change of deviator stress and mean effective stress can be linked as (3-16),

$$\delta p' = -a\delta q \quad (3-16)$$

Therefore, (3-16) can be written as (3-17) based on (3-15),

$$\delta u = \delta p + a\delta q \quad (3-17)$$

where a is the excess pore pressure parameter, which is variable depending on the loading.



(a) Generation of excess pore pressure.

(b) Variation of excess pore pressure.

Fig. 3.24 The identification of excess pore pressure.

Based on the above determinations, the variation of excess pore pressure can be illustrated as Δu in Fig. 3.24(b). From equation (3-4) and (3-5), the relationship between p and q can be expressed as (3-18),

$$p = \frac{\sigma_1 + 2\sigma_3}{3} = \frac{(\sigma_1 - \sigma_3) + 3\sigma_3}{3} = \sigma_3 + \frac{q}{3} \quad (3-18)$$

In this chapter, the CU triaxial compression test as the cell pressure was keeping constant in each case (i.e. $\partial\sigma_3 = 0$). Thus, (3-19) can be reached if $\partial\sigma_3 = 0$.

$$\delta p = \frac{\delta q}{3} \quad (3-19)$$

In Fig. 3.24(b), the TSP line follows (3-18), with the gradient of 3 from an initial stress condition in different confining pressure cases (e.g. 40, 80, 160 kPa). To evaluate the variation of excess pore pressure in the compression process, the TSP line has been drawn with the ESP lines from Fig. 3.20 and Fig. 3.21, and it comes to Fig. 3.26 and Fig. 3.27.

As according to Fig. 3.24(b) and equation (3-17), the variation of excess pore pressure depending on the effective stress path, which relied on the axial compression. Thus, the variation characteristic of excess pore pressure has been illustrated in Fig. 3.25, which is

comparable to the stress-strain relationship shown in Fig. 3.13 and Fig. 3.14. As shown in Fig. 3.25, the variation characteristics of excess pore pressure in this chapter, can be identified into three categories. The a is mainly witnessed in SB and SB-C25, while b in the SB-C50 and c in SB-C75, while some of SB-C50 changes in c.

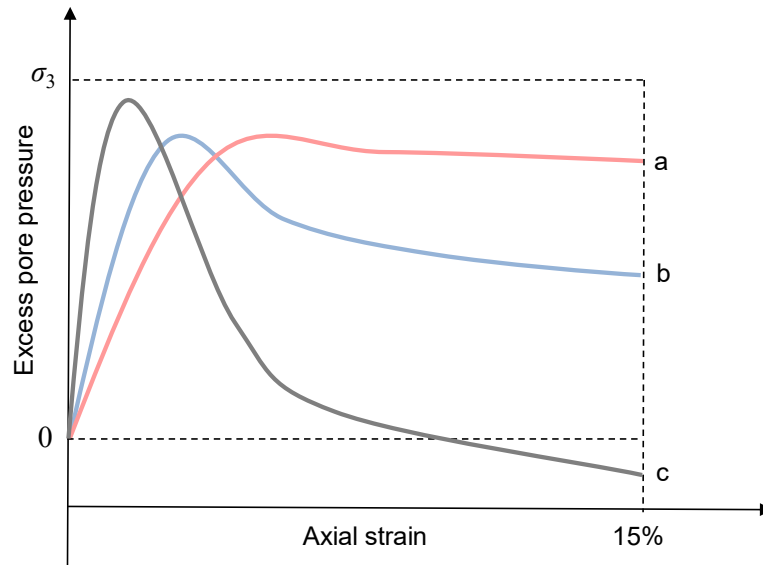


Fig. 3.25 The variation of excess pore pressure versus axial strain.

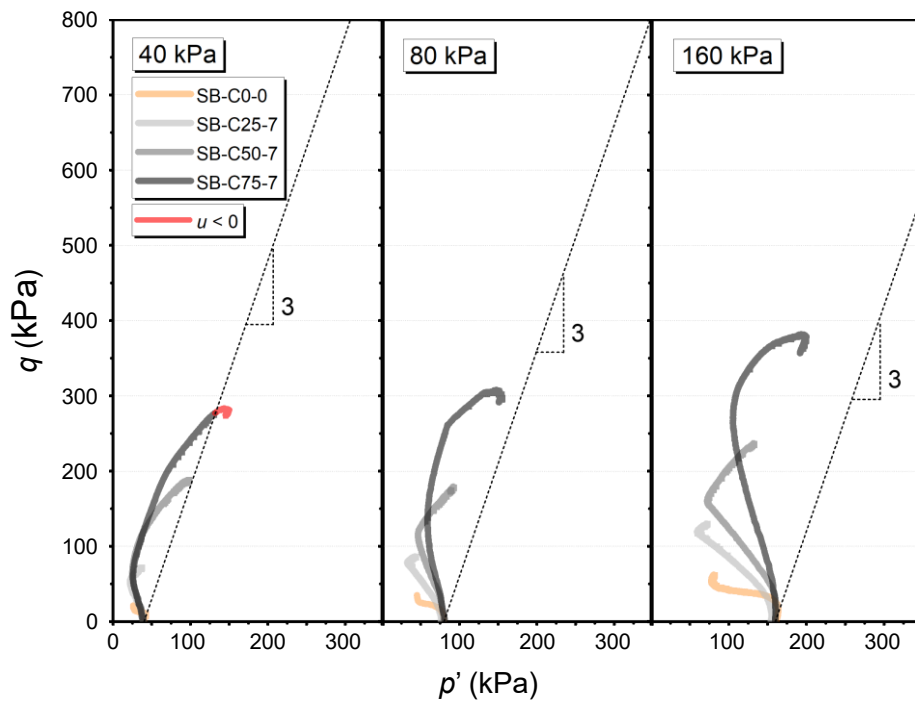


Fig. 3.26 The effective stress path vs total stress path in SB and SB-C 7 D specimens.

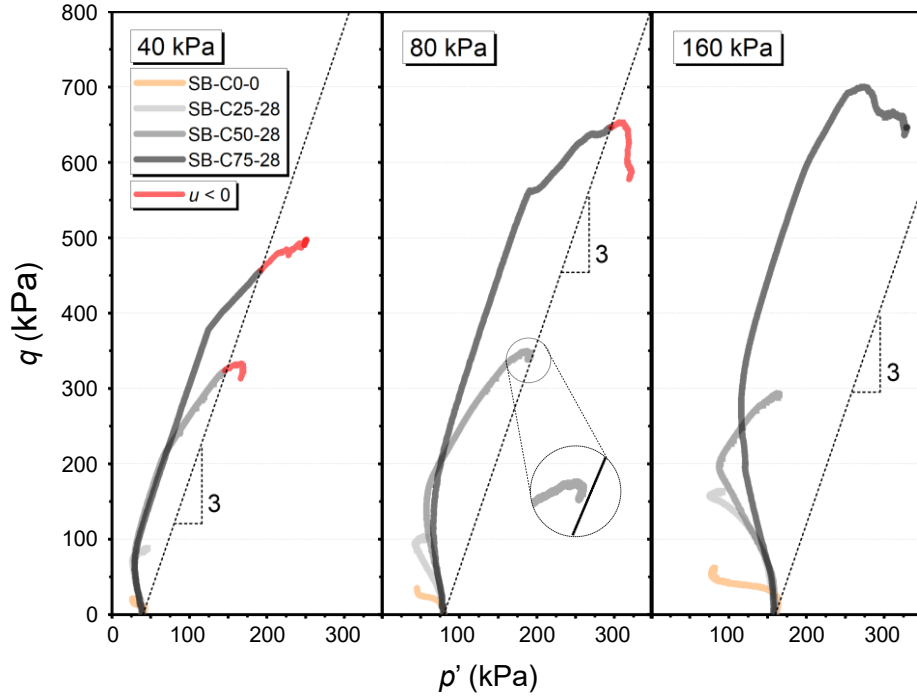


Fig. 3.27 The effective stress path vs total stress path in SB and SB-C 28 D specimens.

As shown in Fig. 3.26 and Fig. 3.27, the variations of excess pore pressure in all SB and SB-C25 are positive during the compression. The peak excess pore pressure was reached around the axial strain of 1.5%, 2% and 3% in $\sigma_3 = 40, 80$ and 160 kPa conditions, respectively. In Fig. 3.25, from the start of compression, there is a rapid increase in u as the soil undergoes deformation. As the compression progresses, the free water in the soil mass gradually drains out from soil particles to accommodate the deformation of the specimen. Then, the rate of u variation slows down, leading to a gradual and stabilized u , because high proportion of adsorbed water within the bentonite remained undisturbed (Wang 2023).

However, as the cement content increased, the flocculation of bentonite will be more obvious as the calcium ions (Ca^{2+}) released from cementitious materials replace the monovalent cations (e.g. sodium ions (Na^+)), that are held at exchangeable sites of bentonite. As a result, the effect of cementation will be more significant, and higher proportion of pore water in the specimen are in the form of free water instead of adsorbed water within the hydrated bentonite structure. Therefore, the variation of excess pore pressure could be in b and c in Fig. 3.25 that keeps decreasing, even below 0 kPa. In Fig. 3.26 and Fig. 3.27, when the ESP line intersect and exceed the TSP line, the u will decrease below 0 kPa, which has been marked in light red colour. In this chapter, the u of four specimens decreased to below 0, and they are SB-C75-7 in $\sigma_3 = 40$ kPa, SB-C50-28 and SB-C75-28 in $\sigma_3 = 40$ kPa, and SB-C75-28 in $\sigma_3 = 80$ kPa.

An important finding is that all these four specimens are relatively high cement content cases in low confining pressure conditions, regardless of the curing time. On the one hand, the cementitious materials filling the voids within the soil mass, as a result, decreasing the void ratio of the specimens (Ryan and Day 2002; Carreto et al. 2015). On the other hand, the cementation and consolidation enhanced the bonding of the particles, leading to small deformation among the cemented soil particle mass during the compression process. Hence, because the free water in the specimen is difficult to flow within the voids. Initially, the excess pore pressure sharply increases and approaches the confining pressure σ_3 , then gradually decreases in similar reduction as in other σ_3 conditions. Therefore, when the σ_3 is low (i.e. 40 kPa), the u will decrease to below 0 earlier, representing the pore water remaining in the voids, neither escape from the voids nor flow through the soil particle voids. Such phenomenon indicating that the soil has not reached an equilibrium state under compression, which compromise the stability of the soil structure and undesirable for the amendment of SB.

3.3.6 Young's modulus E_0 and secant modulus E_{50}

From the Generalized Hooke's Law, the elastic modulus E is determined by the stress σ and strain ε , as expressed in (3-19)

$$E = \frac{\sigma}{\varepsilon} \quad (3-19)$$

The stress condition of elements in the specimen during the triaxial compression has been shown in Fig. 3.28(a). The variation of axial strain $\Delta\varepsilon_1$ is related to the stress increments from axial and lateral, which comes to (3-20), and $\Delta\varepsilon_2$ can be expressed as (3-21).

$$\Delta\varepsilon_1 = \frac{1}{E} [\Delta\sigma_1 - \mu(\Delta\sigma_2 + \Delta\sigma_3)] \quad (3-20)$$

$$\Delta\varepsilon_2 = \frac{1}{E} [\Delta\sigma_2 - \mu(\Delta\sigma_1 + \Delta\sigma_3)] \quad (3-21)$$

In this chapter, $\sigma_2 = \sigma_3$ and the Poisson's ratio μ is normally considered as 0.5 for saturated clay, the secant modulus E_{sec} can be calculated following equation (3-22) from the stress-strain curve, as show in Fig. 3.28(b).

$$E_{\text{sec}} = \frac{\Delta\sigma_1 - \mu(\Delta\sigma_2 + \Delta\sigma_3)}{\Delta\varepsilon_1} = \frac{\Delta\sigma_1 - 2\mu\Delta\sigma_3}{\Delta\varepsilon_1} = \frac{\Delta\sigma_1 - \Delta\sigma_3}{\Delta\varepsilon_1} = \frac{\Delta q}{\Delta\varepsilon_1} \quad (3-22)$$

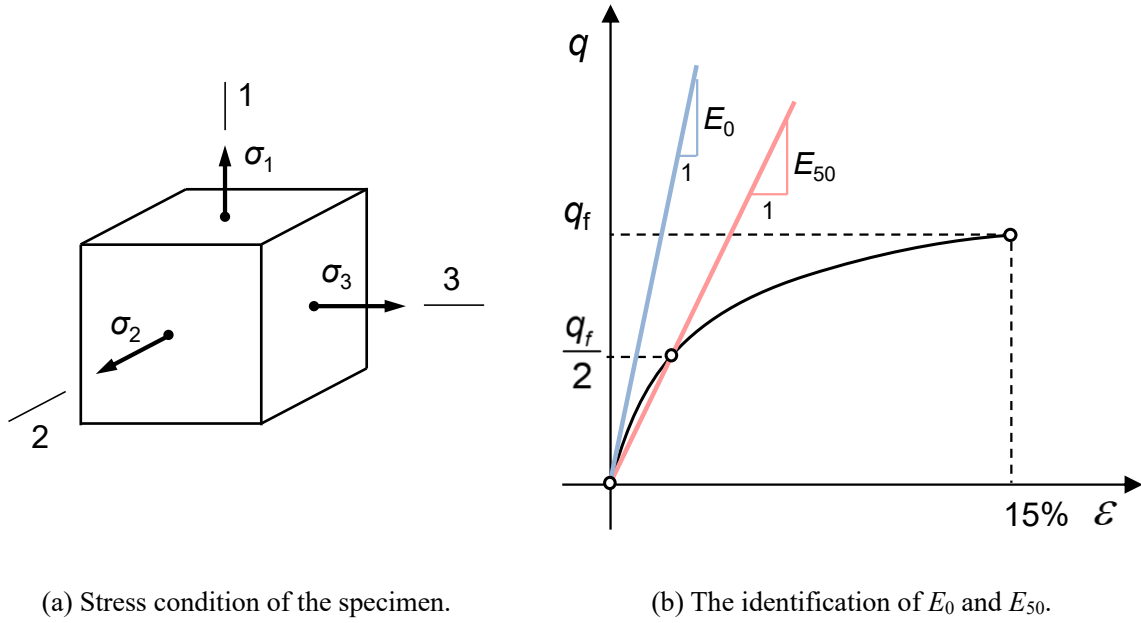


Fig. 3.28 Stress condition of the specimen and the identification of E_0 and E_{50} .

The initial Young's modulus E_0 and secant modulus E_{50} were obtained for all specimens. As shown in Fig. 3.28, the E_0 is typically calculated as the slope of the initial linear portion of the stress-strain curve, representing the initial stiffness of the specimen. The E_{50} is determined by identifying the deviator stress corresponding to 50% of the failure strength q_f and dividing it by the corresponding strain (i.e. slope of the secant line at q_f). E_0 and E_{50} represent the variations in stiffness of the specimen, which denote the deformation resistance properties in axial loading. The results of E_0 and E_{50} are summarized in Table 3.10. The variation of E_0 and E_{50} under the effect of cement content and confining pressure have been plotted in Fig. 3.29. In this chapter, the E_0 and E_{50} were obtained from the stress-strain curves in Fig. 3.13 and Fig. 3.14.

Table 3.10 E_0 and E_{50} of SB and SB-C specimens.

Confining pressure (kPa)	E_0 (MPa)			E_{50} (MPa)		
	40	80	160	40	80	160
SB-C0-0	8.9	25.8	43.7	4.2	24.6	13.2
SB-C25-7	32.2	56.8	70.0	9.4	15.8	44.1
SB-C50-7	61.5	78.8	92.8	19.5	32.0	35.6
SB-C75-7	39.8	86.5	120.6	15.9	73.9	87.3
SB-C25-28	37.5	46.2	77.8	16.7	19.7	33.3
SB-C50-28	55.5	69.2	111.7	26.8	28.4	34.4

SB-C75-28	54.0	97.4	132.1	44.0	74.4	81.5
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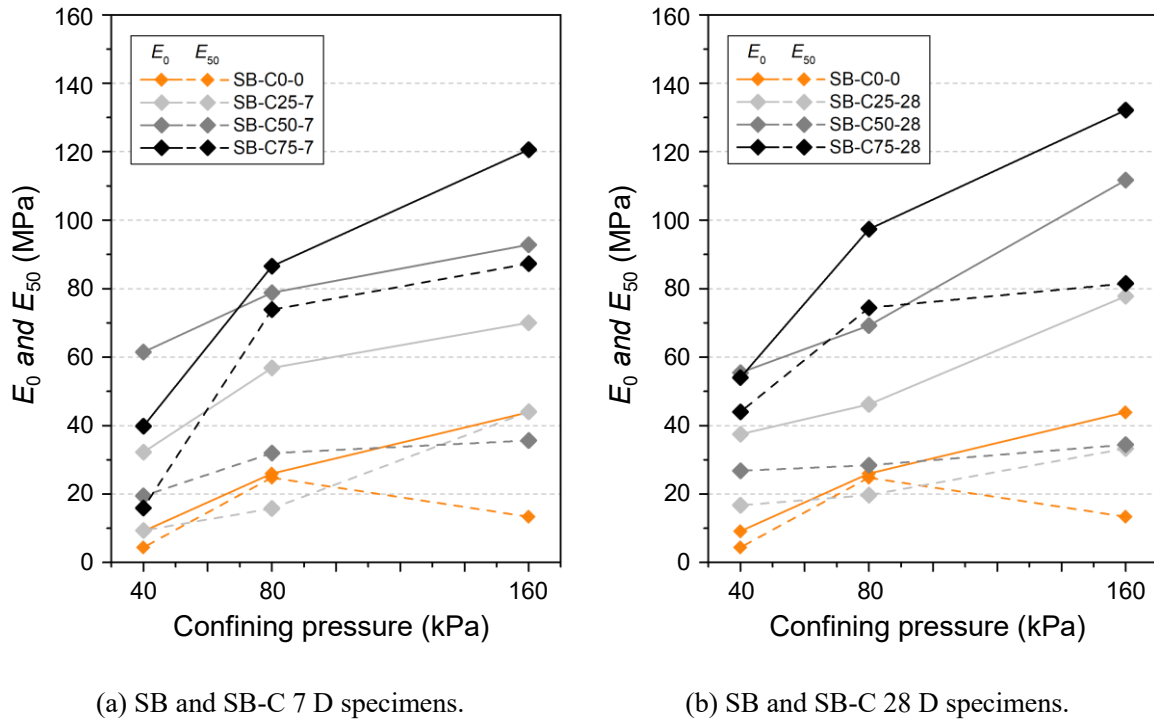


Fig. 3.29 Variation of E_0 and E_{50} under the effect of cement content and confining pressure.

Regarding Fig. 3.29, the moduli (E_0 , E_{50}) of SB-C specimens rise with an increase in the cement content and confining pressure. Furthermore, the augmentation of moduli (E_0 , E_{50}) with the increase in the cement content is more evident at the higher values of S_u . Because at higher cement content, due to enhancement of the formed cementitious bonds, the specimens behave more likely rigid materials. When comparing the E_0 to E_{50} , it is evident that, the E_{50} is less than 50% of E_0 in most of SB-C25 and SB-C75 conditions. It is noteworthy that this value does not align with that of typical elastic material, which tends to be significantly higher. The SB-C75 specimens present higher stiffness retention capacity, as most of the E_{50} were higher than 70%.

The unamended SB-C0 specimens were highly stress dependent (i.e. consolidation effect), that E_{50} sharply decrease under 160 kPa confining pressure (Opdyke and Evans, 2005; Pashang Pisheh and Mir 2019). For the SB-C specimens, the E_{50} decreases which can be attributed to the breakage of interconnected bonds and crystalline structure formed of calcium silicate hydrate (C-S-H) and calcium aluminate hydrate (C-A-H), being capable of supporting compressive stress. Axial loads applied to the SB-C75 specimens are initially supported by this rigid structure (Chew et al., 2004; Evans 1995). After exceeding ultimate strength, which is strongly affected by the small contact area between crystals, this solid-like skeleton is partially

destroyed, leading to the decrease in E_{50} (Carreto et al., 2016; Subramaniam 2019).

3.4 Conclusions for this chapter

In this chapter, the mechanical performance of soil bentonite mixture amended with slag cement was studied based on the results of consolidated-undrained (CU) isotropic triaxial compression tests. The stress-strain characteristics, failure modes, shear strength properties, etc. were evaluated and effects of cement content (e.g. 0, 25, 50 and 75 kg/m³), curing period (e.g. 7 and 28 days) and confining pressure (e.g. 40, 80 and 160 kPa) were discussed. The main results obtained in this chapter can be summarized as follows:

- (1) The major influencing factor on the stress-strain characteristics is the slag cement content. The SB-C0 and SB-C25 were characterized as stress-hardening behavior, and the SB-C75 specimens are strain-softening. The most typical stress-strain characteristics are in SB-C50, as the stress-strain relationship presents both strain-hardening and strain-softening behavior.
- (2) For the failure modes, the cement content from 25 kg/m³ to 50 kg/m³ is one turning point, which represents the failure mode from ideal ductile failure to subpar planner failure, while 50 kg/m³ to 75 kg/m³ representing another crucial turning point that the failure mode changes from subpar planner failure to undesirable split failure.
- (3) The cement amendment has positive effect on S_u and S_r in both 7 D and 28 D specimens. When the cement content is lower than 50 kg/m³ (i.e. 0 and 25 kg/m³), consolidation is the major effect over cementation at both early age and 28 D. On the contrary, when the cement content is higher than 50 kg/m³ (i.e. 75 kg/m³), the cementitious materials from cementation will attenuate the effect of consolidation, especially in 28 D. When comparing S_r with S_u , for SB and SB-C25, the S_r is about the same with S_u . For SB-C50 and SB-C75, the S_r is exceeding 95% on average the S_u , which points to high strength retention characteristics after reaching peak deviator stress q .
- (4) In this chapter, the ultimate undrained shear strength S_u was 334.7 kPa in SB-C75-28 under 160 kPa confining pressure obtained at 2.3% axial strain. With the augment of cement content and confining pressure, the first $S_u > 100$ kPa was witnessed in SB-C50-7 under 160 kPa confining pressure.
- (5) The influences of both cement content and curing time on ϕ' are negligible, because minor increase can be seen in ϕ' , ranging from 58.2° to 61.9°. The c' have linear relationship with the cement content, and the effect of cementation more significant after 28 D curing. The specimen tended to dilate in SB-C50 and SB-C75, while tend to collapse in SB-C0 and SB-

C25, because of the changes in the particle size and gradations. When the specimen dilate, the failure is more likely to be planar and split failure.

- (6) The variations of excess pore pressure in all SB and SB-C25 are positive during the compression. The peak excess pore pressure was reached around the axial strain of 1.5%, 2% and 3% in $\sigma_3 = 40, 80$ and 160 kPa conditions, respectively.
- (7) The u of four specimens decreased to below 0, and they are SB-C75-7 in $\sigma_3 = 40$ kPa, SB-C50-28 and SB-C75-28 in $\sigma_3 = 40$ kPa, and SB-C75-28 in $\sigma_3 = 80$ kPa. All these four specimens are relatively high cement content cases in low confining pressure conditions, regardless of the curing time, indicating that the soil has not reached an equilibrium state under compression.
- (8) The unamended SB-C0 specimens were highly stress dependent (i.e. consolidation effect), that E_{50} sharply decrease under 160 kPa confining pressure. the moduli (E_0, E_{50}) of SB-C specimens rise with an increase in the cement content and confining pressure. Furthermore, the augmentation of moduli (E_0, E_{50}) with the increase in the cement content is more evident at the higher values of S_u .
- (9) When comparing the E_0 to E_{50} , it is evident that, the E_{50} is less than 50% of E_0 in most of SB-C25 and SB-C75 conditions. The SB-C75 specimens present higher stiffness retention capacity, as most of the E_{50} were higher than 70%, implying higher compression resistance.

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Chapter 4

Hydraulic performance of SB mixtures amended with slag cement

4.1 General remarks

The hydraulic performance is a key characteristic which governing the effectiveness of cutoff walls (Britton et al., 2004). The hydraulic performance is quantized to k value (i.e. hydraulic conductivity), which affected by various physical and chemical factors (e.g. stress states, groundwater conditions, material properties, etc.).

The hydraulic conductivity k of soil bentonite mixtures (SB) is highly stress dependent, and the k value of SB is typically between 1×10^{-9} and 1×10^{-10} m/s from laboratory tests. (Evans 1994; Filz et al., 2003; Takai 2014). With the amendment of cement, the hydraulic performance is affected by the curing time, which normally specified as 7, 28 or 90 days. The k value of cement amended soil bentonite mixtures (SB-C) continues to decline over time, which is assumed to be a conservative value (Opdyke and Evans 2005).

During the curing process, the bentonite in close contact with the cement may be chemically affected, as serious cations alteration being an attack on the montmorillonite clay crystal lattice, influencing the swelling performance of the bentonite, which significantly affect the hydraulic performance of SB and SB-C (Chen et al., 2019). However, certain amendment of cement does not affect the pore structure refinement by the bentonite that montmorillonite favorably offers additional nucleation sites for C-S-H and exhibited higher pozzolanic activity as well as, the denser microstructure, leading to more tortuous hydraulic paths to improve the hydraulic performance of SB-C (Yang et al., 2019).

In this chapter, the swelling performance of the bentonite under the effect of cement content has been estimated. Through the triaxial compression apparatus introduced in Chapter 3 and a flexible wall permeameter with falling head system, the hydraulic performance of soil bentonite mixture amended with slag cement was studied. In the hydraulic test on triaxial apparatus, the effect of confining pressure, curing time and cement content on k is evaluated. In the hydraulic test on the flexible wall permeator, the effect of cement content on k is estimated and the long-term hydraulic conductivity performance is discussed, as well as the variations of EC and pH from the effluent. Potential mechanisms (e.g. volumetric strain, void ratio, etc.) that might influence the hydraulic performance are analyzed.

4.2 Materials and methodology

4.2.1 Properties of the soil and binders

Decomposed granite soil was used as the host soil, and the bentonite and slag cement were used as the binders. Different content of slag cement was applied for the amendment of soil bentonite mixtures (SB). Properties of these three materials were introduced in section 3.2.1 and the details of the mixing process of SB and cement amended SB (SB-C) have been described in section 3.2.2. In this chapter, the hydraulic conductivity test containing two parts, using triaxial apparatus and flexible wall, respectively. The details of the results will be introduced in the following sections.

4.2.2 Free swelling test

Free swelling tests were conducted to evaluate the effect of cement on the swelling of bentonite. In this chapter, the free swelling test was corresponding to ASTM D5890-19 (2019). The sodium bentonite powder was drying to constant weight at 105 ± 5 °C for at least 24 h. The reagents in free swelling test were distilled water (DSW), tap water (TW) and cement solution. The cement solution was used to investigate the swelling behavior of bentonite under relatively high electric conductivity (EC) and alkaline conditions (pH).

In the free swelling test, the cement solution had a cement concentration of 25, 50 and 75 kg/m³ (C25, C50, C75), which is the same as the cement to bentonite ratio in SB-C. The cement solution was prepared by blending DSW with different weight of slag cement and hydrated for 24 hours. Before the test, the solution was filtered by a 0.45- μ m filter paper. The passing percent for grain size > 0.075 mm of this bentonite is 90%, as according to JIS A1204 (2020).

During the free swelling test, the atmosphere conditions of this test were at a temperature of 21 ± 2 °C. The test was starting from weighing 2.00 ± 0.01 g of dried bentonite powder onto a weighing paper. Then, 90 mL of distilled water or filtered cement solution was added to a clean 100-mL graduated cylinder. 0.1-g increment of bentonite was dusted on the entire surface of reagent over a period of approximately 30 s. The bentonite particles were allowed to wet, hydrate, and settle to the bottom of the graduated cylinder for 10 min. Additional increments were added in periods of 10 min, allowing the bentonite to swell without air being trapped in, until the entire 2.00-g bentonite has been added.

After the final increment has settled, adhering particles from the sides of the cylinder (mostly at the scale of 90-mL mark) were rinsed into the reagent. The reagent volume was finally raised to 100-mL mark. The graduated cylinders were allowed to stand undisturbed for

24 hours, and the observed volumes of hydrated bentonite were recorded (to the nearest 0.5 mL), as well as EC and pH values of the reagents before and after the test.

4.2.3 Hydraulic conductivity test using flexible wall permeameter

For hydraulic conductivity test using flexible wall, the blended mixtures were scooped into a consolidation cell (with an inner height of 20 mm and a diameter of 60 mm) and consolidated under 39.2 kPa for 48 hours to minimize the volume change during permeation. After consolidation, the specimen in the consolidation cell was immersed in distilled water and saturated in a vacuum deaerator for at least two days. Table 4.1 summarizes the testing scheme in hydraulic conductivity test.

Table 4.1 Testing scheme of hydraulic conductivity test using flexible wall permeameter.

	Bentonite slurry (L/m ³)	Bentonite powder (kg/m ³)	Slag cement (kg/m ³)
SB-C0 (SB)			0
SB-C25	350	97.5	25
SB-C50			50
SB-C75			75

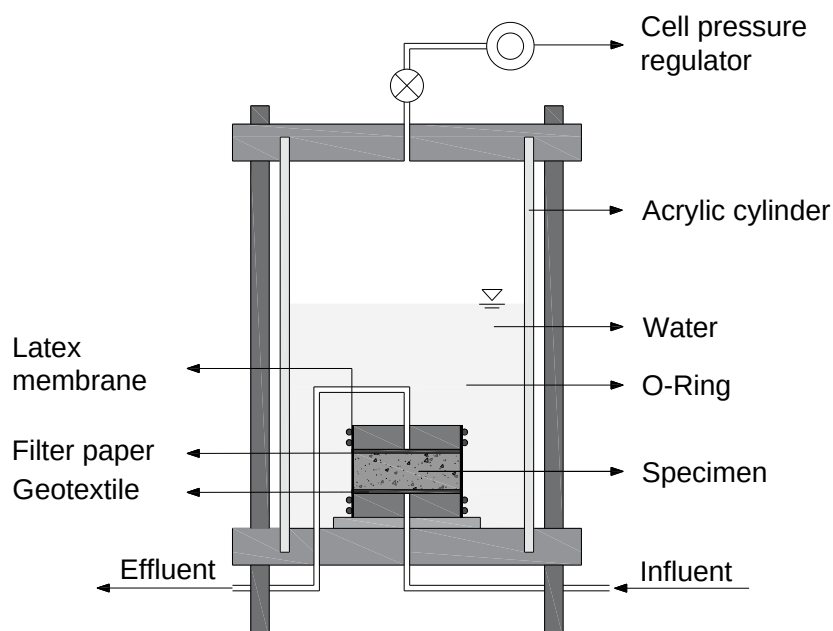


Fig. 4.1 Schematic diagram of flexible wall permeameter.

To investigate the hydraulic performance of SB-C specimens, four cases of flexible-wall hydraulic conductivity test as per ASTM D5084-03 (2011) were conducted at constant standard

temperature conditions (20 °C). To minimize the side-wall leakage, latex membrane was used in close contact outside the specimen with silicone grease spreading evenly on the contact surface (Black and Lee, 1973). After the consolidation and saturation process, the specimen was disassembled from the consolidation cell with no disturbance and installed on the flexible wall permeameter with a falling head system, as illustrated in Fig. 4.1. The cylindrical specimen was placed between saturated (i.e. deaerated) filter papers and geotextiles, which make the up-flow as capillary fluid for sufficient permeation. The experimental procedures of hydraulic conductivity test have been shown in pictures in Fig. 4.2.

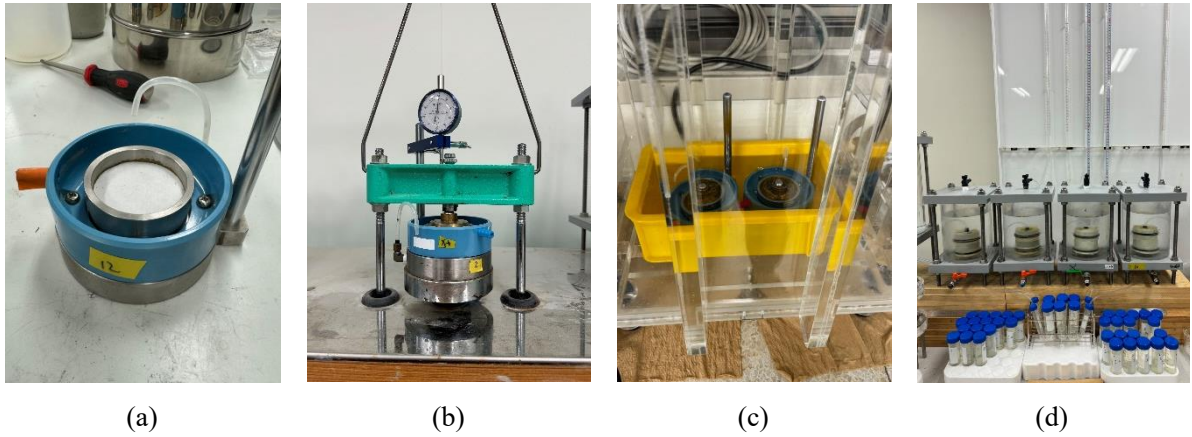


Fig. 4.2 Experimental procedures of hydraulic conductivity test.

* (a) Specimen preparation. (b) Consolidation process. (c) Saturation process. (d) Permeation process.

The hydraulic gradient during permeation was approximately 40-50, and distilled water was used as the permeant solution. Cell pressure (i.e. confining pressure) of 40 kPa was applied to minimize the volume change during permeation, which is equal to the consolidation stress (i.e. 39.2 kPa) in the pre-consolidation process to minimize the effect of confining pressure on the hydraulic conductivity during permeation. The hydraulic conductivity (k), electrical conductivity (EC) and pH value were measured through the permeation process. The hydraulic conductivity k can be calculated following Equation (2),

$$k = \frac{a \cdot l}{A \cdot \Delta t} \ln \frac{h_1}{h_2} = 2.303 \frac{a \cdot l}{A \cdot \Delta t} \log \frac{h_1}{h_2} \quad (4-1)$$

where k is hydraulic conductivity (m/s), a is cross-sectional area of the vertical standpipe (i.e., influent liquid) (m²), l is the height of specimen (m), A is the cross-section area of specimen (m²), Δt is elapsed time interval (s), and h_1 and h_2 are initial water head level and final water head level, respectively (m).

In this study, in an effort to evaluate the effect of cementation on the hydraulic barrier performance of SB-C, the permeation was continued as the pore volumes of flow (PVF) were higher than 2, and the volumes of the influent and the effluent were almost equal.

4.2.4 Hydraulic conductivity test using triaxial apparatus

In Chapter 3, the SB and SB-C specimens were consolidated under triaxial condition, applied through the back pressure and cell pressure. After the specimens have been consolidated under different confining pressure, the hydraulic conductivity was conducted to investigate the influence of cement content, curing and consolidation. The tested specimens are summarized in Table 3.3 in section 3.2.2.

As according to ASTM D5084-03 (2011), the hydraulic conductivity k in an increasing tailwater apparatus can be calculated from (4-2),

$$k = \frac{a_{in} \cdot a_{out} \cdot l}{(a_{in} + a_{out}) \cdot A \cdot \Delta t} \ln \frac{h_1}{h_2} \quad (4-2)$$

where a_{in} is the cross-sectional area of the reservoir containing the influent/inflow liquid (m^2), and a_{out} in the cross-sectional area of the reservoir containing the effluent/outflow liquid (m^2). After the completion of consolidation, the volume of the specimens V_0 changed to V , related to the volumetric strain ε_v (%) (i.e. bulk strain), and the height of specimen l have the relationship with the initial height l_0 , as expressed in (4-3),

$$V = V_0(1 - \varepsilon_v); \quad l = l_0(1 - \varepsilon_1) \quad (4-3)$$

Then, the average diameter of the sample becomes d , the average cross-sectional area A of the specimen comes to (4-4),

$$A = \pi d^2 = \frac{A_0(1 - \varepsilon_v)}{(1 - \varepsilon_1)} \quad (4-4)$$

where A_0 is the initial cross-sectional area after curing for 7 or 28 days, and ε_1 is the axial strain after the consolidation process.

Herein, the lateral strain ε_3 was also considered with the relationship of volumetric strain ε_v . As the height of the specimen l is double of the diameter d (i.e. $l = 100$ mm, $d = 50$ mm), and the Poisson's ratio μ is normally considered as 0.5 for saturated clay. Therefore, in this chapter, the ε_1 can be considered as equal to ε_2 , which also considering the volume conservation

in triaxial consolidation state. And the relationship between ε_v and ε_1 comes to (4-5),

$$(1 - \varepsilon_v) = (1 - \varepsilon_1)^3 \quad (4-5)$$

Based on the above explanations, the hydraulic conductivity k in the hydraulic conductivity test using triaxial apparatus after consolidation process can be evaluated as (4-6),

$$k = \frac{a_{in} \cdot a_{out} \cdot l_0 \cdot (1 - \varepsilon_1)^2}{(a_{in} + a_{out}) \cdot A \cdot (1 - \varepsilon_v) \cdot \Delta t} \ln \frac{h_1}{h_2} \quad (4-6)$$

4.3 Results and discussion

4.3.1 Effect of cement on the free swelling performance of bentonite

The results of free swelling test are summarized in Fig. 4.3. The highest swelling volume was in the DSW, that the swelling volume was approximately 23.2 mL/2g. The swelling volume decreased to 21.8 mL/2g in the tap water. An obvious decrease trend in bentonite swelling volume occurred with the augment of cement content from 25 kg/m³ to 75 kg/m³. The swelling volume in cement reagent were all lower than 22.0 mL/2g, as the lowest was 19.7 mL/2g in 75 kg/m³. It is indicated that solutions with relatively high cement concentration may reduce the swelling performance of bentonite. Previous studies reported that the pH environment (i.e. alkaline environment) and potential cations (e.g. Ca²⁺, Na⁺, etc.) concentration may affect the swelling performance of the bentonite (Anh et al., 2017).

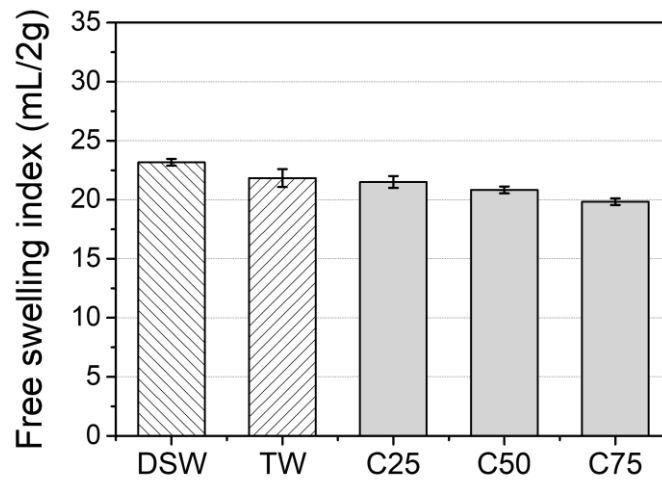


Fig. 4.3 Free swelling volume of bentonite in different reagents.

In this chapter, to investigate the mechanism of this phenomenon, the EC and pH values before and after the test have been measured and summarized in Fig. 4.4 and Fig. 4.5. In Fig. 4.4, the initial pH of DSW was 7, and the pH of TW was almost 7.6. The increase of the pH can be attributed to the hydration of bentonite, as the increasing value was similar in DSW and TW. In cement solutions, the pH variation is minor, that all of the initial pH is between 12.1 to 12.3. And the final pH decreased slightly to the range of 11.8 to 12.2.

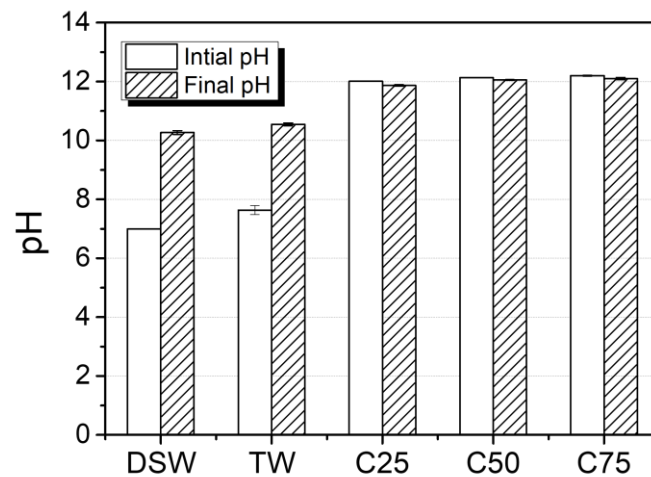


Fig. 4.4 The variation of pH in the free swelling test.

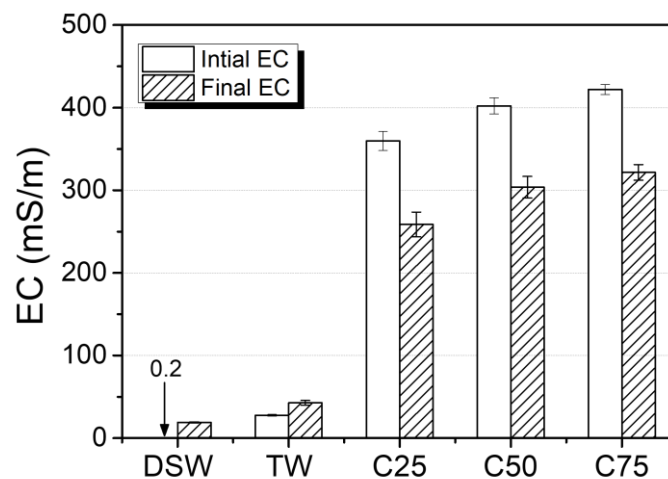


Fig. 4.5 The variation of EC in the free swelling test.

From previous studies, the bentonite alteration can be affected by cation type and the concentration as well as the pH of a solution (Fernández et al., 2014). However, the pH range may not change the montmorillonite fraction of the bentonite after reacting with alkaline solutions (pH = 8.4-13.1), regardless of the solution type (Anh et al., 2017). In the cement

solutions, high pH environments contributed by the cement hydration through highly alkaline materials such as portlandite Ca(OH)_2 , sodium, and potassium hydroxides. The bentonite alteration in these solutions, which involves the dissolution of montmorillonite and the precipitation of secondary minerals, occurs during interactions with hyper-alkaline solutions, significantly affect the EC values, as illustrated in Fig. 4.5.

On the contrary, the swelling volume of bentonite decreased with the increase of EC, suggesting that the swelling performance was primarily affected by the cement solution (e.g. 25, 50 and 75 kg/m^3) concentration rather than the pH value. Such findings can also be found in previous studies, that an increase in electrical conductivity (EC) may restrict swelling performance. As cations in the solution can be tightly adsorbed and held onto the surface of negatively charged clay particles, the adsorbed cations are highly concentrated near the surfaces of particles, hence could diffuse away to equalize concentrations throughout the pore fluid (Takai et al., 2016). The EC, as an indicator for electrolyte concentration of the solutions, is the major effect on the swelling performance of bentonite.

4.3.2 Consolidation state of SB and SB-C specimens

The consolidation process refers to the phase where the soil sample undergoes increasing confining pressure, leading to a gradual consolidation of the soil structure. During the consolidation process, the primary mechanisms include pore compression and deformation, as well as the rearrangement of soil particles within the specimen.

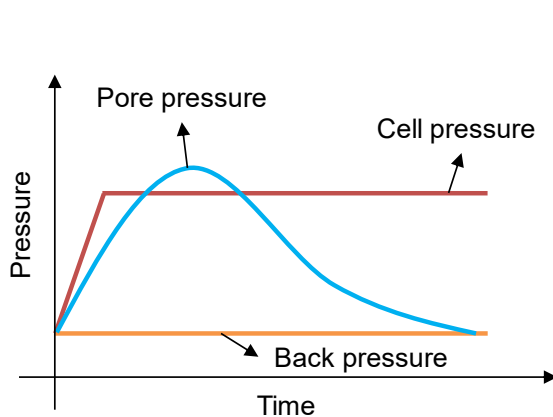


Fig. 4.6 The variation of pore pressure during consolidation process.

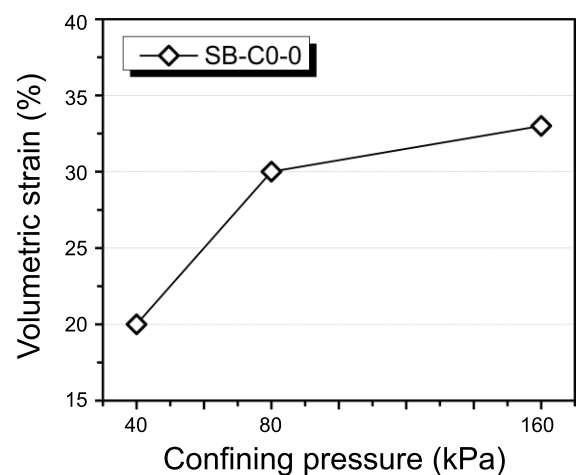


Fig. 4.7 Volumetric strain of SB after consolidation in the triaxial test.

As shown in Fig. 4.6, pore pressure typically increases during the consolidation process as the confining pressure increases. This happens because the increase in confining pressure causes the compression of soil pores, forcing pore water drainage out, arrangement of soil particles and thus increasing pore pressure. As the consolidation proceeding, the rate of increase in pore pressure gradually slows down until it reaches a state of equilibrium, in other words, approaching the back pressure. In this chapter, the consolidation was completed in the triaxial compression apparatus and one-dimensional consolidation apparatus.

After the completion of consolidation, the volume of the specimen will decrease, which is closely related to the consolidation stresses. In the triaxial apparatus, the volumetric strain of SB after consolidation has been shown in Fig. 4.7. Under 40 kPa confining pressure, the volumetric strain was 20%. As the confining pressure increasing to 80 kPa, the volumetric strain increased to 30%. The volume change under 160 kPa was approximately 33%. The volumetric strain of SB-C after consolidation has been shown in Fig. 4.8. The volume change was ranging from 0.1% to 3.0%, which indicates the strong effect of the cementation, making the voids difficult to be consolidated. With increasing cement content and curing days, the influence of confining pressure diminished. In SB-C75 cases, due to the high cementation of the specimens, the impact of consolidation was minimal.

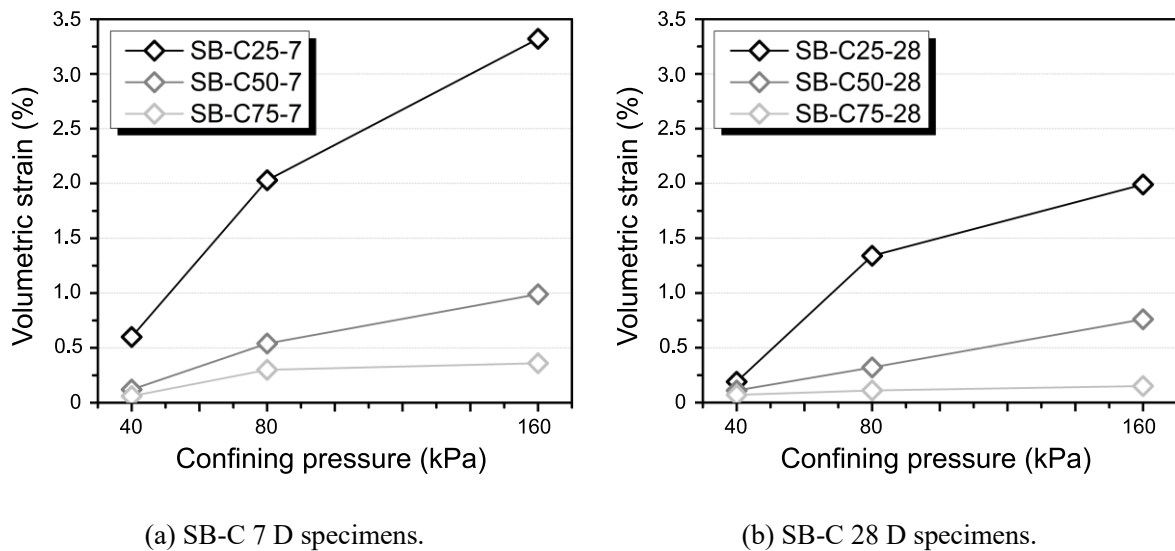


Fig. 4.8 Volumetric strain of SB-C after consolidation in the triaxial test.

In the flexible wall permeator, the void ratio before and after consolidation has been measured and summarized in Fig. 4.9. The decrease in void ratio reflects the compaction of the soil structure and the reduction in pore space available within the specimen. The initial void

ratio of all the specimens were around 1.12. After the completion of consolidation, the decline in void ratio decreased with the augment of cement content, which is similar to volumetric strain variation characteristics in Fig. 4.7 and Fig. 4.8.

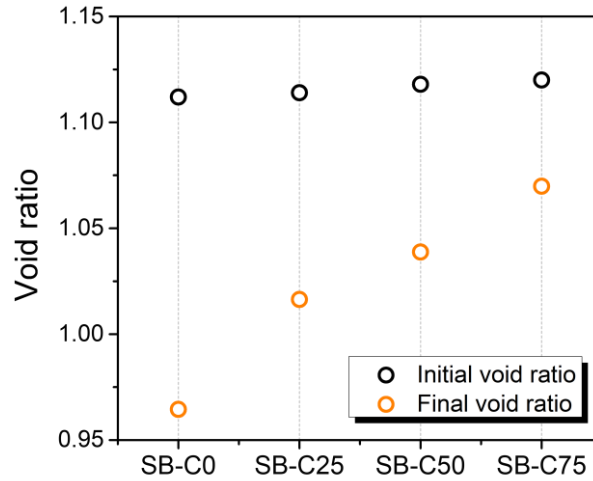


Fig. 4.9 Variation of void ratio before and after permeation in the flexible wall permeator.

4.3.3 Hydraulic performance of specimens using flexible wall permeameter

The hydraulic conductivity k is one of the most important engineering properties of soil cement bentonite (SCB) walls, as the k values varies within the range of 5.0×10^{-9} m/s to 1.0×10^{-8} m/s after 28 D curing (Ryan and Day, 2002). Numerous studies suggested that the permeability of typical soil bentonite cutoff wall could be controlled as low as 1.0×10^{-9} m/s (Gleason et al., 1997; Malusis and McKeehan 2013; Takai et al., 2016). In this study, the SB-C specimens for hydraulic conductivity were permeated without curing, to simulate the actual scenario in the SCB wall construction using the two-phase TRD method.

The variations of k in SB and SB-C versus pore volumes of flow has been illustrated in Fig. 4.10. Herein, pore volumes of flow (PVF) is the cumulative flow volume normalized by the pore volume of each specimen after consolidation. It is obvious that the variation characteristics of these four SB and SB-C specimens were totally different. Initially, the k of SB-C0 floating within the first 0.5 PVF, within which the bentonite adsorbs influent water molecules with the formation of diffuse double layer and osmotic swelling because it did not fully swell during the consolidation process. After permeated for 1.0 PVF, the k value comes to be stable at approximately 7.4×10^{-11} m/s, which means that the immovable water (i.e. adsorbed water in the bentonite double layer and immoveable water between soil particles) did not contribute to the water flow.

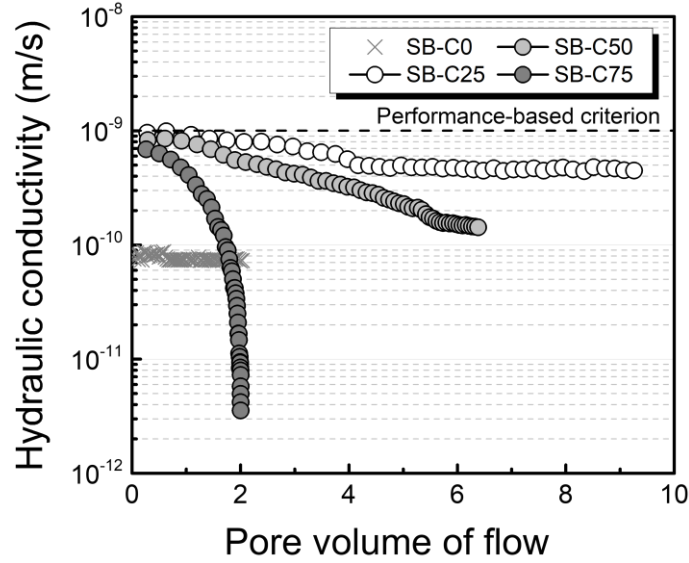


Fig. 4.10 Hydraulic conductivity k versus pore volumes of flow.

The k values of SB-C were similar (6.9×10^{-10} to 9.6×10^{-10} m/s) at the beginning of the permeation (SB-C25 > SB-C50 > SB-C75), which means that the primary cement hydration effect from the beginning was relative to the cement content, as the void ratio of SB-C50, SB-C75 and SB-C100 after consolidation were 1.01, 1.03 and 1.07, respectively. With the progression of permeation, slight increase in k of SB-C25 and SB-C50 can be witnessed within the first PVF. After the consolidation and saturation process, the immovable water was adsorbed onto the bentonite and cementitious material through hydration. When the permeation started, the secondary hydration of cement and cementitious material occurred, and some of the immovable water adsorbed with the water flow, leading to the slight increase of k . After 1.0 PVF, the k value kept decreasing and reached a constant after 4.0 PVF, as the k was 4.6×10^{-10} m/s. The variation trend in SB-C50 was different that, when PVF > 1.0, a gradual decreasing can be seen until the termination of the permeation at 6.4 PVF. In SB-C75, a sharp decrease in k from 10^{-10} to 10^{-12} , as the total PVF was just exceeding 2.0. The k value lower than 10^{-12} implied small pore spaces in the specimen, which impeding water infiltration. These phenomena can be explained by the secondary cementitious materials in the continuous hydration throughout the permeation, which enhanced the filling of pore space and the bonding between solid particles, as a result, the k value declined consistently (Katsumi et al., 2008; Ryan and Day 2002; Carreto et al., 2016;).

The results support that the k values of the amended SB-C with slag cement content of 50, 75, 100 kg/m³ present sufficient k values lower than 10^{-9} m/s, which is the performance-based criterion in this study. This criterion was determined as according to the requirements of

hydraulic conductivity for vertical cutoff walls constructed using TRD method. In the United States, the hydraulic conductivity of SB backfill was measured typically between 1×10^{-9} and 1×10^{-10} m/s in laboratory tests, and the basic value of 1×10^{-9} m/s is required for different soil and ground water conditions (Evans., 2007; Opdyke and Evans., 2005). In Japan, Enforcement Regulations of Soil Contamination Countermeasure Law regulated ‘hydraulic barrier layer’ as a layer with k value less than 1×10^{-7} m/s and thickness larger than 5.0 m. Given that the width of the SB walls ranges from 0.55 m to 1.5 m, and 0.55 m (i.e. 550 mm) representing as a typical width with corresponding depth around 15 m (Evans., 1993; Opdyke and Evans., 2005; Katsumi, et al., 2008). The hydraulic conductivity $k = 1.0 \times 10^{-7}$ m/s with a cutoff wall thickness $L = 5.0$ m is equivalent with the $k \approx 1.0 \times 10^{-8}$ m/s with $L = 0.55$ m in the same hydraulic condition. By considering the travel time of contaminants under the same ground water difference inside/outside the cutoff walls and effective porosity, the criterion was determined as 1.0×10^{-9} m/s (Takai et al., 2016).

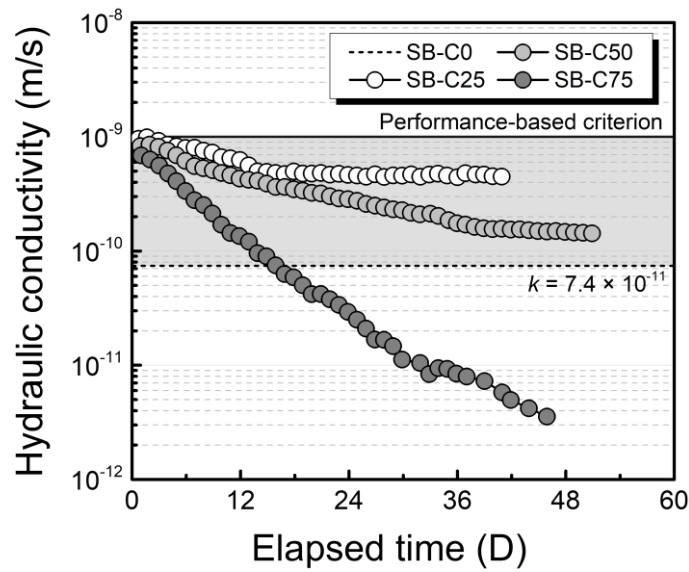


Fig. 4.11 Hydraulic conductivity k versus elapsed time.

Due to the hydration occurring over time, the completion of hydration reactions usually takes 90 days or more in CB walls depending on the cement content, as a result, the hydraulic conductivity continues to decline over time (Opdyke and Evans., 2005). This phenomenon can also be found in this study that the decrease in k values is governed not by PVF but by elapsed time (Takai et al., 2019). The variation of k versus elapsed time have been shown in Fig. 4.11. Given that SB-C0 exhibited a virtually constant k during permeation, the k of SB-C0 (i.e. $k = 7.4 \times 10^{-11}$ m/s) has been drawn as one single dash line in Fig. 4.11, as a comparison for the

hydraulic performance of the SB-C. The pictures of specimens after test are shown in Fig. 4.14.

As illustrated in Fig. 4.11, the decrease in k values occurred at different rates, contingent on the cement content. The variation trend of k in SB-C25 with permeation time is similar to the relationship with PVF depicted in Fig. 4.10, that the equilibrium of k was reached at around 15 days, accompanied by a PVF of 4.0. In SB-C50 case, the total permeation time was 50 days, and the decrease in k value decelerated after 40 days, as the k decreased from 1.5×10^{-11} to 1.4×10^{-11} m/s. The total permeation time of SB-C75 was 46 days, as the k value was in almost linear decreasing regression with the permeation time. The final k of SB-C75 was 3.6×10^{-11} m/s, implying the significant effect of 75 kg/m^3 cement content, that contribute to greater hydration and extremely low hydraulic conductivity.

Fig. 4.12 plots the variation of pH and EC of the effluent versus PVF during permeation. Generally, with the augment of PVF, the progression of cement hydration promotes the solidification, as more secondary cementitious materials had been generated. As a result, the content of desolated cement particle decreased, thus the pH and EC decreased in SB-C cases correspondingly. In Fig. 4.12(a), the effluent pH of SB-C0 was stable around 9.0 through the permeation, which is slightly lower than the pH of bentonite slurry used in this chapter (i.e. $\text{pH} \approx 10.0$). For SB-C specimens, the initial pH was similar between 12.3 to 12.5. As the PVF increased, the pH in SB-C50 and SB-C75 decreased to 8.96 and 9.26 at the final PVF. The pH equilibrium in SB-C25 was achieved at $\text{PVF} = 8.0$, with the equilibrium pH value around 10.0.

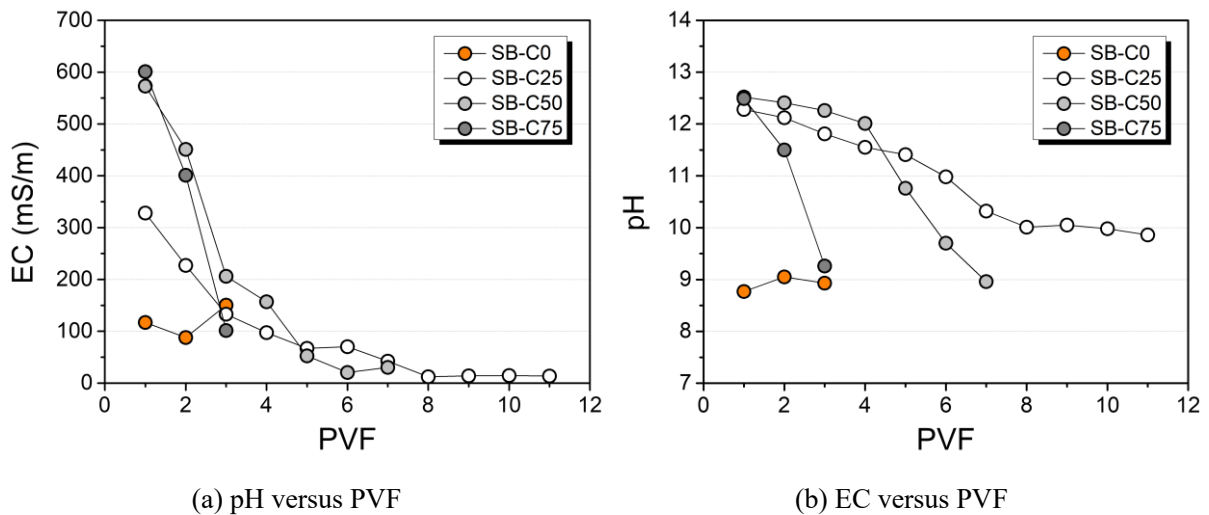


Fig. 4.12 Variation of pH and EC in the hydraulic conductivity test.

In Fig. 4.12(b), the effluent EC of SB-C0 was flexible from 90 to 150 mS/m, which is higher than the EC of bentonite slurry (i.e. $\text{EC} \approx 88 \text{ mS/m}$). For SB-C specimens, the initial EC

of SB-C50 and SB-C75 was similar, at 573 and 601 mS/m, respectively. However, the initial EC of SB-C25 was relatively lower, at 328 mS/m. As the PVF increased, the pH in SB-C50 and SB-C75 decreased to 30 and 101 mS/m at the final PVF. The EC equilibrium in SB-C25 was accomplished at the same PVF, where pH equilibrium was also achieved, with the equilibrium EC value around 13 mS/m. In SB-C25, the final EC was lower than that of SB attributed to the formation of rigid crystalline structure, which limited the migration of cations and thus reducing EC. On the contrary, the pH was higher than that of SB, implied the release of alkaline substances (e.g. Ca(OH)_2) through the permeation.

An important finding is that the equilibrium of k in SB-C25 was accompanied by a PVF of 4.0, but the equilibrium of EC and pH was reached at PVF of 8.0. The EC and pH decreased from PVF of 4.0 to 8.0, indicating the continuous hydration of cementitious materials, within a stable pore structure (i.e. void ratio). The cement hydration may gradually slow down and then almost completely stabilized at PVF = 8.0.

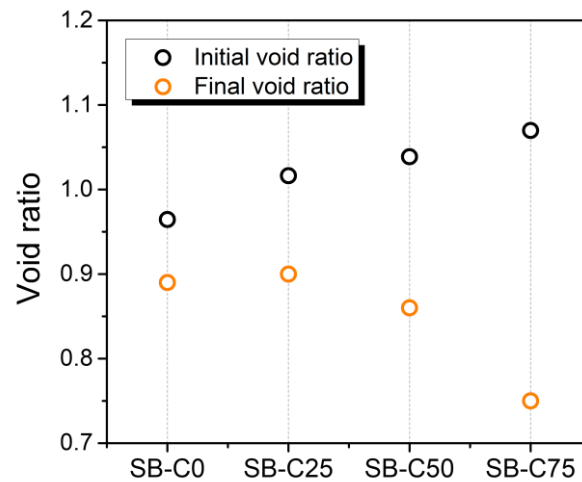


Fig. 4.13 Variation of void ratio before and after the hydraulic conductivity test.

Another important finding is that the final pH of SB-C50 and SB-C75 was relatively low, and similar to the pH of SB-C0. The hydration of cementitious materials filled the voids, as a result, decreased the void ratio. As shown in Fig. 4.13, the void ratio after permeation of these four specimens were 0.89, 0.90, 0.86 and 0.75, respectively. The void ratio of SB-C50 and SB-C75 was relatively low, and the alkaline substances was difficult to be released from the dense pore structure. At the same time, the cations migration was also restricted, which leading to the decrease in the EC. There was a tendency for an equilibrium in EC in SB-C50, along with a similar trend towards equilibrium in the k value, as the final k value was 1.4×10^{-11} m/s.

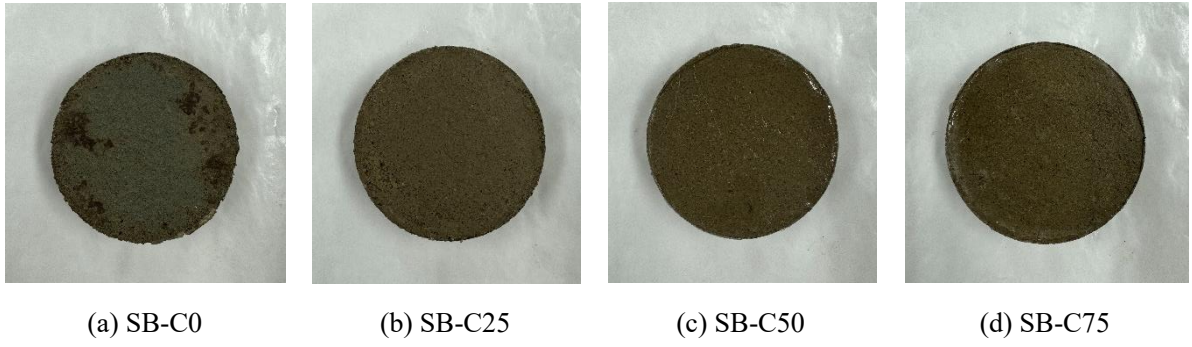


Fig. 4.14 Pictures of specimens after flexible wall permeation test.

4.3.4 Hydraulic performance of specimens using triaxial apparatus

The hydraulic conductivity k of the specimens using triaxial apparatus has been plotted in Fig. 4.15. The k of SB-C0-0 is notably lower compared to SB-C specimens, measuring less than 1.0×10^{-10} m/s (i.e. 8.3×10^{-11} m/s). The confining pressure affect the k of SB-C0-0. Under 80 kPa and 160 kPa of confining pressure, the k decreased to 6.2×10^{-11} m/s and 5.3×10^{-11} m/s respectively. The k of all the SB-C specimens were lower than 1.0×10^{-9} m/s, which is the performance-based criterion as introduced in section 4.3.3. As the cement content increased, the k of SB-C decreased in both 7 D and 28 D specimens. However, the effect of confining pressure was diminished under 80 and 160 kPa, especially in highly cemented 28 D specimens. There was almost no variation of the k in SB-C75-28 under each confining pressure.

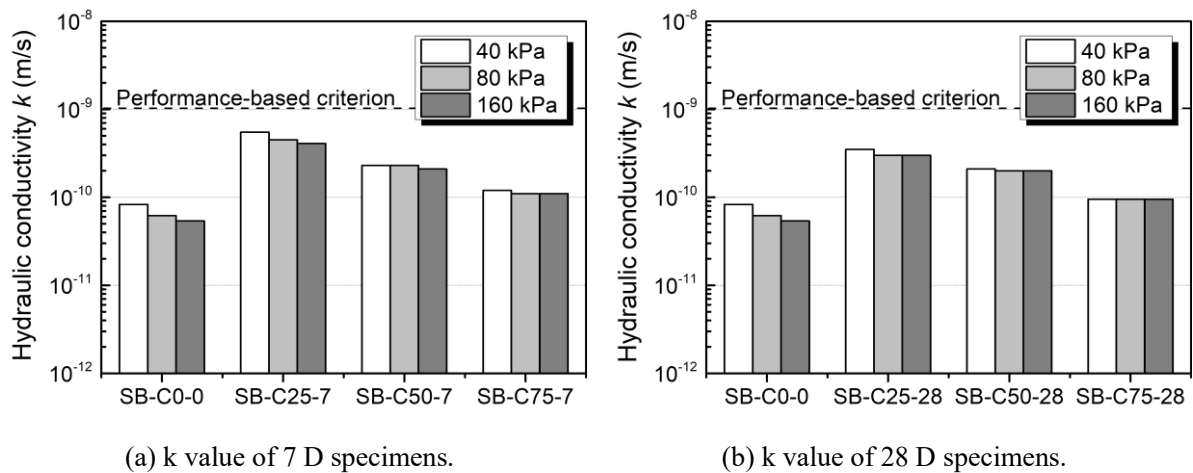


Fig. 4.15 Hydraulic conductivity k values in triaxial apparatus.

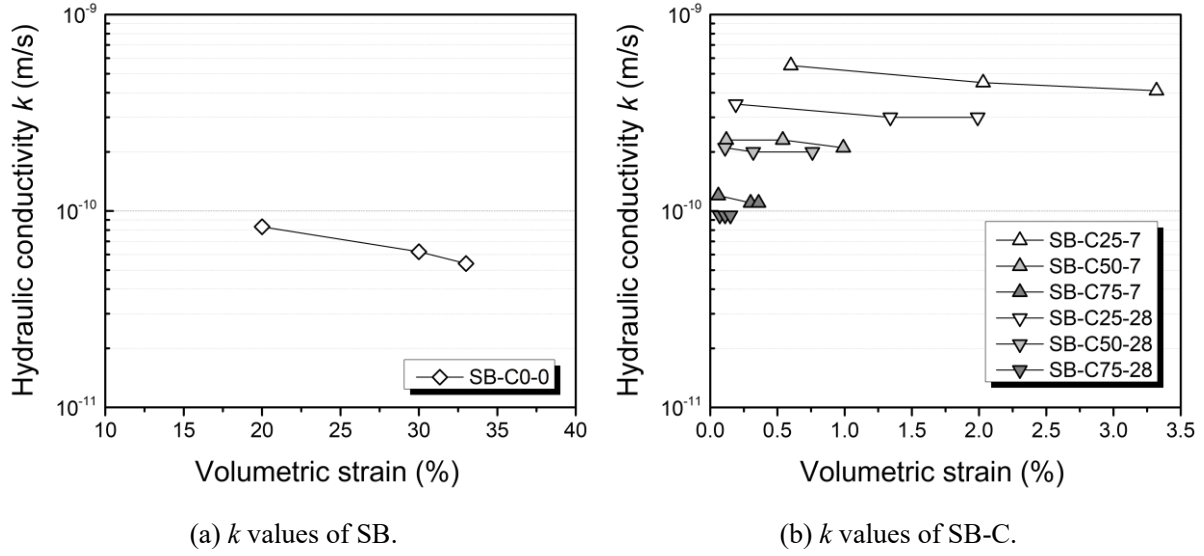


Fig. 4.16 Relationship of k values with volumetric strain.

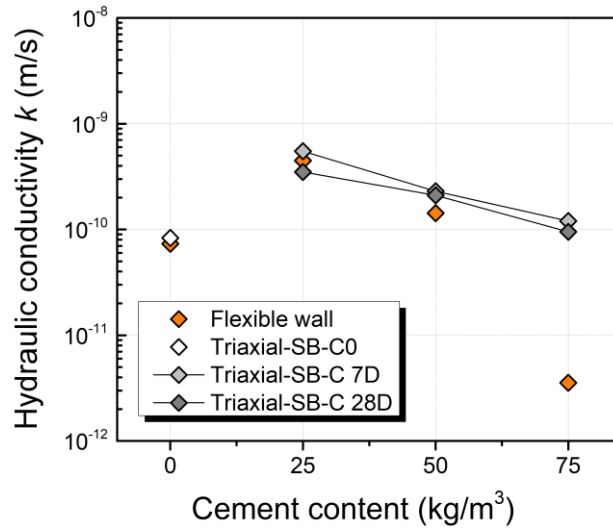


Fig. 4.17 Comparison of k values in using triaxial apparatus and flexible wall permeator.

To estimate the effect of consolidation on k value, the relationship of k values with volumetric strain after consolidation has been plotted in Fig. 4.16. It is obvious that the curing has a positive impact on the k values in all the SB-C specimens, as shown in Fig. 4.16(b). The effect of consolidation has a linear relation with the k of SB-C0-0 and SB-C25-7. The effect diminished in SB-C50 and SB-C75 that, the volumetric strain is within 1%, indicating the minor influence of consolidation regardless of confining pressure. Therefore, when the cutoff wall is constructed using cement, the wall may not be highly compressed horizontally (i.e. small horizontal deformation). Fig. 4.17 compared the hydraulic conductivity results from triaxial apparatus and flexible wall apparatus. Generally, the k values obtained in triaxial test are higher,

except for the SB-C0-0. This phenomenon can be explained by the hydration process of SB-C through permeation, which leading to the extremely lower k values in SB-C25 and SB-C75.

4.4 Conclusions for this chapter

In this chapter, the swelling performance of the bentonite under the effect of cement content was estimated. Also, the hydraulic performance of soil bentonite mixture amended with slag cement was studied based on the results of hydraulic conductivity test using triaxial apparatus and flexible wall permeator. Potential mechanisms (e.g. volumetric strain, void ratio, etc.) that might influence the hydraulic performance are analyzed. The main results obtained in this chapter can be summarized as follows:

- (1) The free swelling volume of bentonite in the distilled water was approximately 23.2 mL/2g, and 21.8 mL/2g in the tap water. The swelling volume decreased in cement solutions, and decreased as the cement content increased, as the EC was increased significantly. The EC has a linear effect on the swelling performance of bentonite, while pH is stable at around 12.0 before and after the test.
- (2) The effect of consolidation is significant in SB specimens as the volumetric strain was over 20%. The volumetric strain in SB-C specimens were lower than 3.5%. The confining pressure affect the consolidation process that the volumetric strain increased with the augment of confining pressure.
- (3) In the hydraulic conductivity test using flexible wall permeator, the k value of SB-C0 was obtained with k value of 7.4×10^{-11} m/s. The decrease of k values in SB-C specimens is governed not by PVF but by elapsed time. The primary k values of SB-C were similar (6.9×10^{-10} to 9.6×10^{-10} m/s) at the beginning of the permeation (SB-C25 > SB-C50 > SB-C75). With the permeation, the k of SB-C25 reached a constant value of 4.5×10^{-10} , while k values of SB-C50 and SB-C75 kept decreasing.
- (4) In the hydraulic conductivity test using flexible wall permeator, the variation of pH and EC in SB was almost stable, indicating a stable state of bentonite swelling and water flow through the specimen. In SB-C specimens, only SB-C25 obtained equilibrium pH and EC. The hydration of cement with influent filled the pore space, as a result, void ratio decreased and more immoveable water generated, leading to extremely low k values as well as sharp decrease of pH and EC.
- (5) In the hydraulic conductivity test using triaxial apparatus, the k values of SB-C0-0 is lower than all the SB-C specimens. The cement content and curing time are the major effect on

the k . The effect of confining pressure (i.e. consolidation effect, volumetric strain) on the k values is significant in SB-C0 and SB-C25, while minor effect was found when the cement content increased to 50 and 75 kg/m³.

- (6) The measured k values are relatively higher using triaxial apparatus, because there was no permeation in the specimens, which is beneficial for the sufficient osmotic swelling in SB specimens and crucial for the hydration of cementitious materials in SB-C specimens. Finally, the slag cement amended soil-bentonite mixtures is considered to have an enough hydraulic barrier performance, as compared to the performance-based criterion (1.0×10^{-9} m/s) in this chapter.

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Chapter 5

Adsorption performance of bentonite on gaseous mercury

5.1 General remarks

The mercury waste is treated by excessive volume of sodium sulfate, which may lead to re-dissolute of mercury. To solve this issue, Fe^{2+} solution (FeSO_4) will be used to cover the excessive mass of Na_2SO_4 and confirm the stabilization/solidification (S/S) of mercury waste. As impermeable layer, the bentonite is highly compacted to obtain extremely low hydraulic conductivity. However, as shown in Fig. 5.1, in cases of rainfall, changeable groundwater and long-term permeation, groundwater will flow into the bentonite layer, as a result, affect the effectiveness (MOEJ, 2021). In this circumstance, the mercury, in the form of element phase, will evaporate from the treated Hg waste, and pose threat to the environment. For long-term safety and reliability of mercury waste landfill, it is vital to investigate the adsorption capacity of impermeable bentonite layer, and also the swelling performance of bentonite when exposed to ferrous ions.

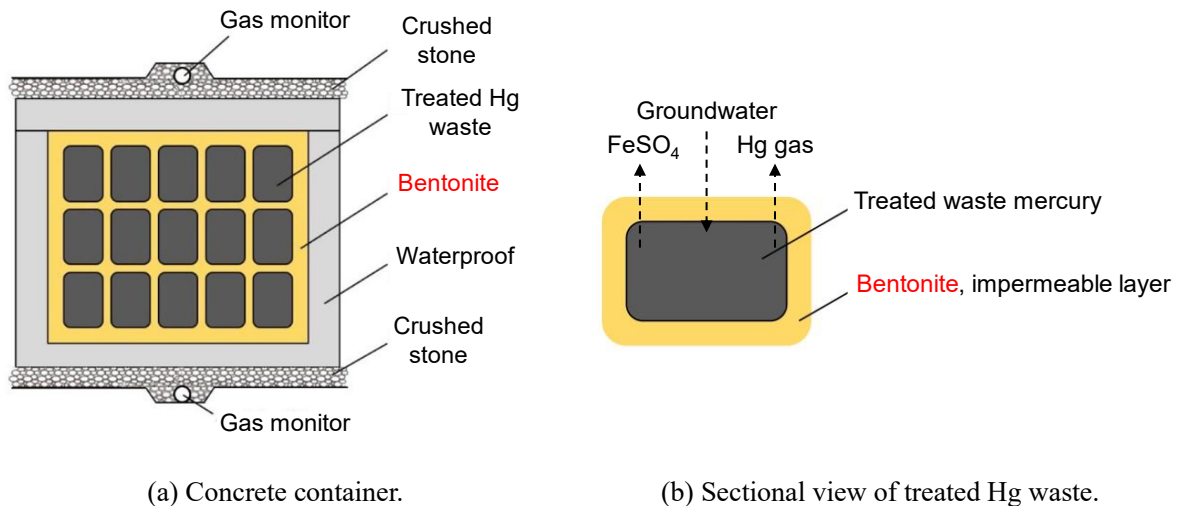


Fig. 5.1 Schematic of concrete container in mercury waste landfill.

In this chapter, the fundamental properties of three types of bentonites were estimated. To investigate the crystal structure and mineralogy of bentonites, X-ray powder diffraction test was conducted. A series of compaction tests were conducted to evaluate the optimal water

content and corresponding maximum dry density. Flexible wall hydraulic conductivity tests were conducted to determine the hydraulic performance of compacted bentonite.

Furthermore, the swelling performance of bentonites in ferrous solution were studied in normal condition and anaerobic condition, which is simulated through Nitrogen (N₂) bubbling and usage of high reduction reagent (i.e. Hydrazine, N₂H₄). Potential mechanisms (e.g. pH, Eh etc.) that might influence the swelling performance are analyzed. Finally, the adsorption capacity of two bentonites (sodium and calcium bentonite) were studied through constant volume adsorption tests. The effect of water content was estimated, and the distribution coefficient was discussed.

5.2 Materials and methodology

5.2.1 Properties of bentonite

In this research, three types of bentonites were used, designated as Turkey, Hokuhyo, and Geoclay. These bentonites were supplied from HOJUN Co., Ltd., and separated in two forms. One is original crushed bentonite, as the grain size distribution curve by JIS A1204 (2020) has been presented in Fig. 5.2. Another is powdered bentonite processed from the crushed bentonite, and the passing percent of grain size > 0.075 mm is over 90%.

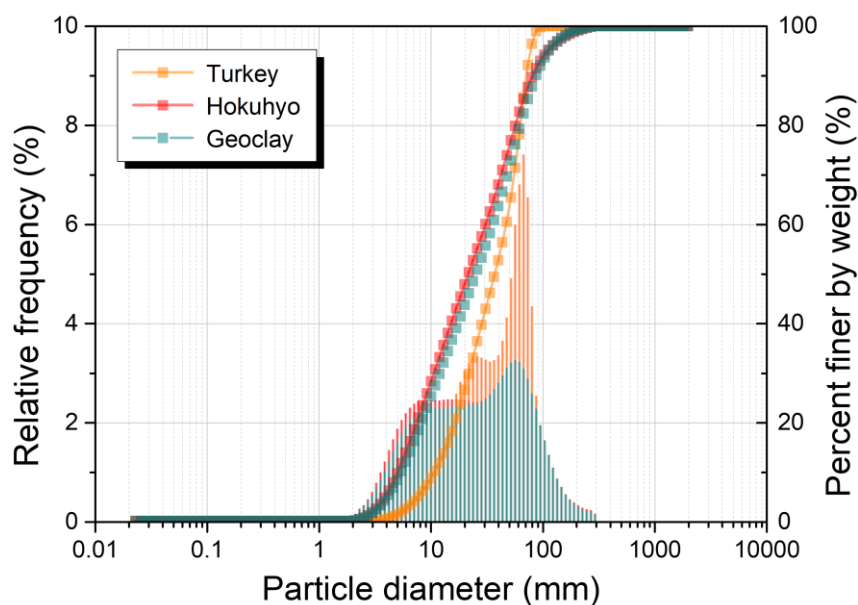


Fig. 5.2 Grain size distribution curve of crushed bentonites.

The fundamental properties of the bentonites have been shown in Table 5.1. The particle density of bentonite was measured in accordance with JIS A1202 (2020). Liquid and plastic

limits were measured in accordance with JIS A1205 (2020). The methylene blue adsorption value was measured using methylene blue titration experiments in accordance with JIS Z2451 (2019). The specific surface area was estimated by using a BET surface area and pore size analyzers (NOVA Touch Serie, Quantachrome Instruments) as according to JIS Z8830 (2001). The cation exchange capacity was measured using the Schollenberger method in accordance with JGS 0261 (2009).

Note that all properties of each type of bentonite were obtained from the same batch of powdered bentonite. The respective fundamental properties demonstrate that three bentonites used in this study have different physicochemical properties. According to the CEC (Cation exchange capacity) results, Turkey, Hokuhyo and Geoclay are classifiable as sodium-type, sodium/calcium-type and calcium-type bentonite. To investigate the crystal structure and minerology, all bentonites were analyzed using X-ray powder diffraction with Cu radiation.

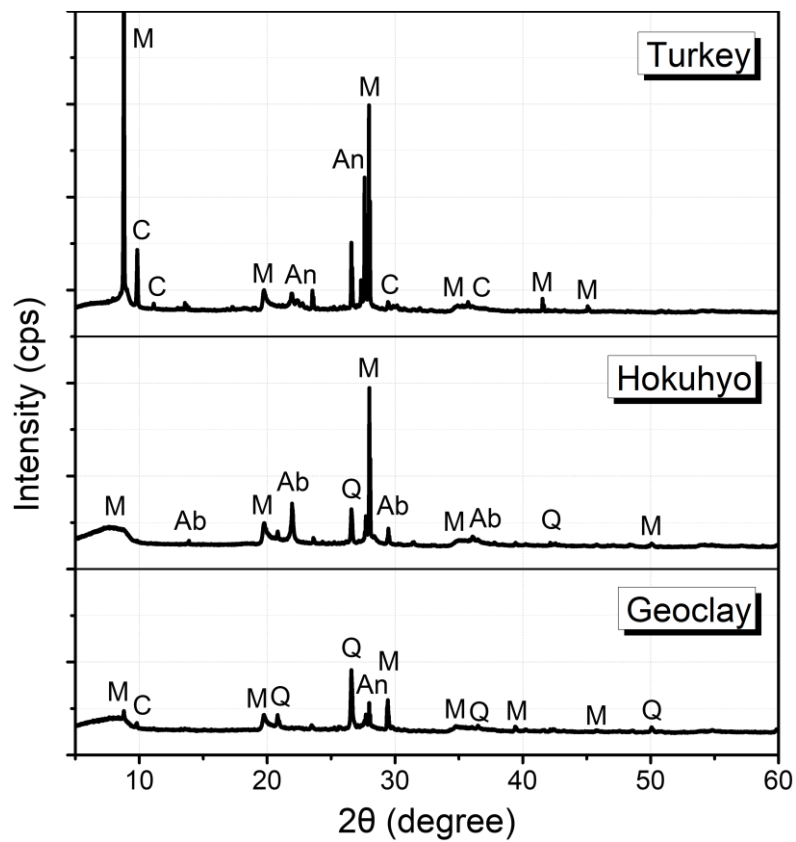
Table 5.1 Fundamental properties of three kinds of bentonite.

Index	Turkey	Hokuhyo	Geoclay	Testing method
Particle density (Mg/m ³)	2.62	2.68	2.67	JIS A1202 (2020)
Liquid limit (%)	522.2	439.5	396.1	JIS A1205 (2020)
Plastic limit (%)	27.8	36.4	27.0	JIS A1205 (2020)
Plasticity index	494.4	403.1	369.1	—
Specific surface area (m ² /g)	44.4	27.6	37.9	JIS Z8830 (2001)
Methylene blue adsorption value (meq/100 g)	71.5	92.5	74.0	JIS Z2451 (2019)
Cation exchange capacity (meq/g)	0.974	1.237	1.537	JGS 0261 (2009)
Na ⁺ exchange capacity (meq/g)	0.451	0.517	0.374	—
Ca ²⁺ exchange capacity (meq/g)	0.046	0.008	0.010	—
K ⁺ exchange capacity (meq/g)	0.407	0.548	1.010	—
Mg ²⁺ exchange capacity (meq/g)	0.070	0.164	0.143	—

Figure 5.3 shows X-ray diffraction patterns of the respective bentonites. Turkey is mainly composed of montmorillonite and clinoptilolite. Hokuhyo and Geoclay were mostly composed of quartz and montmorillonite. Turkey and Geoclay contained additionally the clay mineral of Anorthite, while Hokuhyo containing Albite.

Optimal water content (obtained with Maximum dry density) is crucial for compacted bentonite layer. In this chapter, the optimal water content was obtained through the standard compaction test following c method in JIS A1210 (2020). The compaction curves of the

bentonites have been plotted in Fig. 5.4. Based on Fig. 5.5, the natural water content and compaction properties of bentonites have been presented in Table 5.2.



*M, Montmorillonite; Q, Quartz; C, Clinoptilolite; An, Anorthite; Ab, Albite.

Fig. 5.3 X-ray diffraction patterns of bentonites.

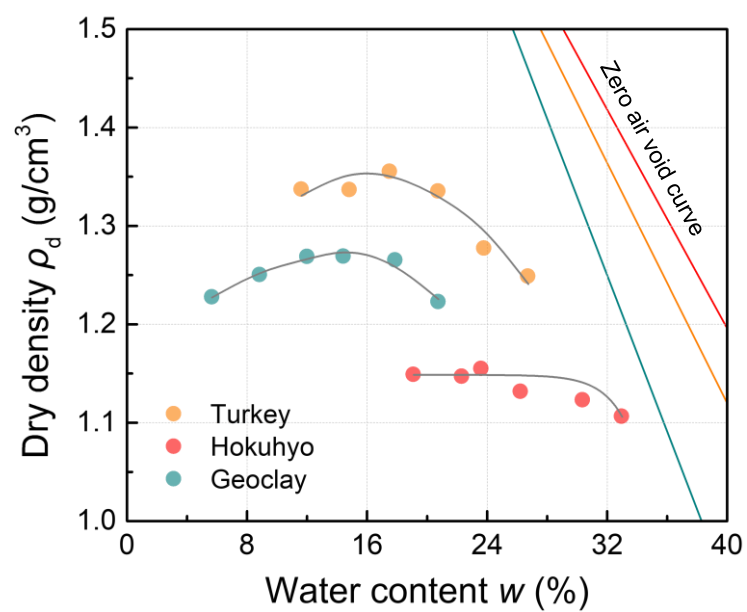


Fig. 5.4 Compaction curves of bentonites.

Table 5.2 Natural water content and compaction properties of bentonites.

	Unit	Turkey	Hokuhyo	Geoclay
Natural water content w_n	%	10.4	10.5	9.5
Optimum water content w_{opt}	%	17.5	23.6	14.4
Maximum dry density $\rho_{d\ max}$	g/cm ³	1.36	1.12	1.27

As shown in Table 5.2, the optimal water contents of three bentonites are 17.5%, 23.6% and 14.4%, while the natural water contents are 10.4%, 10.4% and 9.5% respectively. The maximum dry density of Turkey and Geoclay are much higher than Hokuhyo. The low maximum dry density of Hokuhyo bentonite (1.12 g/cm³) is not ideal for use in compacted bentonite layers.

The hydraulic conductivity test was according to ASTM D5084-03 (2011). The size of specimens was 100 mm in diameter, with 40 mm height. The bentonite samples were compacted to the Dc of 80% at natural water content. The water head was 1 m, and the gradient was 40-60. During the saturation procedure, the cell pressure was set to 105 and 110 kPa, and the back pressure was 25 kPa, because the swelling pressure of Turkey bentonite was around 80 kPa. Also, the saturation method was tried according to ASTM D5887-09 (2009). The hydraulic conductivity k values of bentonites were calculated as according to equation (4-1). The results are summarized in Table 5.3.

Table 5.3 Hydraulic conductivity of bentonites.

	Hydraulic conductivity k (m/s)			
	Case #1	Case #2	Case #3	Avg.
Turkey	4.2×10^{-13}	3.9×10^{-13}	5.0×10^{-13}	4.4×10^{-13}
Hokuhyo	1.5×10^{-10}	1.5×10^{-10}	1.6×10^{-10}	1.5×10^{-10}
Geoclay	1.0×10^{-10}	9.6×10^{-11}	1.2×10^{-10}	1.1×10^{-10}

It is obvious that the best hydraulic performance of pure compacted bentonite is Turkey, as the k value is lower than 1.0×10^{-12} m/s. The k values of Hokuhyo and Geoclay are similar, on the order of 10^{-10} m/s. The k value of Hokuhyo is the lowest among the bentonites.

5.2.2 Free swelling test

To estimate the swelling performance of bentonites in ferrous/ferric solutions. A series of free

swelling tests were conducted. The Fe^{2+} may be oxidized to Fe^{3+} in $\text{Fe-O}_2\text{-H}_2\text{O}$ solution system. Therefore, the bentonites were hydrating and swelling in $\text{Fe}^{3+}/\text{Fe}^{2+}$ solutions. In this chapter, the swelling tests were conducted using 10, 20, 50, 100, 200, 500, 1000 mg/L Fe^{2+} solution, prepared by mixing $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ powder with distilled water. The test procedure has been illustrated in section 4.2.2.

After the final increment has settled, adhering particles from the sides of the cylinder (mostly at the scale of 90-mL mark) were rinsed into the reagent. Then, the reagent volume was finally raised to 100-mL mark. Note that cylinders were sealed with plastic membrane to diminish the oxidation effect on ferrous solution. Finally, the graduated cylinders were allowed to stand undisturbed for 24 hours, and the observed volumes of hydrated bentonite were recorded (to the nearest 0.5 mL), as well as EC and pH values of the reagents before and after the test.

5.2.3 Gaseous mercury adsorption test

Gaseous mercury adsorption tests were conducted using constant volume glass vials. The glass vials used in this research were specially manufactured with a measured volume of 67 mL. In order to decrease the leakage of gaseous mercury from the plug and avoid adsorption on it, fluoride rubber plug was applied and capped on the glass vial, as shown in Fig. 5.5. Outside the fluoride rubber plug, plastic lid was used to rotating on the glass vial for covering of fluoride rubber plug. During the tests, gas-tight syringes with scale of 10 mL and 5 mL were used for injection of gaseous mercury and extraction of gas during the tests. The extracted gas from vials were injected through heating unit for mercury collector tube RH-MA3 and analyzed by thermal decomposition mercury analyzer MA-3000 (Nippon Instruments).



* (a) Fluoride rubber plug, (b) Glass vial, (c) Plastic lid.

Fig. 5.5 The structure of vials used in this chapter.

Standard gaseous Hg generator (i.e. Standard mercury vapor generator) from Nippon Instruments, was applied to obtain accurate elemental Hg vapor in the test. The standard

gaseous Hg generator is a device used to produce and maintain a consistent concentration of mercury vapor for calibration and tests.

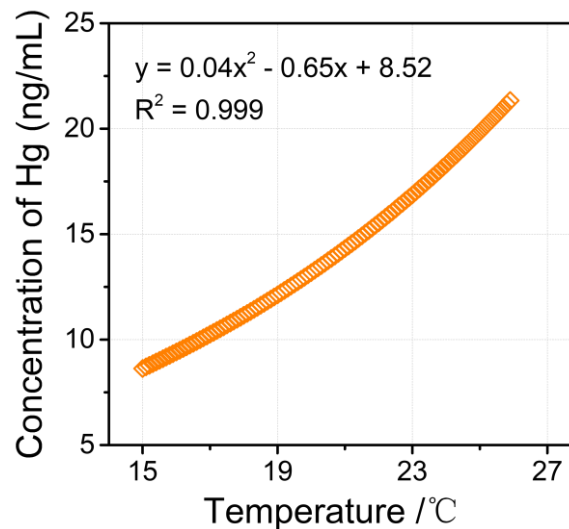


Fig. 5.6 The concentration of gaseous Hg under different temperature.

The concentration of gaseous Hg is temperature dependent. As temperature increases, the vapor pressure of Hg also rises, leading to a higher concentration. This temperature dependence is crucial for accurately measuring and controlling mercury vapor levels to ensure consistent and reliable results. Before each test, the calibration of gaseous Hg should be performed to ensure accuracy, with an R^2 value of 0.999. In the Standard gaseous Hg generator, the temperature showing on the screen varies from 15 to 26°C. The initial concentration of gaseous Hg in each test can be calculated and obtained at designated temperature from the nonlinear regression in Fig. 5.6.

The Hokuhyo is a sodium/calcium bentonite characterized by a relatively low maximum dry density and inferior hydraulic performance. Therefore, in the gaseous mercury adsorption test, Turkey and Geoclay, being sodium and calcium bentonites respectively, were utilized. Before each test, bentonite with different water content were prepared. In order to study the influence of water content, based on the actual situation that the underground water layer may be affected by rainfall, bentonite specimens with natural and optimal water content were tested. To acquire dry specimens, the bentonite specimens with natural water content were put into a drying oven at constant 105 °C for 24 hours. Then the specimens were taken into a drying box to cool down to room temperature. Given that achieving optimal water content was difficult, in this paper, the authors prepare the bentonite with optimal water content through split times of water spraying and mixing from dry bentonite specimens.

Table 5.4 Test conditions in the gaseous Hg adsorption test.

	Turkey	Geoclay
Blank	-	-
Dry ($w = 0\%$)	0.1, 0.5, 1 g	0.1, 0.5, 1 g
Optimum water content w_{opt}	0.1, 0.5, 1 g	0.1, 0.5, 1 g
Maximum dry density $\rho_{\text{d max}}$	0.1, 0.5, 1 g	0.1, 0.5, 1 g

* Three parallel specimens in each group of tests.

The arrangement of tests is illustrated in Table 5.4. In each water content, there are 0.1, 0.5, and 1 g bentonite specimens. To minimize the error, three parallel specimens were tested for each case. After adding designated bentonite specimen into the vial, the vial was sealed with fluoride rubber plug and covered with plastic lid.

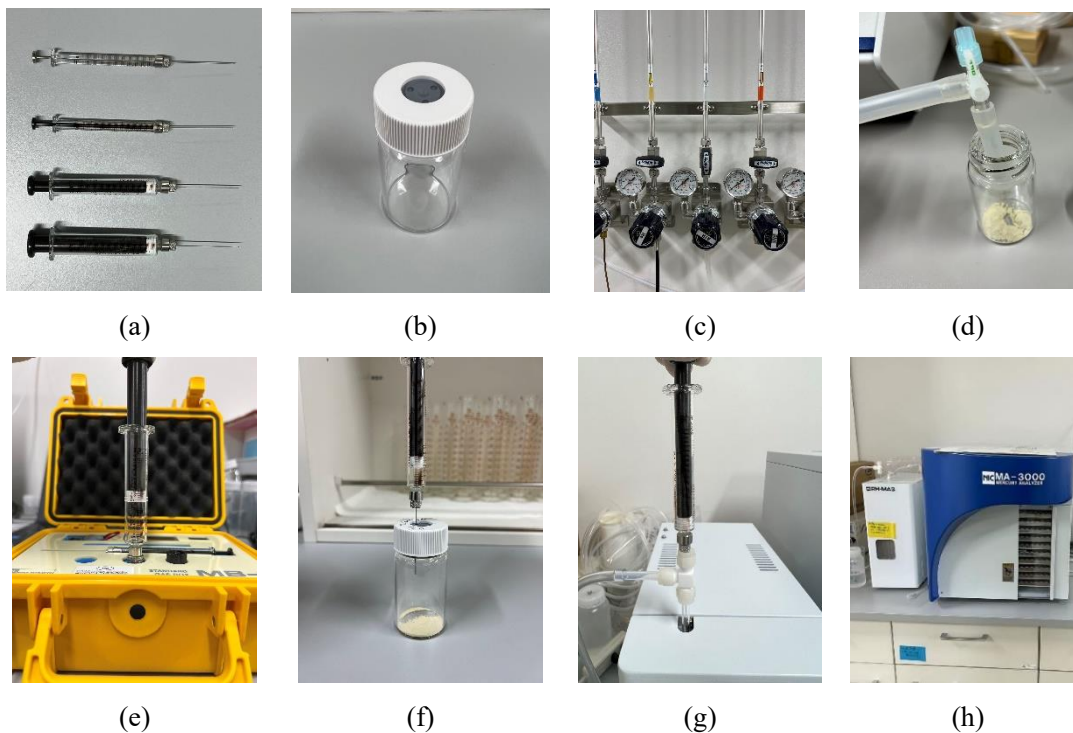


Fig. 5.7 Pictures of gaseous Hg adsorption test.

- * (a) Injector with different volume (50 μL , 100 μL , 5 mL, 10 mL).
- (b) Overview of the assembled glass vial.
- (c) Air and gas supply system (e.g. pure air, N_2 , O_2 , etc.).
- (d) Air flow into the glass vial before test.
- (e) Injection of gaseous Hg from the Standard gaseous Hg generator.
- (f) Injections in the glass vial during the test.
- (g) Injection of air sample into heating unit for mercury collector tube RH-MA3.
- (h) Thermal decomposition mercury analyzer MA-3000.

To avoid higher pressure in the glass vial, 5 mL air was extracted using 10 mL gas-tight syringe. The test started after the injection of 5 mL saturated gaseous mercury at the temperature between 20.0 to 23.0°C. After the test started, 1 mL of gas extracted and measured by mercury analyzer at time intervals of 5 mins, 15 mins, 30 mins, 60 mins and 90 mins (i.e. 5 mins, 20 mins, 50 mins, 110 mins and 200 mins after the test started), respectively. Some experimental pictures of this test have been shown in Fig. 5.7.

Given that the curve of mercury concentration with elapsed time fitting to exponential function with first order constant, the mercury concentration in the vial only decrease within 200 mins (Ishimori et al., 2020). Thus, the parameters based on blank (adsorption and potential leakage) can be calculated by the injection concentration and the final concentration at 200 mins. The adsorbed and potential leakage amount of gaseous Hg in blank test can be calculated as follows:

$$M_B = (C_0 - C_F)V \quad (5-1)$$

In this equation, M_B is the adsorbed and leaked amount of mercury. C_0 and C_F represents the initial concentration and final concentration at 200 mins. V is the volume of the glass vial (67 mL). Then, the distribution coefficient of blank test can be calculated by the relationship between Unit adsorption and leakage amount and final concentration at 200 mins. The formula can be written as:

$$K_B = \frac{M_B}{C_F} \quad (5-2)$$

In this formula, K_B is the distribution coefficient. The distribution coefficient K_B of three blank test within the initial mercury temperature from 19.0 to 20.0-degree C can be fitted to linear function. Then, the adsorption and leakage mercury amount from the vial and plug can be calculated using different temperature within 19.0 to 20.0-degree C. Eq. (1) can also be used in the calculation of bentonite specimens, and the distribution coefficient of bentonite specimens can be formulated as:

$$K_d = \frac{(C_0 - C_F)V - M_B}{C_F} \frac{1}{m} \quad (5-3)$$

In Eq. (3), K_d is the distribution coefficient of bentonite specimens, which represents the adsorption capacity of adsorbents. And m is the weight of bentonite specimens. Henry's Law constant was also analyzed in this based on the gaseous mercury concentration in the vial and

adsorbed on the bentonite. The equation of Henry's Law constant can be summarized as:

$$k_H = \frac{Hg_{(g)}}{Hg_{(s)}} \quad (5-4)$$

In Eq. (4), the k_H is Henry's Law constant. $Hg_{(s)}$ is the concentration of mercury on the solid in ng/cm^3 , and $Hg_{(g)}$ is the mercury concentration in the gaseous phase in ng/cm^3 .

5.2.4 Optimization of anaerobic condition

In order to monitor the anaerobic condition to reveal the actual situations in the mercury waste, bubbling method and strong reduction solution were utilized. Bubbling method was to decrease the dissolved O_2 in the solution. The bubbling method involves passing one kind of gas, typically nitrogen (N_2), through water. This process displaces the dissolved oxygen in the water, reducing its concentration.

The strong reduction solution used in this chapter was N_2H_4 (Hydrazine Hydrate). According to previous research, N_2H_4 solution can be used for reduction environment in solutions. N_2H_4 , containing hydrazine hydrate, is a potent reducing agent used to create a strong reducing environment (Egawa et al., 2021). Its strong reducing properties enable it to effectively reduce oxidizing agents like O_2 , fostering a reducing environment. It normally serves as a reagent for reducing metal ions, oxides, and other oxidizing agents. Additionally, it finds utility in cleaning and removing oxide layers from metal surfaces and in reducing functional groups in organic synthesis.

In this research, 0.04 mol/L N_2H_4 solution was used, combining with bubbling method to both decrease the DO value and ORP value of the ferrous solution. In anaerobic condition, 2 mL 0.04 mol/L N_2H_4 solution was mixing with distilled water to prepare 100 mL solution as blank test. After that, free swelling tests using 10, 20, 50, 100, 200, 500, 1000 mg/L Fe^{2+} solution under anaerobic condition were conducted.

5.3 Results and discussion

5.3.1 Free swelling performance of bentonite in normal ferrous solution

The results of free swelling test in ferrous solutions were measured after 24 hours hydration. The pictures of cylinders after test are shown in Fig. 5.8, and the swelling volume of each case has been plotted in Fig. 5.9.

It is obvious that, as the initial concentration of Fe^{2+} increase from 0 to 1000 mg/L, the

highest swelling volume was achieved at an Fe^{2+} concentration of 100 mg/L, irrespective of the bentonite type. In Fig. 5.8(a), the solution in 10 and 20 mg/L were yellow, and turned to be clear from 50 mg/L to 1000 mg/L. However, the swelled bentonite was mixed with sediments, and it turned to brown within the concentration range of 10 and 20 mg/L and turned to reddish brown from 50 to 200 mg/L.

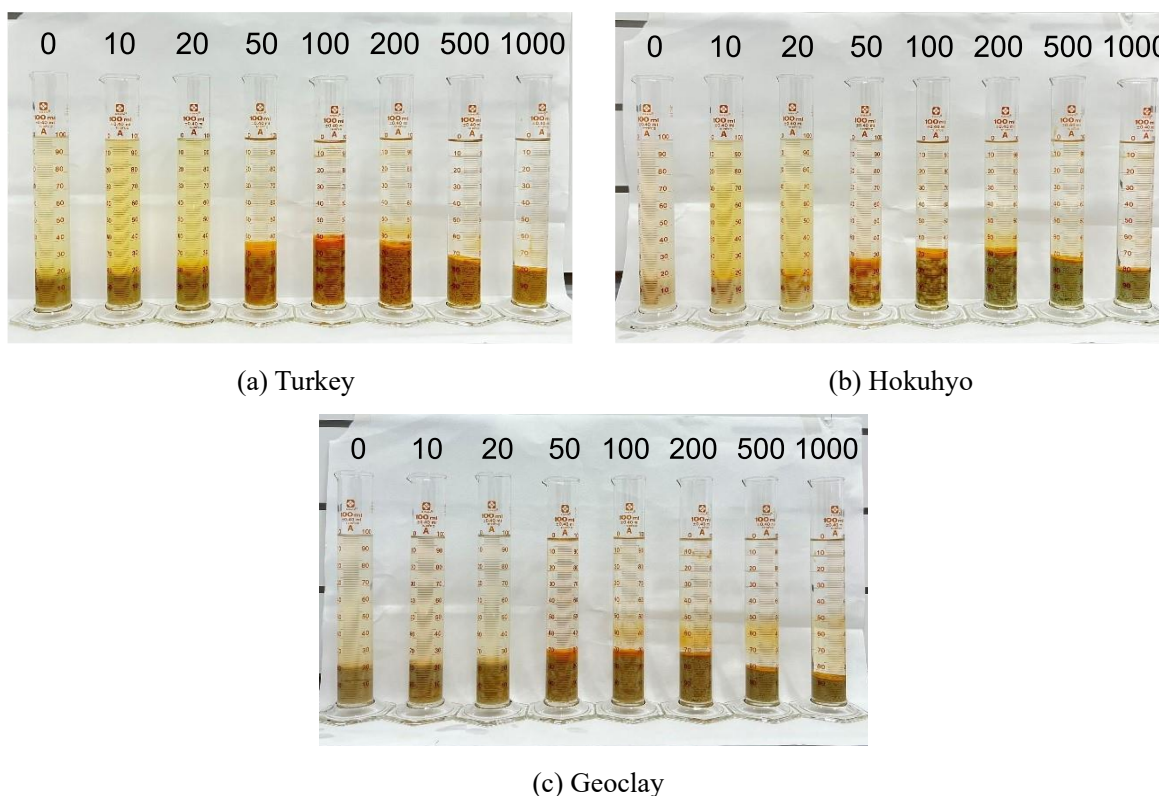


Fig. 5.8 Pictures of free swelling test in different concentrations of ferrous solutions.

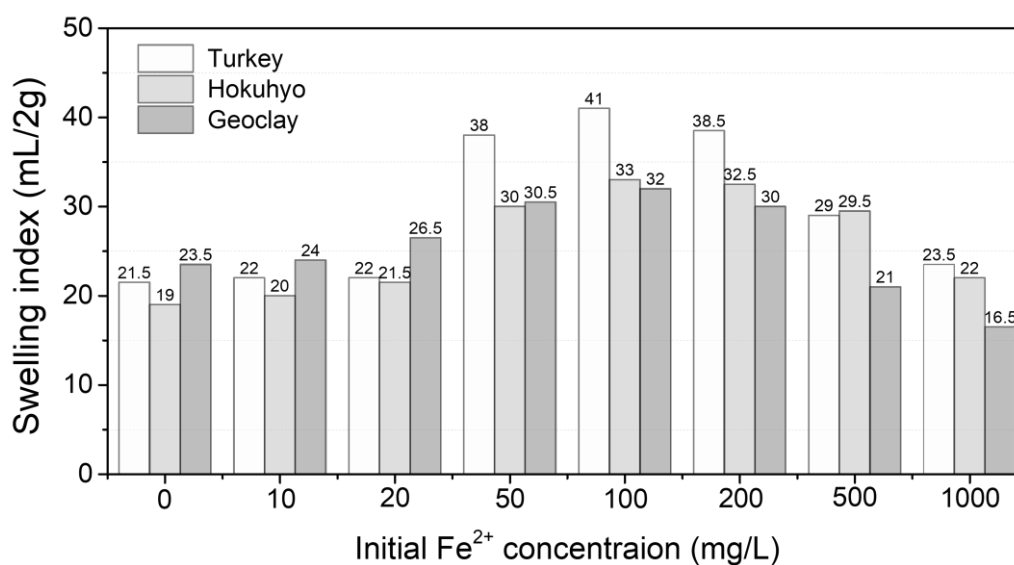
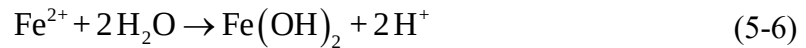
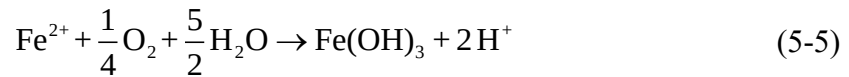


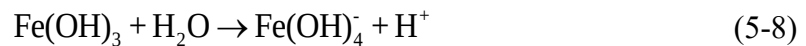
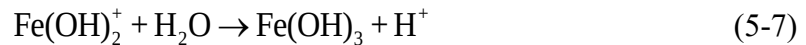
Fig. 5.9 Swelling volume of bentonites in ferrous solution.

In Fig. 5.9, it is obvious that Turkey has higher swelling volume when Fe^{2+} concentration higher than 50 mg/L. Geoclay presents better swelling performance when Fe^{2+} concentration lower than 20 mg/L. However, because Fe^{2+} oxidizes with the dissolved O_2 in the solution, different forms of sediments or colloids are generated at various concentrations. Consequently, this has various oxidation-reduction reactions and affects the swelling performance of different types of bentonites.

The pH values change in different concentration of Fe^{2+} were illustrated in Fig. 5.10, and corresponding Eh-pH diagram has been plotted in Fig. 5.11. From Fe^{2+} concentration of 20 mg/L, the swelling index of Turkey increased to the highest. According to Fig. 5.11, $\text{Fe}(\text{OH})_2^+$ is the main components in solutions. From the Fig. 5.8, more reddish-brown sediments can be seen in Turkey bentonite than in Hokuhyo and Geoclay, which means that $\text{Fe}(\text{OH})_3$ generated during the hydration process. Given that few Fe^{2+} and Fe^{3+} adsorbed between the double layer, oxidation and hydrolysis reactions occur simultaneously as listed in Formula (5-5) and (5-6).



As shown in Fig. 5.10, the pH value decreased below 6 when the concentration was 100 mg/L. In Fig. 5.11, the border between $\text{Fe}(\text{OH})_2^+$, $\text{Fe}(\text{OH})_3$ and $\text{Fe}(\text{OH})_4^-$ is the process of hydrolysis reactions, and the reaction formulas are (5-7) and (5-8).



In the hydrated bentonite, reddish sediments, white sediments and colloid mixed together. The swelling index of sodium bentonite was relatively high in around 100 mg/L, but the swelling pressure is not certain based on this combined mixture.

In high Fe^{2+} concentration, the pH value was lower than 5, which limited the reaction in formula (5-7) and also the oxidation reaction. Therefore, Fe^{2+} , Fe^{3+} , $\text{Fe}(\text{OH})_2^+$ may exist at the same time. The lower pH value also affected the hydration process of bentonite, as a result, the swelling index of calcium bentonite Geoclay decrease to 16.5 as the lowest. During each increment of 0.1 g bentonite, the hydration process was quick that the bentonite settled down sharply, and almost tiny swelling performance can be seen after 24 h hydration.

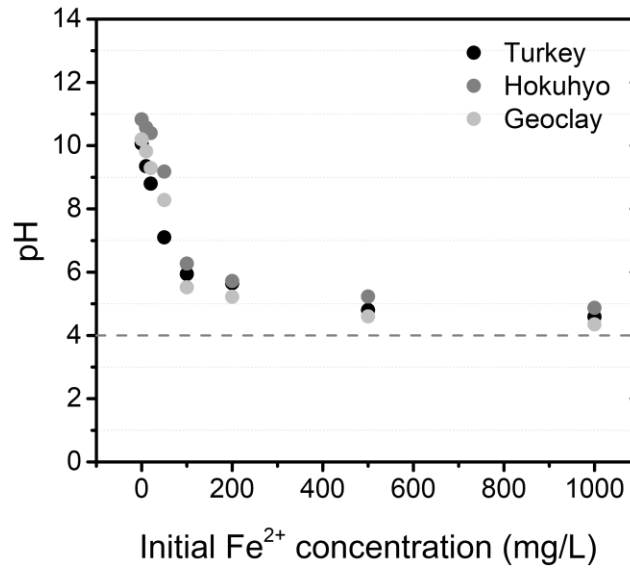


Fig. 5.10 pH changes under different Fe^{2+} concentration.

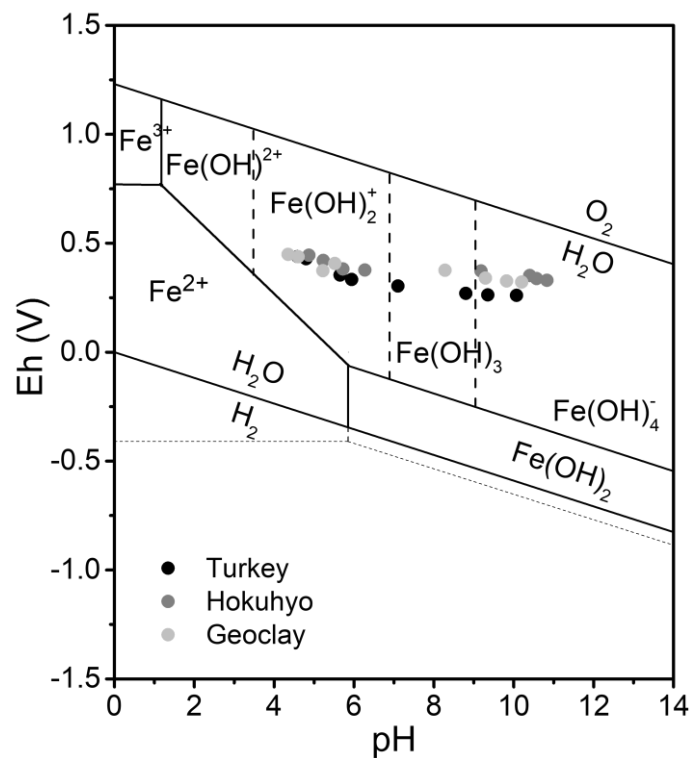


Fig. 5.11 Eh-pH diagram in Fe- O_2 - H_2O system.

5.3.2 Free swelling performance of bentonite in anaerobic ferrous solution

The Fe^{2+} will be oxidized into Fe^{3+} in the solution with dissolved O_2 , as shown in Fig. 5.12, which depicts the appearance variation of 500 mg/L Fe^{2+} solution as time elapsed for 20 mins. In Fig. 5.12, it is obvious that the color of 500 mg/L Fe^{2+} solution has changed within 20 mins,

transferred from clear green to turbid yellow (Fe^{3+} and $\text{Fe}(\text{OH})_3$). It is crucial to simulate the actual solution environment as anaerobic condition and estimate the swelling performance of bentonite in Fe^{2+} solutions. The first method was using Bubbling with N_2 in the distilled water. The initial DO value was 13 mg/L. After ten mins' bubbling, the DO value decreased to below 4 mg/L and keep decreasing to approach 0 mg/L at around 15 mins. However, the final ORP value was 5, indicating remaining dissolved oxygen. When the bubbling is stopped, the DO and ORP increased sharply. Therefore, the bubbling method is not feasible to obtain and maintain an anaerobic environment using distilled water.

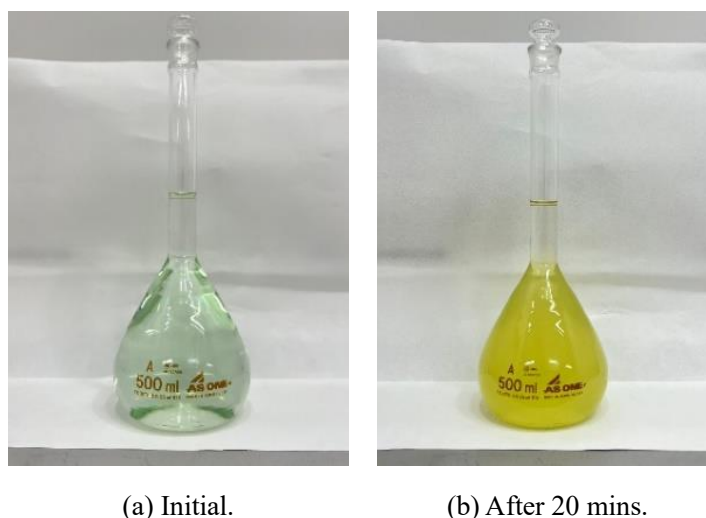


Fig. 5.12 The appearance variation of 500 mg/L Fe^{2+} solution sealed in volumetric flask.

In this chapter, N_2H_4 was used to simulate the anaerobic condition. Fig. 5.13 depicted the ORP value and temperature changes with the increase of N_2H_4 dosage. The temperature decreased to 20 °C, which is in the range for free swelling test. The ORP value decreased to -240 when 2 mL 0.04 mol/L N_2H_4 solution was added. As shown in Fig. 5.11, to achieve Fe^{2+} , the pH value must be below 6.0 in the presence of $\text{Fe}-\text{O}_2-\text{H}_2\text{O}$ system. But in anaerobic condition, the DO value before tests was decreased below 0.01 mg/L. During each increment of bentonite powder, pure N_2 was used for the confirmation of no O_2 dissolution.

The pictures of free swelling test in anaerobic condition (i.e. 2 mL 0.04 mol/L N_2H_4 solution mixing with 98 mL distilled water) and the corresponding results are shown in Fig. 5.14 and Fig. 5.15. The blank test in N_2H_4 confirms almost no effect on the swelling capacity of three types of bentonites that the variation in the swelling volume is minor. In Fig. 5.21, light green solution can be seen in Hokuhyo and Geoclay, indicating the existence of Fe^{2+} . The swelling volume of Turkey, Hokuhyo and Geoclay in normal condition was 41, 33 and 32

mL/2g, respectively. The swelling volume of bentonites in anaerobic condition decreased to 35, 28 and 26 mL/2g, indicating that the N_2H_4 affect the oxidation reaction of Fe^{2+} .

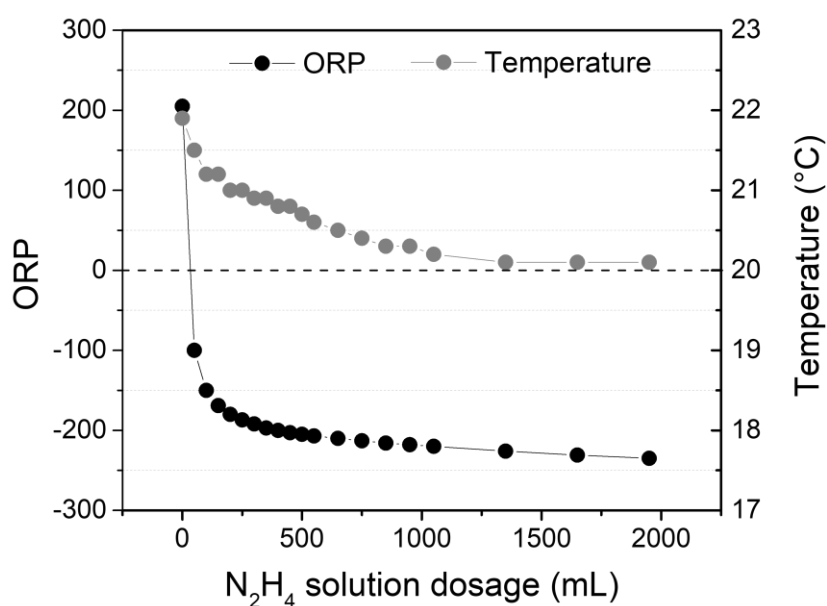
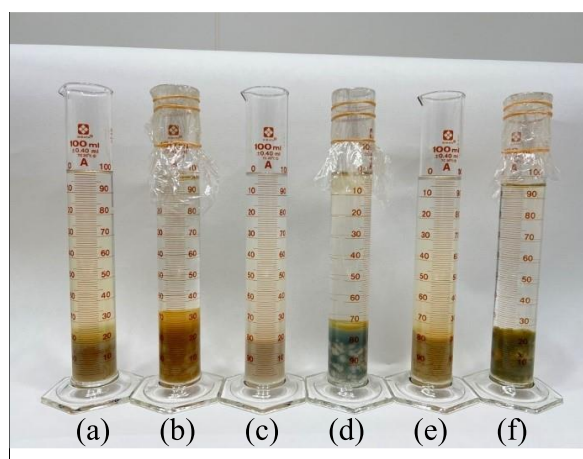


Fig. 5.13 The variations of ORP and temperature in different concentration of N_2H_4 .

The differences with normal condition are Fe^{2+} and $\text{Fe}(\text{OH})_2$, as one kind of light green sediment, which can be seen in Hokuhyo and Geoclay in Fig. 5.14. As sodium bentonite, the swelling volume was the highest and no visible light green sediment can be seen, indicating that the Fe^{2+} has been ion exchanged. Therefore, in the anaerobic condition, the hydrolysis reaction seemed still in effect, but the oxidation reaction was restricted.



* (a) Turkey-blank, (b) Turkey- Fe^{2+} 100 mg/L, (c) Hokuhyo-blank, (d) Hokuhyo- Fe^{2+} 100 mg/L, (e) Geoclay-blank, (f) Geoclay- Fe^{2+} 100 mg/L.

Fig. 5.14 Pictures of free swelling test in anaerobic condition.

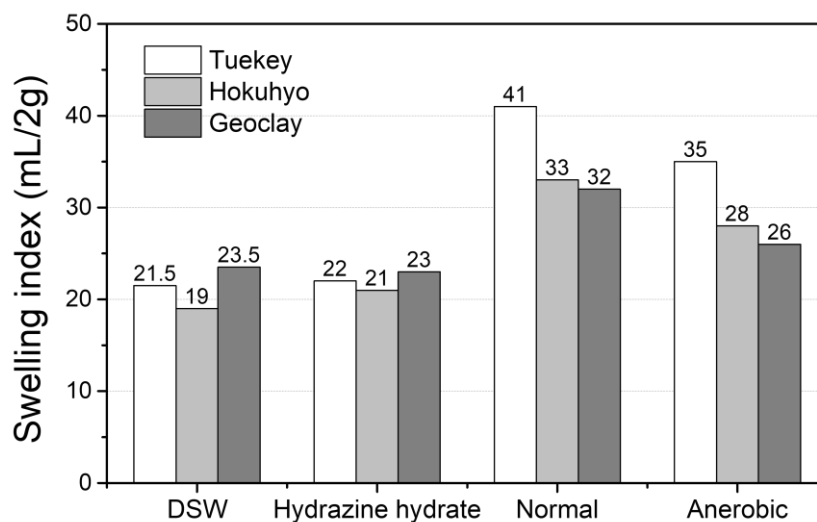


Fig. 5.15 Swelling volume of bentonites in anaerobic ferrous solution.

5.3.3 Adsorption performance of bentonite on the gaseous mercury

The results of the blank test with error bar are shown in Fig. 5.16. In the blank test, the mercury concentration detected at different time intervals gradually decreased within 200 mins. This means that the amount of gaseous mercury leaked from the glass vial or adsorbed on the inner surface gradually increases.

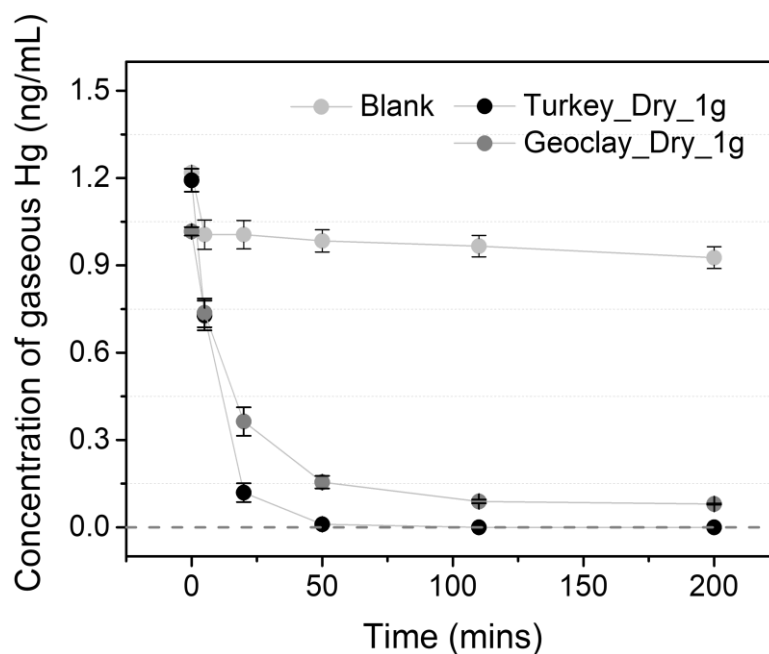


Fig. 5.16 Blank test results.

However, the speed of leakage and adsorb on the vial was the fastest within the first 5 mins

and comes to be stable from 20 mins to 200 mins. As calculated, the speed of leakage after 20 mins was almost 0.03 ng/h. Therefore, the first five mins presented significant effect on the value of leakage and adsorbed amount of mercury gas on the vial, over 90% totally. Given that the measuring time is short, the calculation of adsorption and leakage amount was calculated according to the initial concentration and final concentration. As the measured concentration of gaseous mercury decreases, but no equilibrium witnessed during 200 mins. The blank test using fluoride rubber plug is a non-equilibrium adsorption process (Ishimori et al., 2020).

In this paper, the study of adsorption capacity was all based within 200 mins, to investigate whether the leakage of gaseous mercury from the stabilized mercury solid waste can be adsorbed on the bentonite under different water content. According to Fig. 5.16, the blank adsorption and potential leakage amount of gaseous Hg at different temperature can be simulated. Then, for the adsorption capacity analysis of dry and wet specimens, the actual final concentration can be calculated by direct subtraction. The adsorption value of specimens was calculated according to initial and final concentration with the subtraction of blank test results.

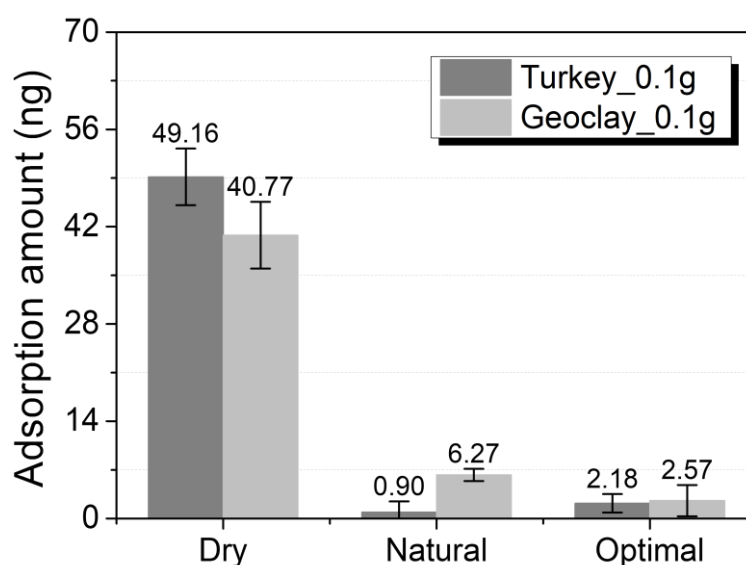


Fig. 5.17 The adsorption amount of 0.1 g specimens with different water content.

The adsorption amount of 0.1 g specimens with different water content is shown in Fig. 5.17. In Fig. 5.17, the dry specimens present better adsorption capacity than natural and optimal water content. The total adsorbed amount of gaseous mercury of dry specimens were 49.16 ng and 40.77 ng for Turkey and Geoclay, respectively. In the natural water content case, the adsorption amount of 0.1 g Turkey bentonite was around 1 ng, represented almost no adsorption capacity. However, the Geoclay bentonite adsorbed 6.27 ng with the unit adsorption amount of

62.7 ng/g. For the optimal water content cases, the adsorption performance of Turkey and Geoclay bentonite was almost the same, with the around 2.50 ng.

The adsorption amount of 0.5 g specimens with different water content is shown in Fig. 5.18. In the dry specimens, the adsorbed mercury gas of Turkey and Geoclay bentonite was 54.74 ng and 39.17 ng, which is similar to the values of 0.1 g dry specimens. The adsorption capacity of Turkey bentonite with natural water content keeps below 1 ng, while the Geoclay bentonite increased half from 6.27 ng to 9.89 ng. For the optimal water content, two bentonite presents analogous adsorption performance, with higher adsorption amount around 6 to 7 ng.

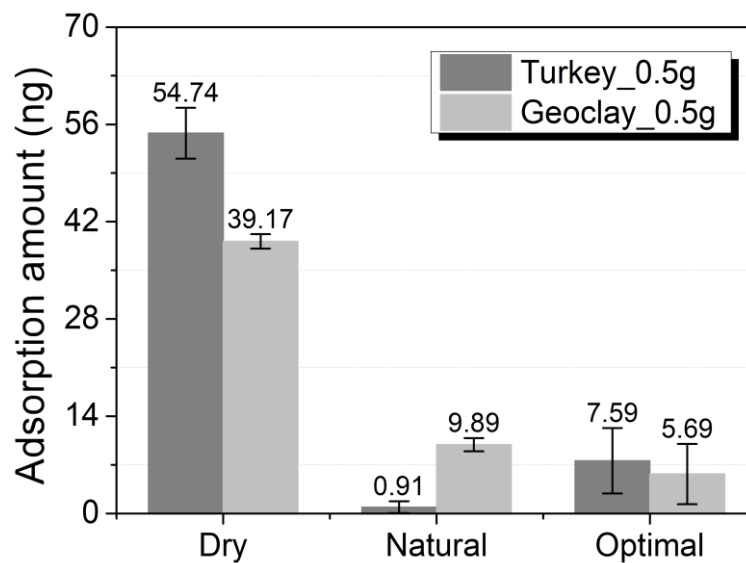


Fig. 5.18 The adsorption amount of 0.5 g specimens with different water content.

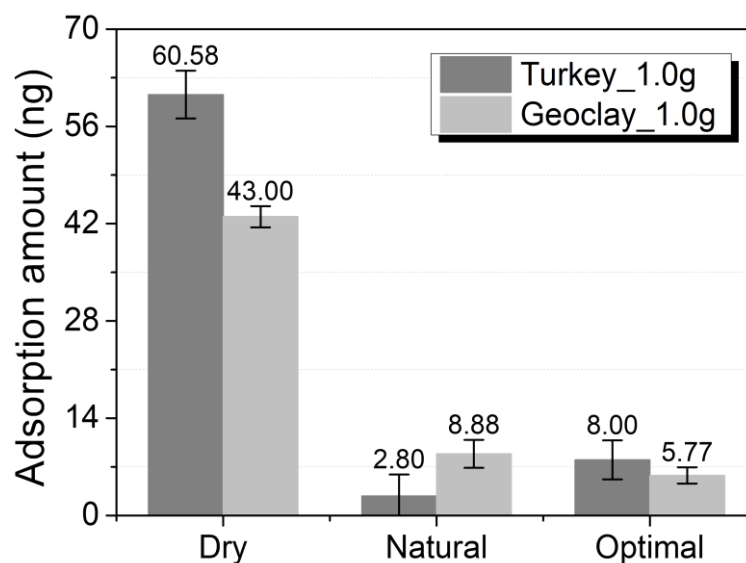


Fig. 5.19 The adsorption amount of 1.0 g specimens with different water content.

Fig. 5.19 depicted the adsorption amount of 1.0 g specimens with different water content. Comparing the results of 0.1, 0.5 and 1.0 g specimens in Fig. 5.17 to Fig. 5.18, dry specimens present the best adsorption capacity. It has been shown that the dry bentonite of Turkey possesses one third of adsorption amount higher than that of Geoclay. The total adsorption amounts of specimens with natural and optimal water content were below 9.00 ng. It means that the water content has significant influence on the adsorption process.

However, it not regular that higher water content may lead to lower adsorption capacity. In contrast, the bentonite specimens with optimal water content have better adsorption properties than natural water content. And the Turkey bentonite is better in natural water content, while Geoclay bentonite presented better performance in optimal water content. This may be caused by the different types of bentonites, and also the different ways of combination with water.

Table 5.5 Summary of gaseous Hg adsorption capacity of bentonites (Unit: ng).

Weight of bentonite (g)	Turkey			Geoclay		
	0.1	0.5	1.0	0.1	0.5	1.0
$w = 0$	49.16	54.74	60.58	40.77	39.17	43.00
w_n	0.90	0.91	2.80	6.27	9.89	8.88
w_{opt}	2.18	7.59	8.00	2.57	5.69	5.77

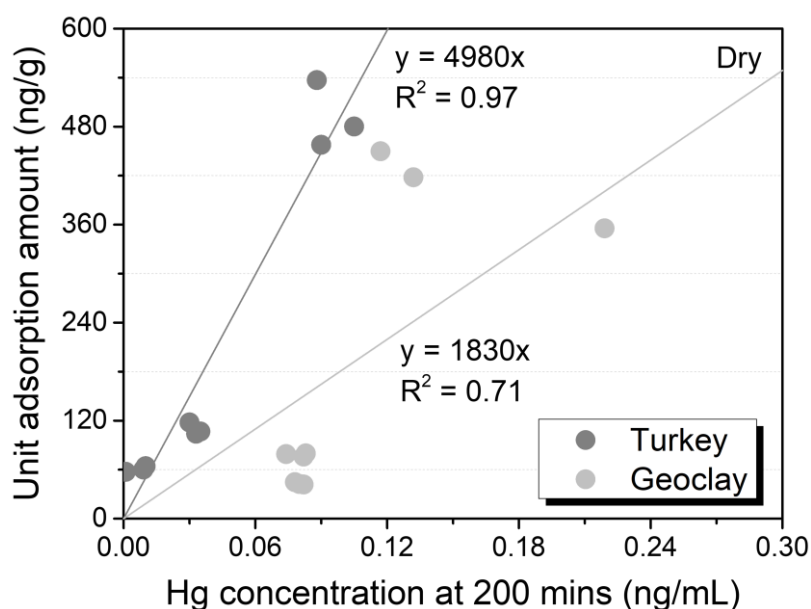


Fig. 5.20 The distribution coefficient curve of dry specimens.

The dry specimens present the best adsorption performance, and the final concentration at 200 mins was almost all below 0.15 ng/mL. According to equation (5-2) and the blank test

results, the distribution coefficients of dry specimens were calculated according to the results in Table 5.5, as depicted in Fig. 5.20. The distribution coefficient of Turkey was 4980 mL/g, and the curve was well fitted with the R^2 of 0.9651. The distribution coefficient of Geoclay was 1830 mL/g, approximately one third of Turkey. However, this curve was not in good fitting, with the R^2 around 0.70.

Other research also studied on the adsorption capacity of bentonite, and the distribution coefficient was almost 7140 mL/g (Ishimori et al., 2020). Some other adsorbent materials like silica sand, decomposed granite soil, mordenite possess the distribution coefficient of 56.3 mL/g, 2070 mL/g, 3490 mL/g respectively (Ishimori et al., 2020).

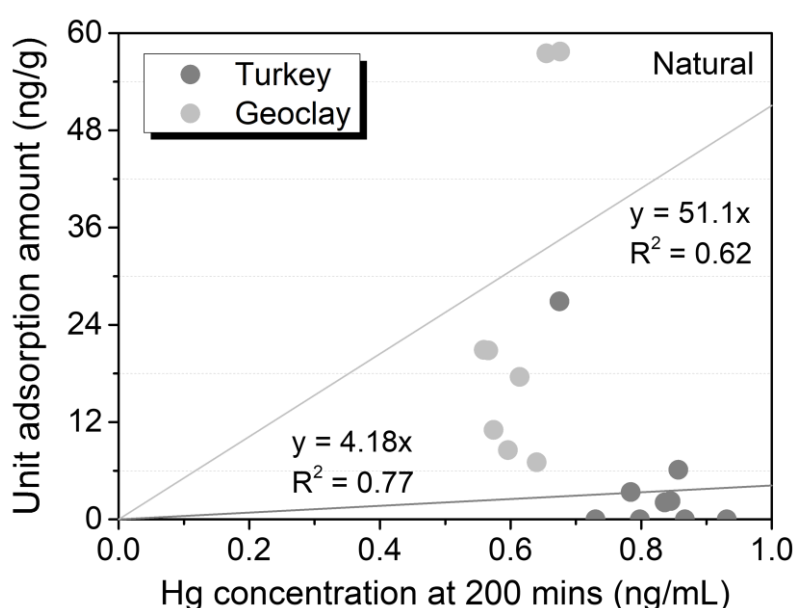


Fig. 5.21 The distribution coefficient curve of specimens with natural water content.

Based on the results of adsorbed amount and final concentration, the distribution coefficient was calculated through linear fitting. The distribution coefficient curve of specimens with natural water content is shown in Fig. 5.21. The distribution coefficient of Turkey was 4.18 mL/g. When the bentonite mixed with water, the electric double layer was connected by H_2O molecular. The bentonite swells gradually but fewer and smaller particle surface provided for the adsorption of elemental mercury gas. The distribution coefficient of Geoclay was higher, as the value was 51.1 mL/g.

The distribution coefficient curve of specimens with optimal water content is shown in Fig. 5.22. The R^2 were all lower than 0.70, which mean that the linear fitting was not good. It can also be witnessed from the vertical alignment of the adsorption test results. As the consideration of poor R^2 values in Fig. 5.21 and 5.22, the relationship between mercury concentration at 200

mins and the weight of specimen was not comprehensive.

As mentioned above, for optimal water content, the Turkey is better than Geoclay. On the contrary, the distribution coefficient of Geoclay was 32.0 mL/g, higher than that of Turkey as 22.3 mL/g. It can be concluded from the results that, in the dry cases, the Turkey bentonite have much better adsorption performance than Geoclay. In the cases of natural and optimal water content, the Geoclay bentonite present higher adsorption capacity than Turkey bentonite, especially in natural water content. Through comparing the results of dry specimens and wet specimens, from Fig. 5.20 to Fig. 5.22, the R^2 of wet specimens were relatively low, three of four were lower than 0.70, which possibly caused by the difficulty of keeping homogeneous of wet specimens, and the potential leakage from the vials during test.

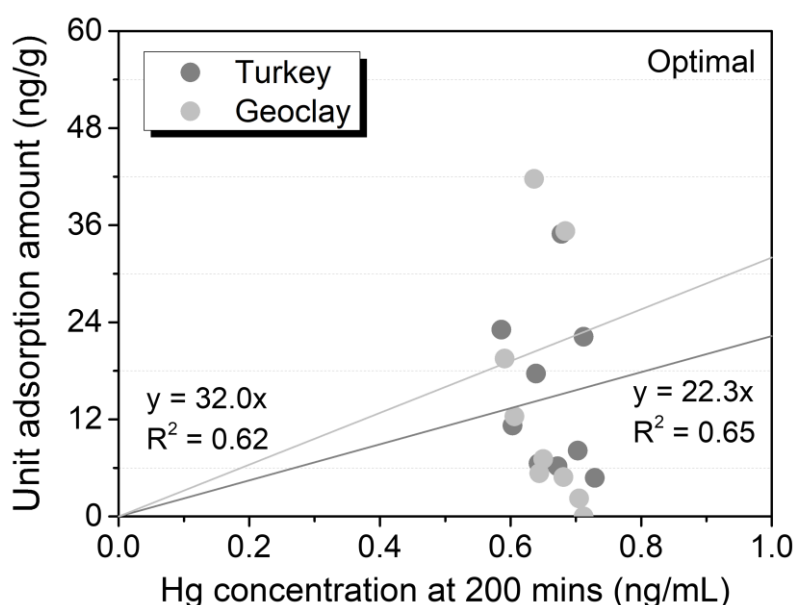


Fig. 5.22 The distribution coefficient curve of specimens with optimal water content.

Table 5.6 Summary of distribution coefficient of bentonites with different water content (Unit: mL/g).

	$w = 0$	w_n	w_{opt}
Turkey	4980	4.2	22.3
Geoclay	1830	51.1	32.0

In general, wet specimens have relatively low adsorption capacity. The Turkey bentonite is better for utilization if this kind of Na-Bentonite presents lower hydraulic conductivity than Ca-Bentonite, although the Geoclay bentonite have better adsorption effect in dry specimens and specimens with optimal water content. As the leaching of treated Hg waste meets the criterion that lower than 0.005 mg/L using dissolution test and lower than 0.001 mg/m³ using Headspace

Method (Japanese regulatory leaching test, JLT-13), the adsorption capacity of the bentonites satisfies the real situations.

5.4 Conclusions for this chapter

In this chapter, three types of bentonites (i.e. sodium, sodium/calcium and calcium bentonite), namely Turkey, Hokuhyo and Geoclay, were introduced. The optimal water content and maximum dry density were obtained from series of compaction tests. The hydraulic performance of pure bentonites permeated with distilled water were also discussed. The swelling performance of bentonites in ferrous solution were studied in normal condition and anaerobic condition, and potential mechanisms (e.g. pH, Eh etc.) that might influence the swelling performance were analyzed. Finally, two bentonites (Turkey and Geoclay) were applied in constant volume adsorption test to investigate the influence of water content for the adsorption capacity on gaseous mercury. Dry specimens and specimens with natural and optimal water content were tested, and the adsorption properties of these two bentonites were compared. The main results obtained in this chapter can be summarized as follows:

- (1) According to the compaction test results, the optimal water content of Turkey, Hokuhyo and Geoclay were 17.5, 23.6 and 14.4%, as the corresponding maximum dry density were 1.36, 1.12 and 1.27 g/cm³, respectively. The hydraulic conductivity of bentonites was 4.4×10^{-13} , 1.5×10^{-10} and 1.0×10^{-13} m/s.
- (2) Turkey represents the best swelling performance in the free swelling test in both normal and anaerobic conditions. As the concentration of Fe²⁺ increased, the swelling volume of the bentonites followed a similar trend, initially increasing to a peak and then decreasing. The turning point was at a concentration of 500 mg/L.
- (3) The hydrolysis of Fe²⁺ with water and oxidation with dissolved O₂ are the main effect on the swelling volume. The mechanism can also be seen from the appearance of the swelled bentonite after test. At certain Fe²⁺ concentration of 500 mg/L, the Fe²⁺ maximize the ion exchange and expansion of the bentonite layers, resulting in the highest swelling volume. At this point, the solution contains a mix of Fe²⁺ and Fe³⁺ compounds, including Fe (OH)₂⁺ and Fe(OH)₃ compounds, leading to the appearance of bentonite as mix of yellow and reddish-brown.
- (4) When the Fe²⁺ concentration is lower than 500 mg/L, the ion exchange reacted in the double layer of bentonites, causing an increase in the interlayer distance and swelling volume. The solution appears yellow, due to the partial oxidation of Fe²⁺ to yellow Fe³⁺ and Fe(OH)₄⁺ in

the solution by dissolved oxygen. When the Fe^{2+} concentration is higher than 500 mg/L, the high ionic strength of the solution interferes with the bentonite ion exchange and swelling capacity, even lower than distilled water. Also, few sediments were generated because of the high ionic strength.

- (5) The anaerobic condition was applicable by using bubbling before test and N_2H_4 solution to simulate strong reduction environment. In the anaerobic condition, the swelling volume of bentonites was lower than under normal conditions, but higher than in distilled water.
- (6) In the gaseous Hg adsorption test, fluoride rubber plug, and plastic lid were used in this research to avoid the leakage of gaseous mercury from the vials. However, from the results, the gaseous mercury leaked from the plug and adsorbed on the vials approaching 0.3 ng within 200 mins, almost 90% in the first 20 mins.
- (7) The water content significantly affected the adsorption capacity of these two bentonites. The adsorption amount decreased from 40~60 ng to 0~10 ng sharply, when the water content increased to optimal and natural water content. At the same time, the distribution coefficient in 200 mins also plunged from thousands to tens mL/g.
- (8) Comparing the two bentonites, the dry specimens of Turkey have better adsorption performance than Geoclay, with the distribution coefficient of 4980 mL/g and 1830 mL/g respectively. When the water content increased, the Geoclay presented higher adsorption capacity, especially in optimal water content.

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Chapter 6

Practical implications

6.1 General remarks

The study of soil-bentonite mixtures amended with slag cement has significant practical implications for the construction and performance of vertical cutoff walls, commonly used in geoenvironmental engineering for contaminant containment and groundwater control. Vertical cutoff walls are critical barriers designed to prevent the lateral migration of contaminants in subsurface environments, particularly around waste disposal sites, landfills, and contaminated sites. This chapter will focus on the practical aspects of using soil-bentonite-slag cement mixtures, emphasizing mechanical and hydraulic performance, relating to the application in real scenarios.

The utilization of soil-bentonite mixtures in specially engineered mercury waste landfills presents an innovative and effective approach to managing mercury waste. This practical implementation leverages the unique properties of bentonite, such as its low permeability, high swelling capacity, and cation exchange capabilities, to create a robust containment barrier. This chapter integrates the results from fundamental properties, free swelling tests, and adsorption tests to illustrate the practical applications.

6.2 Soil-bentonite-slag cement mixtures as cutoff wall material

The mechanical performance of soil-bentonite mixtures amended with slag cement is pivotal for ensuring the structural integrity and durability of vertical cutoff walls. Consolidated-undrained (CU) isotropic triaxial compression tests have provided valuable insights into the stress-strain characteristics, failure modes, shear strength properties, and the effects of cement content, curing period, and confining pressure on these mixtures.

The addition of slag cement significantly influences the stress-strain behavior of soil-bentonite mixtures. As discussed in Chapter 3, SB-C mixtures with lower cement content (0 to 25 kg/m³) exhibit stress-hardening behavior, while higher cement content (75 kg/m³) results in strain-softening behavior. The optimal mixture (SB-C25) demonstrates a combination of both behaviors, indicating a balanced mechanical response. This balance is crucial for vertical cutoff walls, as it ensures adequate flexibility to accommodate ground movements while maintaining

sufficient strength to resist deformation (Zentar et al., 2016). Failure modes are also affected by cement content. The transition from ideal ductile failure to subpar planner failure occurs between 25 and 50 kg/m³, and from planner to undesirable split failure between 50 and 75 kg/m³. These findings suggest that a careful selection of cement content is necessary to avoid brittle failure, which could compromise the integrity of the cutoff wall under varying stress conditions (Yilmaz et al., 2018). Shear strength properties, including the undrained shear strength (S_u) and residual shear strength (S_r), are enhanced by cement amendment. This doctoral study found that the cement content significantly influences S_u and S_r , with higher contents leading to greater strength. For instance, the ultimate S_u of 334.7 kPa was observed in SB-C75-28 under 160 kPa confining pressure. This enhanced strength is vital for the stability of vertical cutoff walls, particularly in areas subject to high lateral pressures, regardless of side-effect as brittle at failure with no deformability.

Hydraulic conductivity (k) tests using triaxial apparatus and flexible wall permeator have provided essential data on the permeability and self-healing properties of these mixtures. The hydraulic conductivity of soil-bentonite mixtures is inherently low, a desirable characteristic for cutoff walls. The study found that the k value of unamended SB-C0 was 7.4×10^{-11} m/s, and the k values for amended mixtures decreased over time due to cement hydration. The primary k values of SB-C25, SB-C50, and SB-C75 were similar initially but decreased significantly with permeation time. For instance, the k value of SB-C25 reached a constant value of 4.5×10^{-10} m/s, while the k values of SB-C50 and SB-C75 continued to decrease, demonstrating the long-term hydration capabilities of the mixtures (Xiao et al., 2020). Hydraulic conductivity tests using flexible wall permeator showed stable pH and EC values in SB specimens, indicating stable swelling and water flow through the bentonite. In SB-C specimens, only SB-C25 achieved equilibrium pH and EC. The hydration of cement filled the pore spaces, decreasing the void ratio and generating immobile water, leading to extremely low k values and sharp decreases in pH and EC. These findings emphasize the importance of proper cement content to maintain low permeability and effective barrier performance (Benson et al., 2011). Triaxial apparatus permeation tests indicated higher k values for SB-C specimens compared to SB-C0-0, suggesting that cement content and curing time are major factors influencing k . The effect of confining pressure was significant in SB-C0 and SB-C25 but less so in SB-C50 and SB-C75. This highlights the need for appropriate confining pressure during the construction and operation of vertical cutoff walls to ensure optimal hydraulic performance (Zhang et al., 2018).

The findings from this doctoral study verified the suitability of soil-bentonite-slag cement mixtures for vertical cutoff walls, providing essential guidelines for their practical

implementation. The enhanced mechanical properties, including improved stress-strain behavior, failure modes, shear strength, and elasticity, coupled with good hydraulic performance and self-healing capabilities, making 25 kg/m³ cement content ideal for use in vertical cutoff walls. Therefore, the use of soil-bentonite-slag cement mixtures in vertical cutoff walls offers significant advantages in terms of mechanical and hydraulic performance. The addition of slag cement enhances the structural integrity and durability of vertical cutoff wall, ensuring they can withstand varying stress conditions and maintain low permeability over time (Gleason et al., 2004; Di Emidio et al., 2018). These findings provide a robust foundation for the practical implementation of soil-bentonite-slag cement mixtures for preservation of geoenvironment.

6.3 Pure bentonite utilization in mercury waste landfill

The compaction tests of different bentonites, namely Turkey, Hokuhyo, and Geoclay, revealed significant variations in their optimal water content and maximum dry density. For practical implementation in mercury waste landfills, these properties are crucial as they directly influence the hydraulic conductivity of the bentonite layer. The hydraulic conductivity values are critical for the design of compacted bentonite layers in mercury waste landfills. The extremely low permeability of these bentonites, especially Turkey and Geoclay, ensures minimal fluid migration through the landfill barrier. In practical applications, achieving and maintaining the optimal water content during compaction is paramount. This can be managed through precise control of moisture conditions during the construction of the landfill barrier, ensuring that the bentonite remains at its most effective state for minimizing permeability (Egloffstein, 2001).

The swelling performance of bentonites in the presence of ferrous solutions, under both normal and anaerobic conditions indicated that Turkey bentonite exhibited the best swelling performance, maintaining its volume even under varying concentrations of Fe²⁺. The swelling capacity of bentonite is crucial for maintaining the integrity of the containment barrier. The study showed that the swelling volume initially increased with Fe²⁺ concentration up to 500 mg/L, then decreased beyond this point due to the high ionic strength interfering with ion exchange and swelling capacity. Under anaerobic conditions, the swelling volume of bentonites was lower than under normal conditions but still higher than in distilled water. This indicates that bentonites can perform effectively in reducing environments, which might be present in certain landfill conditions. Maintaining an optimal swelling capacity under various environmental conditions ensures that the bentonite layer remains effective in isolating mercury waste, preventing the formation of preferential flow paths that could lead to contaminant

leakage (Katsumi et al., 2008).

The adsorption capacity of bentonites for gaseous mercury is a critical factor in their application for mercury waste landfills. The study revealed significant variations in the adsorption performance of Turkey and Geoclay bentonites, influenced by water content. Dry specimens of Turkey bentonite exhibited superior adsorption performance, with a distribution coefficient of 4980 mL/g compared to 1830 mL/g for Geoclay. However, as the water content increased to natural and optimal levels, the adsorption capacity decreased sharply. In mercury waste landfill, maintaining the bentonite at an optimal moisture level is essential to balance its swelling and adsorption properties. The findings suggest that while dry bentonite is more effective in adsorbing mercury, engineered solutions might include a combination of bentonite layers with varying moisture content to maximize both adsorption and swelling performance (Yuan and Weng, 2006).

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Chapter 7

Conclusions and future research

7.1 Major conclusions

This study was conducted to evaluate the feasibility of using slag cement as alternative cementitious binder for the amendment of mechanical and hydraulic performance of soil-bentonite mixtures. The other scope is to investigate the swelling performance of three bentonites, and adsorption performance of two bentonites on gaseous mercury, and the feasibility in using as compacted bentonite layer with certain adsorption capacity.

In Chapter 1, the overall study background was clarified, also the objectives and the contents of the study were presented.

In Chapter 2, the properties of bentonite were introduced, and the mechanism of swelling and self-healing was explained. Current studies on enhancement on mechanical performance of SB, hydraulic performance of amended SB, along with gaseous mercury adsorption and utilization of bentonite were summarized.

In Chapter 3, the mechanical performance of soil bentonite mixture amended with slag cement was studied based on the results of consolidated-undrained (CU) isotropic triaxial compression tests. The stress-strain characteristics, failure modes, shear strength properties, etc. were evaluated and effects of cement content (e.g. 0, 25, 50 and 75 kg/m³), curing period (e.g. 7 and 28 days) and confining pressure (e.g. 40, 80 and 160 kPa) were discussed.

The stress-strain characteristics of soil-bentonite mixtures are primarily influenced by the content of slag cement. Mixtures with lower cement content, such as SB-C0 and SB-C25, exhibit stress-hardening behavior, while those with higher cement content, such as SB-C75, demonstrate strain-softening behavior. SB-C50 displays a combination of strain-hardening and strain-softening behavior, representing a typical stress-strain relationship. The failure mode transitions from ideal ductile failure to subpar planner failure between cement contents of 25 kg/m³ and 50 kg/m³, and from subpar planner failure to undesirable split failure between 50 kg/m³ and 75 kg/m³.

Cement amendment enhances both undrained shear strength (S_u) and residual strength (S_r), particularly in specimens with higher cement content. For instance, SB-C75-28 exhibits a S_u of 334.7 kPa under 160 kPa confining pressure at 2.3% axial strain. The effects of cement content and curing time on the effective stress angle (ϕ') are negligible, while the cohesion (c') shows a linear relationship with cement content, with cementation effects becoming more significant after 28 days of curing. The specimens tend to dilate with increase slag cement content, while the soil-bentonite specimens and 25 kg/m³ slag cement amended specimens tend to collapse with good deformability. The tendency of dilation is not acceptable that it leads to the particle compression within themselves, and mostly caused totally destroy of the particles, in general planar and split failures.

Excess pore pressure variations are positive in all specimens during compression, with peak values reached at different axial strains under varying confining pressures. Several specimens, particularly those with high cement content and low confining pressure, exhibit negative pore pressure, indicating relative low void ratio. Furthermore, SB-C0 specimens demonstrate high stress dependency, with significant reductions in the modulus of elasticity (E_{50}) under high confining pressure. However, specimens with cement amendment show increased moduli with higher cement content and confining pressure, with SB-C75 specimens displaying higher stiffness retention capacity, implying greater compression resistance. By considering all the aspects of mechanical performance, 25 kg/m³ is the best slag cement amendment concentration for SB mixtures.

In Chapter 4, the swelling performance of bentonite under varying cement content was assessed, alongside the hydraulic conductivity of soil-bentonite mixtures amended with slag cement. Potential mechanisms (e.g. volumetric strain, void ratio, etc.) that might influence the hydraulic performance are analyzed.

The free swelling volume of bentonite increased in cement solutions, inversely correlating with cement content, while significant variations in electrical conductivity (EC) were observed. The consolidation process of SB significantly influenced volumetric strain, with higher confining pressures leading to increased strain, while reverse in SB-C. The effect of consolidation is significant in SB specimens as the volumetric strain was over 20% while in SB-C specimens were lower than 3.5%. In the hydraulic conductivity test using flexible wall permeator, the k value of SB-C0 was obtained with k value of 7.4×10^{-11} m/s. The decrease of k values in SB-C specimens is governed not by PVF but by elapsed time. The primary k values of SB-C were influenced by cement content. With the permeation, the k of SB-C25 reached a

constant value of 4.5×10^{-10} , while k values of SB-C50 and SB-C75 kept decreasing because of the continuous hydration, which can be seen in the variations of EC.

In the hydraulic conductivity test using triaxial apparatus, the k values of SB-C0-0 is lower than all the SB-C specimens. The cement content and curing time are the major effect on the k . The effect of confining pressure (i.e. consolidation effect, volumetric strain) on the k values is significant in SB-C0 and SB-C25, while minor effect was found when the cement content increased to 50 and 75 kg/m³. The measured k values are relatively higher using triaxial apparatus, because there was no permeation in the specimens, which is beneficial for the sufficient osmotic swelling in SB specimens and crucial for the hydration of cementitious materials in SB-C specimens. Finally, the slag cement amended soil-bentonite mixtures is considered to have an enough hydraulic barrier performance, as compared to the performance-based criterion (1.0×10^{-9} m/s) in this study.

In Chapter 5, The swelling performance of three bentonites in ferrous solution were studied in normal condition and anaerobic condition, and potential mechanisms (e.g. pH, Eh etc.) that might influence the swelling performance were analyzed. In constant volume adsorption test, two bentonites (Turkey and Geoclay) were applied to investigate the influence of water content for the adsorption capacity on gaseous mercury.

The bentonite type was determined through CEC test and XRD patterns. Turkey, as sodium bentonite, represents the best swelling performance in the free swelling test in both normal and anaerobic conditions. As the concentration of Fe²⁺ increased, the swelling volume initially increasing to a peak at a concentration of 500 mg/L and then decreasing. The hydrolysis of Fe²⁺ with water and oxidation with dissolved O₂ are the main effect on the swelling volume. At certain Fe²⁺ concentration of 500 mg/L, the Fe²⁺ maximize the ion exchange and expansion of the bentonite layers, resulting in the highest swelling volume. At this point, the solution contains a mix of Fe²⁺ and Fe³⁺ compounds, including Fe (OH)₂⁺ and Fe(OH)₃ compounds. When the Fe²⁺ concentration is lower than 500 mg/L, the ion exchange reacted in the double layer of bentonites, causing an increase in the interlayer distance and swelling volume. When the Fe²⁺ concentration is higher than 500 mg/L, the high ionic strength of the solution interferes with the bentonite ion exchange and swelling capacity, even lower than distilled water.

The anaerobic condition was successfully simulated by using bubbling before test and N₂H₄ solution. In the anaerobic condition, the swelling volume of bentonites was lower than under normal conditions, but higher than in distilled water. In the gaseous Hg adsorption test, the water content significantly affected the adsorption capacity of these two bentonites. The

adsorption amount decreased from 40~60 ng to 0~10 ng sharply, when the water content increased to optimal and natural water content. Similarly, the distribution coefficient is also plunged from thousands to tens mL/g. The dry specimens of Turkey have better adsorption performance than Geoclay, with the distribution coefficient of 4980 mL/g and 1830 mL/g respectively. When the water content increased, the Geoclay presented higher adsorption capacity, especially in optimal water content. This adsorption capacity meets the leaching concentration as it is extremely low under anaerobic conditions.

In Chapter 6, the practical aspects of using soil-bentonite-slag cement mixtures were discussed, emphasizing mechanical and hydraulic performance, and relating to the application in vertical cutoff wall. Also, the results from fundamental properties, free swelling tests, and adsorption tests were integrated to illustrate the practical applications in mercury waste landfill.

7.2 Future research

7.2.1 Soil-bentonite-slag cement mixtures as cutoff wall material

To refine the mechanical and hydraulic properties of soil-bentonite-slag cement mixtures, sophisticated models combining with test results can be studied to predict the behavior of these mixtures under various loading and environmental conditions. Advanced numerical modeling, combined with comprehensive laboratory and field testing, could provide deeper insights into the stress-strain behavior, failure mechanisms, and shear strength properties of these mixtures.

To compare the mechanical properties of OPC and slag cement amended SB. It is recommended to utilize OPC and conduct the triaxial compression test to distinguish the mechanical performance from these two cementitious materials, as the slag cement is much more eco-friendly.

For the long-term hydraulic performance, further studies need to be applied to investigate the void changes in the specimen. The hydration of cement fills the voids, but ordinary methods could not calculate the pore space accurately. Sufficient micro analysis and calculation is necessary. To understand the long-term performance and durability of soil-bentonite-slag cement mixtures, long-term field study is one method to monitor the performance of these mixtures under real conditions. Factors such as freeze-thaw cycles, chemical exposure, and biological activity can significantly affect the durability and integrity of the cutoff walls. This is essential for developing maintenance strategies and ensuring the long-term effectiveness of vertical cutoff walls in containing contaminants.

The integration of advanced modeling techniques with experimental research can significantly enhance the understanding and prediction of the behavior of soil-bentonite-slag cement mixtures. Future research can focus on developing models that can accurately simulate the mechanical and hydraulic performance of these mixtures under various conditions. Machine learning also offer tools for analyzing large datasets and identifying results that might not be evident through traditional analysis methods. In this way, the test results could be thoroughly analyzed, and the mechanism could be further discussed.

Additionally, optimizing the proportions of slag cement and bentonite to achieve the best balance between strength and permeability is crucial. While higher cement content generally improves strength, it can negatively affect the swelling and self-healing properties of bentonite, leading to increased hydraulic conductivity. Research should explore the use of supplemental additives (e.g. nanomaterials) or alternative binders that could enhance strength without compromising hydraulic performance. While slag cement has shown promise as a binder for soil-bentonite mixtures, other materials could offer similar or improved benefits. Future research can investigate the potential of alternative binders such as fly ash, metakaolin, and other pozzolanic materials. These binders might provide similar mechanical benefits while potentially offering advantages in terms of better availability. Comparative studies evaluating the performance of different binders in terms of mechanical strength, hydraulic conductivity, and environmental sustainability are needed.

7.2.2 Pure bentonite utilization in mercury waste landfill

First is using fractal theory to analyze existing results, elucidating the complex relationships between different influencing factors. The results could be better discussed in this method. Optimization of anaerobic condition is crucial to real estimate the swelling and hydraulic performance of bentonite. Hydraulic conductivity tests in anaerobic conditions to assess bentonite effectiveness under permeation of Fe^{2+} solution is necessary. Understanding the hydraulic and barrier mechanism could enhance the practical implements in mercury waste landfills. Finally, improving homogeneity of bentonite specimen in mercury adsorption tests is very important to accurately analyze bentonite adsorption mechanisms. Studying microstructural changes under wet conditions could also further explain the mechanism and put forward research.

Appendix

Failure pictures of the specimens.



SB-C25-7 40 kPa



SB-C25-7 80 kPa



SB-C25-7 160 kPa



SB-C25-28 40 kPa



SB-C25-28 80 kPa



SB-C25-28 160 kPa



SB-C50-7 40 kPa



SB-C50-7 80 kPa



SB-C50-7 160 kPa



SB-C50-28 40 kPa



SB-C50-28 80 kPa



SB-C50-28 160 kPa



SB-C75-7 40 kPa



SB-C75-7 80 kPa



SB-C75-7 160 kPa



SB-C75-28 40 kPa



SB-C75-28 80 kPa



SB-C75-28 160 kPa