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Soil strengthening using thermo-gelation biopolymers

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HIGHLIGHTS

- Thermo-gelation biopolymers are introduced as new construction materials.
- Micro interaction between thermo-gelation biopolymers and soils is investigated.
- Strength change with time, biopolymer quantity, and water content is evaluated.
- Thermo-gelation biopolymers form hydrogen bonding with clayey particles.
- Sandy soil shows hysteretic strength path along drying and wetting.

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ABSTRACT

A new biopolymeric construction material for soil treatment/improvement is introduced in this study in an effort to develop an environmentally-friendly construction engineering approach to replace the use of conventional materials that have high environmental impact. Thermo-gelation biopolymers dissolve and form a suspension in heated (*i.e.*, 85–90 °C) water, and then coagulate (*i.e.*, gelate) upon a decrease of temperature (*i.e.*, below 50 °C). Gellan gum and agar gum are typical thermo-gelation biopolymers with potential as soil strengthening construction materials due to their hydrogen bonding characteristics, and were used to treat two types (*i.e.*, clayey and sandy) of soil in different quantities and treatment conditions. The results showed that thermal treatment is an important prerequisite as well as air-drying (*i.e.*, hardening), and produced higher strengthening (up to 12 MPa) and durability in an immersed condition. Moreover, gellan gum is preferable to agar gum for soils with significant fine contents due to the interaction (*e.g.*, hydrogen bonding) between biopolymers and fine particles, which produces firm biopolymer–soil matrices. Consequently, thermo-gelation biopolymers have strong potential application as construction materials for both land (*i.e.*, dry) and waterfront purposes.

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1. Introduction

Studies on ground improvement have been performed and utilized throughout the history of human civilization. With the current high demand for civil infrastructure, ground improvement techniques have become an important element in geotechnical engineering projects. Such techniques have been developed in accordance with advances in current technology and human resources and make many civil engineering projects feasible.

For admixture type ground improvements, materials such as cement, epoxy, acrylamide, phenoplasts, polyurethane, and glass water are typically used for soil improvement [1]. These materials,

however, give rise to environmental concerns owing to their harmful nature [2,3]. The development of eco-friendly materials for soil improvement is thus necessary.

Biopolymers are biodegradable polymers produced by living organisms such as algae, fungus or bacteria. They consist of polysaccharides, which are compounds consisting of monosaccharides linked at certain locations. They are broadly distributed in nature and serve as skeletal structure-forming substances, assimilative reserve substances, and water-binding substances [4]. With their natural behavior, polysaccharides act as thickening agents, stabilizers, sweetening, and gel-forming agents. Accordingly, most applications utilizing biopolymers are in the fields of food production, agriculture, cosmetics, medicine, and pharmaceuticals [5–7]. Recently, some researchers have studied the utilization of biopolymers in the field of geotechnical engineering [8–10]. Also, recent studies have shown that biopolymers such as β -1,3/1,6-glucan,







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xanthan gum, can successfully improve the mechanical properties [11,12] and erosion resistance of soil, while remaining the porosity and hydraulic conductivity high [13]. Compared to cement mixture improvement, biopolymer-treated soils have higher strengths, even though the amount of biopolymer used is less than the amount of cement in the soil. Moreover, in terms of economic costs, based on the costs of materials and pollution effects, biopolymer-treated soils have advantages over cement-treated soil [11].

Nevertheless, the durability of biopolymer-treated soil against water has not yet been comprehensively discussed by researchers and remains uncertain. Therefore, this study introduces a thermogelation biopolymer group as a new construction material with the aim of improving the durability of soil immersed in water, and to provide high performance building components such as bricks, blocks, and indoor finishing materials. The biopolymers used in this study are agar gum and gellan gum.

2. Material and methods

2.1. Materials

2.1.1. Agar gum

Agar gum is a biopolymer composed of polysaccharides (complex sugar) made of linked galactose molecules. Agar gum is a galactan made of alternating sequences of (1–4)-linked 3,6-anhydro- α -t-galactose and (1–3)-linked β -D-galactose residues [14]. Agar gum has been used as a food additive material since it was discovered decades ago. Its properties as a stabilizer, thickener, emulsifier, flavor enhancer, and absorbent make it one of the more important additives for food products, including use in bakery products, confections, dairy products, and meat and fish products by forming gels to thicken and provide texture [15]. In addition, agar gum is also used in the fields of dentistry, microbiology, and medicine [16].

The agar gum used in this study was manufactured by Samchun Chemical Company. Essentially, agar gum is extracted from *rhodophyceae* (an algae group), such as the *Gelidium* and *Gracilaria* species [16], and the biopolymer is highly dependent on the species and environmental conditions of the source *algae*. Agar gum is a hydrophilic colloid that consists of two different compounds: a firmly gelling complex sugar called *agarose* and a weakly gelling charged polymer called *agaropectin* [17]. Agar gum starts to dissolve in boiling water at 85 °C and forms a gel when cooled to 32-43 °C [15]. It has a neutral polymer chain with a limited reactivity to other materials. Agar gum is white to pale yellow, shiny, semi-translucent, tasteless, and odorless [17].

2.1.2. Gellan gum

Gellan gum, usually employed as a substitute gelling agent for agar gum, is a high molecular weight polysaccharide fermented from the *Spingomonas elodea* microbe (formerly known as *Pseudomonas elodea*). Gellan gum is a linear anionic heteropolysaccharide made of four molecules: (1,3)- β -D-glucose, (1,4)- β -D-glucoronic acid, (1,4)- β -D-glucose, and (1,4)- α -L-rhamnose [18]. It has the properties of a thickening or gelling agent and is often used as a food additive. Products using gellan gum are typically confections, bakery fillings, dairy products, jams and jellies, microwavable foods, puddings, and toppings [16].

Gellan gum's chemical structure contains four linked monosaccharides: one molecule of rhamnose that can be found in plants, one molecule of oxidized glucose named glucuronic acid, and two molecules of glucose (common sugar). In usual commercial production, gellan gum is modified to become a deacylated polymer that is soluble in water at temperatures above 90 °C, and it form gels when suitable cations are present or when cooled to gelling temperatures (*i.e.*, 30–70 °C depending on the gel concentration, presence of cations, and cooling rate) [19,20].

2.1.3. Agar gum and gellan gum gels

Hydrogen bonds play an important role in the formation and structure of agar gum and gellan gum gels during thermo-gelation. Before gelation, continuous stirring and heating in water allows the biopolymer chains to disperse thoroughly and form a hydrocolloid solution. Then, as the solution cools, the biopolymer chains twist together and form double helices with other chains or molecules to form a rectangular matrix via hydrogen bonding [21,22].

The compressive strengths of pure agar and gellan gum gels have been reported to show similar values (*i.e.*, 30–50 kPa) in low concentration ranges (*i.e.*, 1–2%) [23]. We prepared pure gel cubes (*i.e.*, 40 mm × 40 mm) in the laboratory with higher concentration values (*i.e.*, $w_b/w_w = 2-8\%$, where w_b/w_w is the weight of biopolymer to the weight of water) by dissolving biopolymers into heated (*i.e.*, 100 °C) distilled water. The hot biopolymer solution coagulated to a firm gel upon cooling and the compressive strength was immediately measured when the gel temperature reached room temperature.

Fig. 1 shows that the compressive strength of agar gels remains constant (*i.e.*, 50 kPa) above 2% concentration, while the compressive strength of the gellan gel increases significantly up to 300 kPa at $w_b/w_w = 7.5\%$. Thus, it is expected that gellan

gum would be more effective in soil strengthening in higher biopolymer concentrations (*i.e.*, w_b/w_w). However, the reduced workability of gellan gum (*e.g.*, thorough mixing) due to the increased viscosity and gelation temperature (*i.e.*, faster gelation) induced by higher gellan concentration becomes a drawback [24].

2.1.4. Clayey soil (Korean residual soil)

Red Yellow Soil is a Korean residual soil formed from the weathering of rocks, and is locally known as hwangtoh due to its reddish-yellow appearance. In this study, Red Yellow soil was obtained from Gochang, Korea. It mostly consists of silt, halloysite, kaolinite, quartz, and illite. It has a plastic limit and liquid limit of 35% and 52.4%, respectively, and a specific gravity of 2.60. The soil can be classified as sandy lean clay (*CL*) due to its particle size distribution (*i.e.*, $D_{60} = 0.065$ mm, $D_{50} = 0.055$ mm, and $D_{10} = 0.040$ mm), as shown in Fig. 2. Further descriptions of Red Yellow soil can be found in Chang and Cho [11]. Red Yellow soil represents clayey soil in this study.

2.1.5. Sandy soil

Sandy soil used in this study was surface soil sampled from Cheonan, Korea. By particle size distribution data (Fig. 2) sandy soil is classified as poorly graded sand with silt (SP–SM). With a C_u (uniformity coefficient) of 3.02 and C_c (coefficient of gradation) of 0.92 due to its grain size characteristics (*i.e.*, $D_{60} = 0.56$ mm, $D_{50} = 0.031$ mm, and $D_{10} = 0.16$ mm), sandy soil represents coarse type soil in this study.

2.2. Experimental program

2.2.1. Thermo-gelation biopolymer-soil mixing

The biopolymers were dissolved in hot (*i.e.*, 100 °C) distilled water to prepare a thermally-treated biopolymer solution that was suitable for mixing due to its low viscosity [15,20]. The biopolymers were poured into a 100 °C solvent and stirred constantly via a magnetic stirrer (WiseStir® MSH-20D) at the desired biopolymer



Fig. 1. Compressive strength of pure gellan gum and agar gum gels with different biopolymer to water concentrations (w_b/w_w) .



Fig. 2. Particle size distribution of clayey soil and sandy soil.

Table 1

Specimen mixing and caring conditions.

Soil	Biopolymer		Caring conditions	
	Туре	w_b/w_s		
Clayey soil	Gellan gum	1%, 3%	Case 1. Air-dried (room temp: 20 ± 2 °C)	
	Agar gum	1%, 3%	Case 2. Submerged (water temp: 18 ± 2 °C)	
Sandy soil	Gellan gum*	1%	Case 3. Dried after submersion of 28 days	
	Agar gum	1%, 3%	Case 4. Submerged after air-drying of 28 days	

 * 3% gellan gum mixing condition is avoided for sandy soil due to the extremely high viscosity of $w_b/w_w = 10\%$ gellan gum solution, which is insufficient for thorough mixing.

concentration (*i.e.*, w_b/w_w), which was determined by the biopolymer to soil ratio in weight (*i.e.*, w_b/w_s) and the initial water content (*i.e.*, w_w/w_s) for proper biopolymersoil mixing. The initial water content for mixing was set at 60% for clayey soil and 30% for sandy soil. For both agar and gellan gum biopolymers, 1% and 3% w_b/w_s conditions were considered in this study. Thus, clayey soil was mixed with $w_b/w_w = 1.6\%$ and 3.3% biopolymer solutions, while 5% and 10% biopolymer solutions were prepared for sandy soil.

Both soils were heated (over 60 °C) before mixing to prevent rapid cooling due to the temperature difference between soils and biopolymer solutions. Four different types of mixtures were prepared: clayey soil-agar gum, clayey soil-gellan gum, sandy soil-agar gum, and sandy soil-gellan gum (Table 1).

The mixing process was quickly performed on an isothermal hotplate (90 °C) to minimize temperature loss so as not to allow the biopolymer to cool and form a gel while mixing. Hot soil-biopolymer mixtures were molded into cubic (40 mm × 40 mm) molds. Vibration (600 rpm) was applied for proper specimen compaction and a spatula was used to flatten the surface. The specimens were then placed at room temperature to be cooled and stabilized through biopolymer gelation. With cooling, the biopolymer solutions change from a solution into highly viscous biopolymer gels, which are formed within soil specimens via interparticle interaction (e.g., hydrogen bonding). Finally, the specimens were extracted from the mold and subjected to the specific conditions described in the next section.

The $w_b/w_s = 3\%$ case of gellan treated sandy soil mixture was not used due to the extremely high viscous gellan gum solution (*i.e.*, $w_b/w_w = 10\%$), which causes poor non-uniform mixing due to the immediate gelation induced by the high gellan concentration (*i.e.*, higher gellan concentration increases the gelation temperature) [20,24,25]. No problems were encountered with high concentration (*i.e.*, $w_b/w_w = 10\%$) agar gum solution mixing with sandy soil.

2.2.2. Specimen caring condition

Generally, the hydrogel strength of thermo-gelation biopolymers strongly depends on the water content [26]. Thus, the prepared specimens were treated according to four different conditions: (1) air dried (in open air at room temperature, 20 ± 2 °C), (2) submerged (set under 18 ± 2 °C water), (3) submerged and air dried, and (4) air dried and submerged. These conditions were set to evaluate the water content dependency and hydro-irreversibility of the strength of the thermo-gelation biopolymers treated soils. For the air dried case and the submerged case, the unconfined compressive strength of the specimens was measured after 1, 3, 7, 14, 21, and 28 days of treatment. After 28 days of treatment, air dried specimens (*i.e.*, case 1) were submerged under water for 7 days (*i.e.*, case 4), while submerged specimens (*i.e.*, case 2) were air dried for 28 days (*i.e.*, case 3). The unconfined compressive strengths of case 3 and 4 specimens were also measured. Details of specimen mixing and treatment conditions are summarized in Table 1.

2.2.3. Biopolymer-soil mixing without thermal treatment

Clayey soil specimens were also prepared without thermal treatment to evaluate the effects of thermal treatment for case 1 (air dried) and case 4 (submerged after drying of 28 days). Both agar gum and gellan gum were dissolved in distilled water and mixed with soil at room temperature without any heat source. The w_b/w_s of biopolymer–clayey soil specimens (*i.e.*, 40 mm × 40 mm × 40 mm cube) was controlled to be 1% and the initial water content was set to be 60%.

2.2.4. Uniaxial compressive test on biopolymer-treated soil

The unconfined compressive strengths of biopolymer-treated soil specimens were measured by a universal testing machine (UTM) device (INSTRON 5583). Three specimens were used for a single test condition (*i.e.*, soil and biopolymer type, biopolymer content, treatment condition and time) and their average was taken to represent the unconfined compressive strength of the biopolymer-treated soils. Axial strain was controlled at 1%/min, and stress–strain relationships were obtained automatically. Submerged specimens were extracted from water immediately

before testing. Wet specimens could be subjected to a compression test without any trimming or external covering (*e.g.*, membrane) as they were self-standing and had negligible moisture loss (*i.e.*, less than 2% during 30 min of exposure).

2.2.5. Microscopic observation

Microscopic observations were performed to understand the interactions between biopolymers (*i.e.*, agar gum and gellan gum) and soil particles inside the soil, using a scanning electron microscope (SEM; Magellan400) to take images showing the inter-particle structure and connection between the soil and biopolymer.

3. Results and analyses

The compressive strength of the thermo-gelation biopolymertreated soil was investigated in terms of treatment time and conditions (*i.e.*, de-hydration conditions), biopolymer type and concentration (*i.e.*, w_b/w_s = biopolymer weight/soil weight), and soil type via analyses of experimental findings, as described in the following sections.

3.1. Effect of thermo-gelation

Fig. 3 shows the stress-strain behaviors of clayey soils for different biopolymer treatment methods (*i.e.*, non-treated, 1% gellan without thermal, and 1% gellan with thermal) after 28 days of drying at room temperature.

Dried natural (*i.e.*, without any additives) clayey soil shows a compressive strength of 1050 kPa, while both 1% gellan and agar gum without thermal treatment enhanced the soil strength up to



Fig. 3. Stress–strain relationship of clayey soil specimens with different biopolymer treatments. (a) Gellan gum treatment. (b) Agar gum treatment. Biopolymer treatment shows higher strength than natural dried soil and thermal treatment shows maximum enhancement, whereas gellan gum shows a higher strengthening performance than agar gum treatment.

Table 2			
Peak strength and el	astic modulus (E_{50}) values of 1% biopolyme	er-treated clayey soils after 28 days of dryin	ng at room temperature.
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Biopolymer	Peak strength [kP	Peak strength [kPa]			E ₅₀ [MPa]			
	Natural (0%)	1% mixed	1% mixed		1% mixed			
		Without thermal	With thermal		Without thermal	With thermal		
Gellan gum Agar gum	1050 1050	2500 1800	4660 3190	53 53	265 188	466 270		



Fig. 4. Strengthening behavior of natural and 1% gellan gum mixed (with and without thermal treatment) clayey soils. Thermal treatment produces higher strength values regardless of water content variation.

2500 kPa (gellan gum) and 1800 kPa (agar gum). Meanwhile, thermal treatment produced higher strengthening (*i.e.*, 4660 kPa for gellan gum and 3190 kPa for agar gum), and caused the soil to become more brittle with higher elastic modulus values (Table 2).

Fig. 4 shows the strengthening behavior of natural clayey soil, 1% gellan gum mixed clayey soil without thermal treatment, and 1% gellan gum mixed clayey soil with thermal treatment during

28 days of dehydration (*i.e.*, decreasing water content). The unconfined compressive strength of clayey soil with 1% gellan gum mixed at room temperature was double that of natural soil, while thermal treatment promoted the strengthening effect of the gellan gum biopolymer twofold above that of the soil without thermal treatment. Therefore, heating should be an important concern in the usage of thermo-gelation biopolymers for soil treatment in construction and building engineering purposes.

Thermal treatment produced a more significant effect when 1% gellan gum mixed clayey soil specimens (*i.e.*, without and with thermal treatment) were immersed in water after 28 days of air drying (*i.e.*, case 4). Clayey soil–gellan gum mixtures without thermal treatment stated to dissolve immediately (Fig. 5a) and became fully degraded after 2 h of submergence (Fig. 5b). However, clayey soil–gellan gum mixtures with thermal treatment (Fig. 5c) retained their shape during 7 days of submergence (Fig. 5d) with a volumetric swelling ($\Delta \varepsilon_v = 0.1$) due to the volume expansion of the gellan gels in soil via water adsorption. Therefore, we can conclude that thermal treatment is a necessary and effective condition for soil treatment using agar and gellan gum biopolymers.

3.2. Effects of soil type, biopolymer, and biopolymer content

Fig. 6 shows the strengthening behavior of air dried biopolymer-treated clayey soils (a) and sandy soils (b) with time. The compressive strength of both thermo-gelation biopolymer-treated clayey soil and sandy soil increased significantly with up to 7 days



Fig. 5. Difference on durability under submergence. Gellan gum-clayey soil specimens without thermal treatment completely dissolve after 1 day (a and b). However, specimens prepared via thermal treatment (c) remain stable even after 28 days of immersion (d).

of drying, and showed a relatively low amount of strengthening afterwards. It is obvious that higher biopolymer content renders higher compressive strength values.

For clayey soil (Fig. 6a), gellan gum always shows higher strength than agar gum under the same w_b/w_s conditions (both 1% and 3%). Gellan gum is a repeating tetrasaccharide connected by α -1.3-glycosidic bonds having numerous hydroxyl or hydrogen edges [27], which are suitable for binding with clayey particles, while the combination of agarose and agaropectin forming agar gum [28] reduces points for external binding. It can thus be concluded that gellan gum is more suitable for forming firm biopolymer-clayey soil matrices than agar gum, regardless of biopolymer content.

Meanwhile, sandy soil shows similar strength values at w_b/v_b $w_{\rm s}$ = 1% for both biopolymer types (*i.e.*, agar gum and gellan gum) (Fig. 6a). This can be explained in terms of the absence of interaction between soil particles and biopolymers. Electrically neutral sandy particles do not have direct interaction with biopolymers, while in clayey soil, clay interacts directly with biopolymer molecules via hydrogen bonding [25]. In sandy soil, biopolymers coagulate, forming a surface coating on sandy particles, filling pore spaces, or enlarging particle-to-particle contact areas. Moreover, for dried sandy soil, biopolymer gels form thin coatings on sand particle surfaces with slight adhesion. Thus, mechanical friction becomes more critical than the inherent strength of the biopolymer gels. Therefore, sandy soil can be expected to show similar compressive strength values regardless of the biopolymer type in a dried condition.

The compressive strengths of thermo-gelation biopolymer-treated soils under a submerged condition remained almost constant regardless of time (Fig. 7). The compressive strength values of thermo-gelation biopolymer treated clayey soil and sandy soil were remarkably higher than those of non-treated natural soils, which could not even be measured with the UTM device. In contrast with the strength behavior of dried soil, submerged specimens show similar strength values $(150 \pm 60 \text{ kPa})$ regardless of soil type. The water content of submerged specimens remained constant at the initial water content at specimen mixing (i.e., 60% for clayey soil and 30% for sandy soil), which indicates that biopolymer gels also remained constant between soil particles without dehydration. Therefore, the compressive strength of pure biopolymer gels shown in Fig. 1 becomes an important parameter in the strengthening behavior of saturated biopolymer-soil mixtures.

In detail, the w_b/w_w for $w_b/w_s = 1\%$ and 3% clayey soil treatment was 1.6% and 5.0%, and 3.3% and 10% for sand treatment, respectively. The strengths of $w_b/w_w = 1.6\%$ agar gum and gellan gum gels were almost equal (*i.e.*, 40 kPa) [23], while $w_b/w_w = 5\%$ gellan gum gel (170 kPa) was higher than $w_b/w_w = 5\%$ agar gum gel (70 kPa) (Fig. 1), resulting in the same strength order observed with thermo-gelation biopolymer treated clayey soils in Fig. 7(a) (i.e., gellan gum 3% > agar gum 3% > gellan gum 1% > agar gum 1%).

For sandy soil (Fig. 7b), the strength of $w_b/w_w = 3.3\%$ agar gum gel (60 kPa) is higher than the $w_b/w_w = 1.6\%$ (40 kPa) applied for clayey soil. Thus, the strength of $w_b/w_s = 1\%$ agar gum treated saturated sand is higher than $w_b/w_s = 1\%$ agar gum or gellan gum treated clayey soils. Meanwhile, the strength of $w_b/w_w = 3.3\%$ gellan gum gel (100 kPa) is slightly higher than $w_b/w_w = 10\%$ agar gum gel (75 kPa) (Fig. 1). This demonstrates that the strength of $w_b/$ $w_s = 1\%$ gellan gum treated sandy soil is similar to or slightly higher than the strength of $w_b/w_s = 3\%$ agar gum treated sandy soil, under saturation.



soil (a) and sandy soil (b) dried in air at room temperature (20 ± 2 °C).



Fig. 7. Unconfined compressive strength with time for biopolymer treated clayey soil (a) and sandy soil (b) placed in submerged condition (temp: 18 ± 2 °C).

15000

(a) Clayey soil

∎Gellan 3%

3.3. Effect of water content variation

Submerged specimens (*i.e.*, agar gum and gellan gum treated clayey and sandy soils) were subjected to an additional 28 days of drying at room temperature. The compressive strength of the submerged to air dried specimens was increased by the drying process, as shown in Figs. 8 and 9, in accordance with the general trend of compressive strength-water content variation seen for the normally dried condition (*i.e.*, case 1; Fig. 6).

The submerged zones in Figs. 8 and 9 indicate the highest water content conditions of submerged specimens (case 2), whose compressive strength values were identical to the results in Fig. 7. As specimens undergo the drying process (both in the normally dried (case 1) and when dried after submersion (case 3)), the increase in compressive strength follows a single compressive strength-water content trend regardless of the time delay (*i.e.*, 28 days) between case 1 and 3 specimens. This applies for both gellan gum and agar gum treated clayey soil and sandy soil.

Meanwhile, when air dried specimens were submerged for 7 days (case 4), the specimens absorbed the surrounding water, as both agar gum and gellan gum are hydrophilic biopolymers [29]. The compressive strength decreased gradually due to the water absorption during the submersion; however, the trend of compressive strength-water content during submersion does not match the trend of compressive strength-water content for drying conditions (*i.e.*, cases 1 and 3).

The final compressive strengths of clayey soil specimens (*i.e.*, case 4) were indeed higher than those of normally submerged

specimens (*i.e.*, case 2), while sandy soil specimens showed degradation of strength to a level even below that of case 2 specimens (Table 3). For thermo-gelation biopolymer-treated soils, the volumetric shrinkage that occurs during the air drying process produces a denser soil structure due to the enhanced biopolymer gel-soil particle interactions inside the soil. Thus, the potential amount of water that the specimen can absorb during submergence will be less than its initial water content, because the rigid structure of the thermo-gelation biopolymer-treated soil is irreversible. The pore filling process of hydro-expanded biopolymer gels will be further limited because the firm biopolymer matrices formed during normal air drying increase the inter-particle resistance to volumetric expansion.

3.4. Interaction between thermo-gelation biopolymers and soils

Fig. 10 shows Scanning Electron Microscope (SEM) images of untreated (a), gellan gum treated (b and c), and agar gum treated (d) clayey soil. As shown in the figure, the gellan gels inside the soil appear to create web-like matrices that interact directly with the soil particles. On the other hand, agar gum appears to form massive soil-biopolymer aggregates via gelation. This difference is due to the differences in the molecular structure of the gellan and agar gums. Gellan gums have a shorter molecular structure and are charged, and consequently direct interactions with fine soil particles (*e.g.*, hydrogen bonding) are achievable [25]. However, agar gums have a longer molecular structure, making direct interactions



Fig. 8. Change in compressive strength of gellan gum (a) and agar gum (b) treated clayey soil in relation to water content and treatment conditions (drying and submerging). Two zones are shown to represent the compressive strength of the specimens under particular treatment conditions.



Fig. 9. Change in compressive strength of gellan gum (a) and agar gum (b) treated sandy soil with relation to water content and treatment conditions (drying and submerging). Two zones are shown to represent the compressive strength of the specimens under particular treatment conditions.

Table 3

Strength [kPa] Soil Biopolymer	Case 2 (submerged)				Case 4 (air-dried \rightarrow submerged)			
	Clayey soil		Sandy soil		Clayey soil		Sandy soil	
	1%	3%	1%	3%	1%	3%	1%	3%
Gellan gum	73	230	250	-	210	520	51	-
Agar gum	64	138	148	245	185	534	58	75

Compressive strength values of normally submerged (case 2) and air-dried to submerge (case 4) biopolymer mixed clayey and sandy soil specimens.

more difficult. This structure causes agar gums to coat and coagulate soil particles, because the longer structures wrap around the soil particles [14].

This implies gellan gum will result in higher strength than agar gum treatment of clayey soil given the same biopolymer content (*i.e.*, w_b/w_s). Clayey soil is a lean clay (CL) with fine content up to 70%, while sandy soil is a poorly graded sand with silt content less than 5% (SP–SM). Thus, gellan monomers can form firm gellan gum–clayey soil matrices via microscopic interactions (*e.g.*, hydrogen bonding), producing higher strengthening than agar gum for both air dried (Fig. 6a) and saturated (Fig. 7a) conditions.

Meanwhile, sandy soil has relatively high pore sizes due to its particle size distribution. In a submerged condition, it can be assumed that the volume of biopolymer gels approximately corresponds to the volume of voids via pore filling. Therefore, the strength should be governed by the strength of the biopolymer gels (Fig. 1) inside the soil, for saturated coarse soil. This explains why the strength with 1% gellan gum treatment is higher than that obtained with 1% and 3% agar gum treatment in saturated sandy soil (Fig. 7b).

From Fig. 7 and Table 3, it is seen that the thermo-gelation biopolymer-treated soils still have compressive strengths even though they are in a high water content condition. When the test is performed under unconfined conditions, no soil (especially sandy soil) can maintain its structure at high moisture conditions, and it can thus be concluded that pure soils do not have compressive strengths under such conditions. Therefore, the strengthening induced by thermo-gelation biopolymer gels inside soil should be highlighted.

3.5. Discussions

Generally, the compressive strengths of clayey soil and sandy soil increased with higher biopolymer content for both agar gum and gellan gum treatment (Figs. 6 and 7). In a submerged condition, the approximately constant strength difference between 1% and 3% biopolymer treatments (for each agar and gellan gum) regardless of dehydration and volumetric shrinkage, indicates that biopolymer content to soil (w_b/w_s) is an important strengthening parameter for thermo-gelation biopolymers.

The order of magnitude of compressive strength for both air dried and submerged cases shows that gellan gum is much better than agar gum in soil strengthening due to the higher strength of the former, as already shown in Fig. 1. Meanwhile, the compressive strength of thermo-gelation biopolymer-treated clayey soil is lower than that of sandy soil for the same amount of w_b/w_s in a submerged condition, which appears to be affected by differences in initial water content and soil type.

For saturated soils, the higher initial water content of clayey soil (*i.e.*, 60%) produced a looser particle composition and lower biopolymer gel concentration (*i.e.*, w_b/w_w) inside the soil. Moreover, the thermo-gelation biopolymer–clayey soil mixture exists as a well-dispersed slurry, while the biopolymer–sandy soil mixture becomes a composite of coarse solid particulates and viscous gels. Thus, the strengthening mechanism for thermo-gelation biopolymer–treated sandy soil involves both inter-particle friction and

adhesion, while clayey soil shows an undrained shear behavior governed by the viscosity of the biopolymer–clayey soil mixture. Furthermore, drying accompanies closer bonding distance via volumetric shrinkage and absence of water from the soil, which induces stronger and more hydrogen bonding between biopolymers and clayey soil particles, rendering higher strength, as shown in Figs. 6 and 10.

It can be deduced that the compressive strength of thermo-gelation biopolymer-treated soils is mainly governed by the water content instead of the treatment time, as shown in Figs. 8 and 9. Under a normal air dried condition, the water content decreased, and this resulted in an increase in compressive strength. Meanwhile, under a normal submerged condition, because the environment did not permit evaporation, the water content remained almost constant (*i.e.*, 60% and 25% for clayey soil and sandy soil, respectively), resulting in constant compressive strength regardless of the submersion time. When submerged specimens were dried after a 28 day delay, the compressive strength-water content behavior followed the general trend that had been previously observed.

Regardless of the treatment time, the compressive strength complies with the water content of the specimen, and consequently, 'treatment time' was determined to not be a significant parameter in the scope of this study. Thus, biopolymer treatment of soil for construction engineering has the advantage of immediate effectiveness, while cement treatment requires a relatively long time for hydration.

The presence of water plays an important role in the uniform dispersion of biopolymer in the soil mixture before thermo-gelation. During cooling, biopolymers form double helix hydrogels between soil particles to form a firm biopolymer–soil matrix. During dehydration, the loss of water increases the concentration (*i.e.*, w_b/w_w) of the biopolymer hydrogel, inducing overall soil strengthening as shown in Figs. 1 and 4. In this case, the ultimate strength of the biopolymer hydrogels and the interaction between the hydrogels and soil particles (*e.g.*, hydrogen bonding) governs the overall strength of the biopolymer–soil mixture.

When a dried biopolymer–soil mixture is subjected to re-wetting (*i.e.*, case 4), dried biopolymers swell via hydrophilic water absorption, reducing the overall strength of the biopolymer–soil mixture. However, clayey soil shows less reduction than sandy soil compared to their initial (*i.e.*, submerged, case 2) condition due to their different structures.

In practice, with drying, clayey soil experiences a large change in dry density (γ_d) (*i.e.*, 0.9–1.8 g/cm³, Fig. 11a), irreversibly affecting its soil water content recovery and resulting in a final water content (*i.e.*, 30 ± 5%, Fig. 8) after re-wetting that is markedly lower than the initial water content (*i.e.*, 60%). Meanwhile, sandy soil displays only a slight rise in dry density (*i.e.*, 1.5–1.6 g/cm³ in average, Fig. 11b), resulting in a recovered final water content after 7 days of re-wetting that is close to the initial water observed in the submerged zones in Fig. 9.

Moreover, the strength values in Table 3 indicate that strengthening efficiency and durability of thermo-gelation biopolymer treatment is maximized in clayey soil, especially with preliminary hardening (*i.e.*, dehydration). The severe reduction in strength of sandy soil indicates that sandy soils are more sensitive to the struc-



Fig. 10. SEM images of (a) natural (non-treated), (b and c) gellan gum treated, and (d) agar gum treated clayey soil.

tural disruption of the biopolymer-soil matrix caused by re-wetting, due to the absence of direct hydrogen bonding between the biopolymers and coarse soil particles.



Fig. 11. Change in compressive strength and dry density (*i.e.*, structural composition) of thermo-gelation biopolymer-treated clayey soil (a) and sandy soil (b).

Nevertheless, the reduction in strength caused by re-wetting of thermo-gelation biopolymer treated clayey soil is less than the reduction exhibited by ordinary fly ash stabilized clays [30]. Moreover, the clayey soils subjected to air-drying and submergence (case 4) showed higher strength than submerged only (case 2) clayey soils, while sandy soil showed a higher reduction in strength with re-wetting (Table 3). This indicates the importance of the formation for hydrogen bonding between thermo-gelation biopolymers and clay particles. Further studies are recommended to investigate the biopolymer and soil particle interactions, their enhancement, and assessment of the durability of thermo-gelation biopolymer treated soils subjected to wetting and drying.

It should be noted that the hydrogels of agar gum and gellan gum are thermo-reversible [22]. However, the gel–sol transition temperature of melting is higher than that of cooling (*i.e.*, setting). More specifically, the melting temperature of low-acyl gellan gum is higher than 70 °C, while the set temperature is below 45 °C [16]. Thus, for soil treatment purposes we expect a stable thermal environment for thermo-gelation biopolymers, because the annual maximum subsurface soil temperature (in summer) is known to be less than 40 °C in most areas, expect arid deserts [31].

Finally, the strength of thermo-gelation biopolymer treated soils are clearly higher than non-treated natural soils, regardless of the water content. Therefore, thermo-gelation biopolymer treatment can be possibly applied for environmentally-friendly purposes such as soil erosion control on waterfronts and slope or embankment protection.

4. Conclusions

This study addressed the strengthening behavior of thermogelation biopolymers to facilitate their use as a construction material for soil improvement. Agar gum and gellan gum were used as thermo-gelation biopolymers, and clayey soil and sandy soil were used to represent fine-type soil (CL) and coarse type soil (SP–SM), respectively. Through a series of experimental and analytical studies, the factors influencing the strengthening behavior of thermo-gelation biopolymer-treated soils were discussed and the main findings are summarized as follows:

Thermo-gelation biopolymer soil treatment has significant impacts, enhancing soil strength and soil durability. Thermal treatment induces higher strength as well as structural stiffness. Moreover, thermally treated specimens showed high structural durability under immersion, which is a remarkable characteristic compared to the durability provided by existing soil treatment/ improvement methods.

- Time is not an important treatment consideration, while water content is a critical strengthening parameter for thermo-gelation biopolymer soil treatment. More specifically, as the water content decreases, the compressive strength increases, and if the water content remains constant, so does the compressive strength of thermo-gelation biopolymer-treated soils. Meanwhile, thermal treatment and air drying of the thermo-gelation biopolymer-treated soils before submergence maximizes their structural stability when immersed in water. Therefore, thermal mixing and air drying are necessary pretreatments when applying thermo-gelation biopolymer-soil mixtures as construction materials for waterfront purposes.
- Biopolymer and soil type compatibility is also an important finding from this study. Test results show that soil with higher fine content is more suitable for forming firm biopolymer-soil matrices due to the microscopic interactions (*e.g.*, hydrogen bonding) between fine particles and biopolymers.
- The structure of the initial biopolymer-soil matrix, which is determined by the initial biopolymer content (w_b/w_s) , is also an important consideration. Different biopolymer content changes the thickness and bonding characteristics of the thermo-gelation biopolymer gels inside the soil. The inter-particle interaction and bonding characteristics are determined by the biopolymer content inside the soil. Even though thermo-gelation biopolymers show similar dry density values after drving, the remarkably enhanced strength of clavey soil implies that interactions between biopolymers and fine particles (e.g., hydrogen bonding) is the major governing parameter of soil strengthening induced by thermo-gelation biopolymer treatment. Moreover, considering the strength behavior of pure thermo-gelation biopolymer gels, gellan gum treatment produces higher strengthening than agar gum mixing for high $w_b/$ $w_{\rm s}$ conditions regardless of the soil type and moisture content of thermo-gelation biopolymer-treated soils.
- As mentioned above, biopolymer type and quantity (*i.e.*, *w_b/w_s*), water content, and soil type are important parameters with regard to the strengthening behavior of thermo-gelation biopolymer soil treatment. Moreover, the drying and wetting process is also a non-negligible concern.

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