

Utilization of carrageenan as an alternative eco-biopolymer for improving the strength of liquefiable soil

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Abstract. The liquefaction of soil occurs when a soil loses strength and stiffness because of applied stress, such as an earthquake or other changes in stress conditions that result in a loss of cohesion. Hence, a method for improving the strength of liquefiable soil needs to be developed. Many techniques have been presented for their possible applications to mitigate liquefiable soil. Recently, alternative methods using biopolymers (such as xanthan gum, guar gum, and gellan gum), non-traditional additives, have been introduced to stabilize fine-grained soils. However, no studies have been done on the use of carrageenan as a biopolymer for soil improvement. Due to its rheological and chemical structure, carrageenan may have the potential for use as a biopolymer for soil improvement. This research aims to investigate the effect of adding carrageenan on the soil strength of treated liquefiable soil. The biopolymers used for comparison are carrageenan (as a novel biopolymer), xanthan gum, and guar gum. Then, sand samples were made in cylindrical molds (5 cm x 10 cm) by the dry mixing method. The amount of each biopolymer was 1%, 3%, and 5% of the total sample volume with a moisture content of 20%, and the samples were cured for seven days. In terms of observing the effect of temperature on the carrageenan-treated soil, several samples were prepared with dry sand that was heated in an oven at various temperatures (i.e., 20°C to 75°C) before mixing. The samples were tested with the direct shear test, UCS test, and SEM test. It can increase the cohesion value of liquefiable soil by 22% to 60% compared to untreated soil. It also made the characteristics of the liquefiable increase by 60% to 92% from very loose sandy soil (i.e., $\phi=29^\circ$) to very dense sandy soil. Carrageenan was also shown to have a significant effect on the compressive strength and to exceed the liquefaction limit. Based on the results, carrageenan was found to have the potential for use as an alternative biopolymer.

Keywords: biopolymer; carrageenan; liquefaction; liquefiable soil; soil strengthening

1. Introduction

Soil liquefaction is a process that transforms saturated sand into a non-cohesive state as a result of repetitive shakings, such as that caused by earthquake waves (Syukri 2011). When co-seismic surface movements exceed a threshold value, liquefaction occurs in places prone to major earthquakes that are composed of saturated sand layers with low density (Tsaparli *et al.* 2016). This promotes a decrease in the soil's bearing capacity, and consequently, produces significant soil degradation. Therefore, a technique for enhancing the strength of liquefiable soil must be developed.

Numerous approaches for mitigating liquefiable soil have been proposed, including induced partial saturation

(Eseller-Bayat *et al.* 2011) and bio-chemical grouting (Baharuddin *et al.* 2013, van Paassen *et al.* 2009). Bio-chemical grouting is seen as a potential technique for strengthening liquefiable soil. Those experiments on 100 m³ soil yielded shear strengths ranging from 1 Mpa to 12 MPa. However, unanswered questions remain regarding the utilization of bacterial cells in the biochemical grouting technique. Complex microbial activities make it difficult to deploy the approach on a commercial basis (Anbu *et al.* 2016). In addition, large quantities of chemicals suppress bacterial development (Nemati *et al.* 2005); hence, soil bacterial activity is ineffectual (Sharma 2016).

Increasing the strength of soil that has the potential to be liquefied can be also done by increasing the liquefaction resistance value. Research conducted by Simatupang and Okamura (2017), demonstrates that the increase in liquefaction strength owing to EICP treatment was mostly a function of the effectiveness of calcite formation that precipitated at the inter-particle contact points, rather than the total amount of calcite created. Hence it is required to develop a mechanism that can connect the inter-particles contact point between particles.

Recently, alternative methods using biopolymers, such as xanthan gum, guar gum, beta-1,3/1,6-glucan, dextran and gellan gum, non-traditional additives, have been introduced (Ayeldeen *et al.* 2017, Chang *et al.* 2015, Chang and Cho

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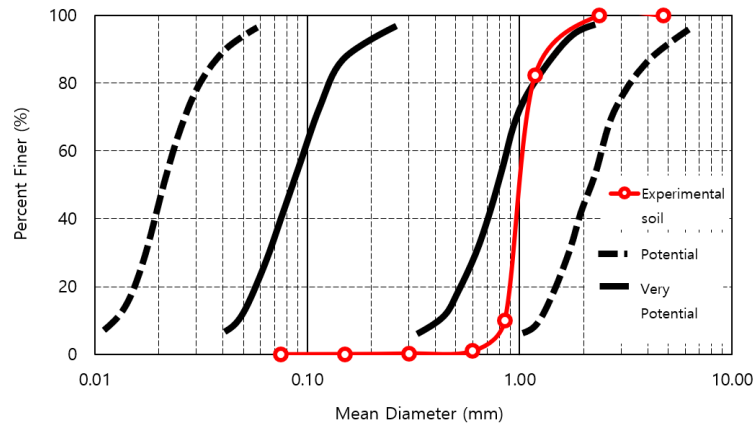


Fig. 1 Grain-size distribution of sandy soil used in this study

2014, Jeon *et al.* 2017). The results indicated the ability of xanthan gum, guar gum, and gellan gum to be used as improvement materials for soil treatment. They were chosen as materials for soil improvement mostly because they have highly viscous properties, so it is hoped that they can bind the soil particles and improve soil that has problems, especially in terms of strength (Chang *et al.* 2016).

Carrageenan is a hydrocolloid extracted from red seaweed. In the industry, carrageenan is also capable of possessing highly viscous qualities. Thus, in this case, carrageenan has rheological similarities to xanthan gum and guar gum. At room temperature, it has the remarkable ability to generate a variety of gel textures. In other industrial applications, it can be employed as a suspending, gelling, emulsifying, stabilizing, and water-retaining agent (Zia *et al.* 2017). Thus, carrageenan may have the potential to be utilized as a biopolymer for soil improvement based on its rheological and chemical structure. However, few investigations have been made into its application as a biopolymer for soil improvement.

In this study, several tests making comparisons among xanthan gum, guar gum, and carrageenan as potential materials were conducted to know the effect of adding carrageenan on the soil strength of treated liquefiable soil. By taking measurements through direct shear test (DS) and unconfined compressive strength (UCS) tests, the comparison of the reinforcing effect between the types of biopolymers and carrageenan on soil specimens was observed. A SEM analysis was also performed to investigate the macro- and micro-interactions of soil-biopolymer mixtures.

2. Materials and experimental procedures

2.1 Soil tested

Keisha sand No. 4 (coarse) is a natural silica sand produced in Oishida Town, Yamagata Prefecture, Japan (manufacturer: Tohoku Sisago Co., Ltd.). It is a white natural silica sand with round particles and good fluidity. Soil property tests were carried out to observe the characteristics of the sandy soil and to confirm that the soil

Table 1 Basic property values of Keisha sand no. 4 (liquefiable soil)

Parameter	Value
Specific gravity	2.630
Soil classification	Sandy soil is classified according to USCS as poorly graded sand (SP) because the C_u and C_c values do not meet the requirements
e_{max}	0.834
e_{min}	0.591
C_u	1.533
C_c	1.019
ρ_{dmax}	1.659 g/cm ³
ρ_{dmin}	1.438 g/cm ³
$D_{10}; D_{50}$	0.600 mm; 0.890 mm

had potentially liquefied. The results of the property values of the soil used in this study are shown in Table 1. Based on it, this sandy soil is classified according to the Unified Soil Classification System (USCS) as poorly graded sand (SP) with maximum void ratio (e_{max}), minimum void ratio (e_{min}), coefficient of uniformity (C_u), coefficient of curvature (C_c), and specific gravities (G_s) of 0.834, 0.591, 1.53, 1.02 and 2.63, respectively.

Fig. 1 shows the ranges in grain size distribution for the potential and high potential conditions of liquefiable soils based on Tsuchida (1970); the red line is the grain size distribution of the soil used in this study. The area within the two inner curves in the figure represents sands and silty sands, the soils with the lowest resistance to liquefaction, and the red line is in the inner curves. The graph also shows that the gradation curve tends to be upright, which means there is a slight/uniform variation in grain size. Poorly graded sand with a uniform grain size tends to form unstable soil grain configurations. In addition, the shape of the sand is well rounded, in an earthquake, the soil does not

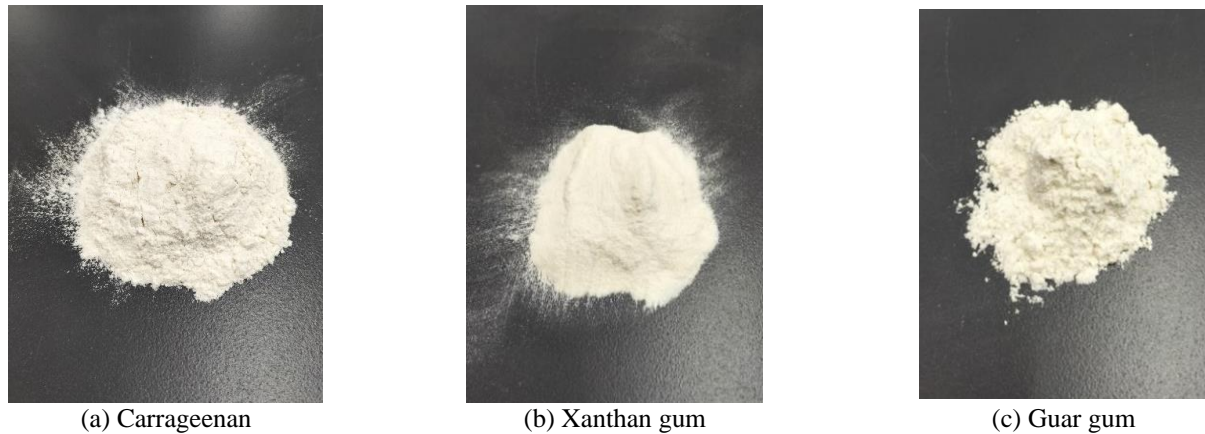


Fig. 2 Visualization of three kinds of biopolymers

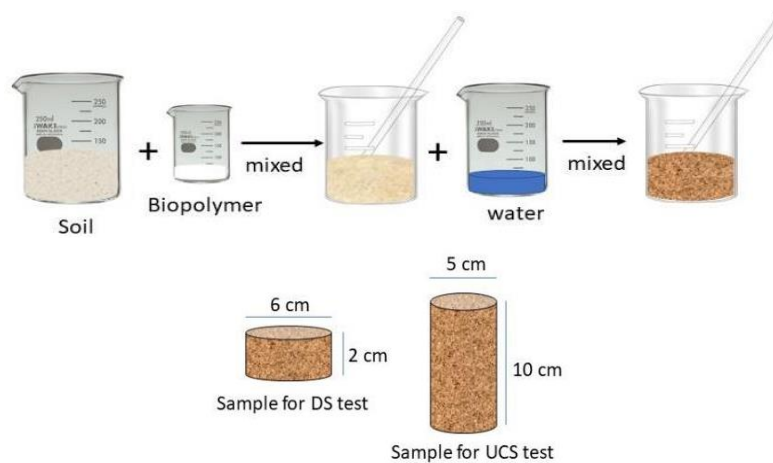


Fig. 3 Sample preparation of dry mixing for DS and UCS test

create an interlocking configuration. Thus, the soil can be released easily and the liquefaction process occurs. Based on the results of the property tests, therefore, the soil used in this study is liquefiable soil.

2.2 Carrageenan

Carrageenan is a hydrocolloid extracted from red seaweed belonging to the Eucheuma (*kappaphycus*), Chondrus, Gigartina, and Hypnea species. It is in the cell wall and intercellular matrix of seaweed plant tissue. It is a high molecular weight polysaccharide with a 15% to 40% ester-sulphate content. It is formed by alternate units of D-galactose and 3,6 anhydro-galactose (3,6-AG) joined by α -1,3 and β -1,4 -glycosidic linkage (Zia *et al.* 2017). Carrageenan's strong thickening, gelling, and stabilizing properties have led to its widespread use as a food additive over the past several decades (Artoft *et al.* 2008, Huang *et al.* 2007). According to a recent study, carrageenan has the potential to serve as a soil stabilizer (Nakamatsu *et al.* 2017). In this study, the carrageenan (κ) was obtained from IndoGum (Indonesia) with molecular weight is 788.65 g/mol. The biopolymers used in this study are shown in Fig. 2.

2.3 Specimen preparation

The main biopolymer used in this study was carrageenan (as a novel biopolymer). In addition, an untreated soil without any biopolymer (as a control), xanthan gum-treated soil, and guar gum-treated soil (both obtained from IndoGum, Indonesia) were also employed for comparison. In order to completely fill the intergranular spaces with biopolymer gel and achieve a fully saturated condition for the carrageenan-treated soil mixtures, based on the preliminary test results, the water content for mixing was fixed at 20% of the dry sand weight. Then, the liquefiable soil was mixed with the biopolymers using the dry mixing method according to Chang *et al.* (2015).

Different concentrations of biopolymers were applied (i.e., w/w=1%, w/w=3%, and w/w=5%), respectively. Purified biopolymers were thoroughly mixed with dried liquefiable soil before adding water (i.e., 20%). Then, the biopolymer-soil mixtures were placed into cylindrical (5 cm x 10 cm) molds for the unconfined compressive strength (UCS) test, and cylindrical (6 cm x 2 cm) molds for the direct shear (DS) test. Three samples were prepared for each condition. The soil was compacted in the molds in three layers with 25 blows per layer. After all the soil was



Fig. 4 Illustration of DS and UCS tests

placed in the molds, the samples were cured in mold at a temperature of $30^{\circ}\text{C} \pm 2$ for seven days. After curing time, there is a phase change as assessed by the change in the sample's saturation level. In the DS sample, the degree of saturation (S) ranges from 25% to 27%, and in the UCS sample, it ranges from 52% to 57%. In terms of the effect of temperature on the carrageenan-treated soil, several samples were prepared with dry sand (without biopolymers) that was heated in an oven at various temperatures (i.e., 20°C to 75°C) before the mixing process. The molding and mixing processes were the same as in the dry mixing method, but a laboratory hot plate was used for mixing in order to minimize the reduction in temperature. The experimental procedure for the dry mixing method is shown in Fig. 3.

2.4. Test procedure

The DS test is based on ASTM D3080 (Standard test method for direct shear tests of soils under consolidated undrained conditions) with normal loads of 8.84 kPa, 17.69 kPa, and 35.39 kPa. The direct shear test was carried out utilizing a square shear box. The biopolymer-treated sand samples, with a diameter of 6 cm and height of 2 cm, were placed into a direct shear test apparatus with porous stone above and below it. During the test process, the displacement due to shear forces and changes in the thickness of the specimen were recorded for each load. The test findings are depicted in a graph illustrating the connection between shear stress and shear displacement for each normal load.

The UCS test is based on ASTM D 2166 (Standard Test Method for Unconfined Compressive Strength of Cohesive Soil). All the geometric dimensions and the specimen mass were measured, and the specimen's top and bottom surfaces were trimmed slightly to prevent uneven stress distribution during testing. Samples were loaded to failure and their residual compressive strength was measured. Three measurements were averaged for each case, and all data points deviated from the mean by less than 5%. The testing processes of the DS and UCS tests are described in Fig. 4.

A JSM-7001FA device was also used to capture Scanning Electron Microscopy (SEM) images. To prevent electron scattering on the surface of the specimens, the samples were dried and coated with platinum (Pt) for 90 seconds using JEC-3000FC (JEOL Ltd. Japan) to prevent

charging. The interaction of the biopolymer with the soil was then documented.

The apparatus used for viscosity test was Set Rion Viscotester VT-03F using rotor no. 5 ($r=3.06$ cm). The shear rate used in this study was 4.35. The experimental conditions and procedures were following Webber *et al.* (2012). The same concentration applied for each kind of biopolymer (i.e., $w/w=1.5\%$) and dissolved it into 30 mL of water. The measurement of the viscosity value is carried out at various temperatures (i.e., 20°C , 60°C , and 75°C). Stirrer also used for mixing and also as a hot plate in order to minimize the reducing in temperature.

3. Strengthening parameters of carrageenan treated soil

3.1 Direct shear test result of biopolymer-treated soil

The shear strength of the carrageenan-treated soils at different concentration levels, obtained by direct shear testing, is shown in Fig. 5(a). In terms of a comparison with the existing biopolymers, Figs. 5(b) and 5(c) show the xanthan gum-treated soil and guar gum-treated soil, respectively. The parameters of the shear strength of the biopolymer-treated sample were then compared to the untreated sample reported in our previous study (Putra *et al.* 2018). The figures clarify that the shear strength values of the biopolymer-treated soil increase with the higher biopolymer contents for all kinds of biopolymers. The cohesion and internal friction angle values for the carrageenan-treated soil with different carrageenan contents (i.e., 1%, 3%, and 5%) are given in Fig. 6. The graph also shows the values for xanthan gum-treated soil and guar gum-treated soil for the sake of comparison. The concentration of each biopolymer is the same as the carrageenan content. The results in Fig. 6 show that the cohesion level of the treated soil tends to increase with an increase in the carrageenan content. For each sample, the increase in the cohesion value from that of the original untreated cohesionless soil (i.e., $c=0$ kPa) was in the range of 22% to 60%. Furthermore, the treatment of liquefiable soil using biopolymers seems to influence the internal friction angle. It causes an increase of 60% to 92% and a change from very loose sandy soil (i.e., $\phi=29^{\circ}$) to very dense sandy soil (Swiss Standard SN 670 010b). The

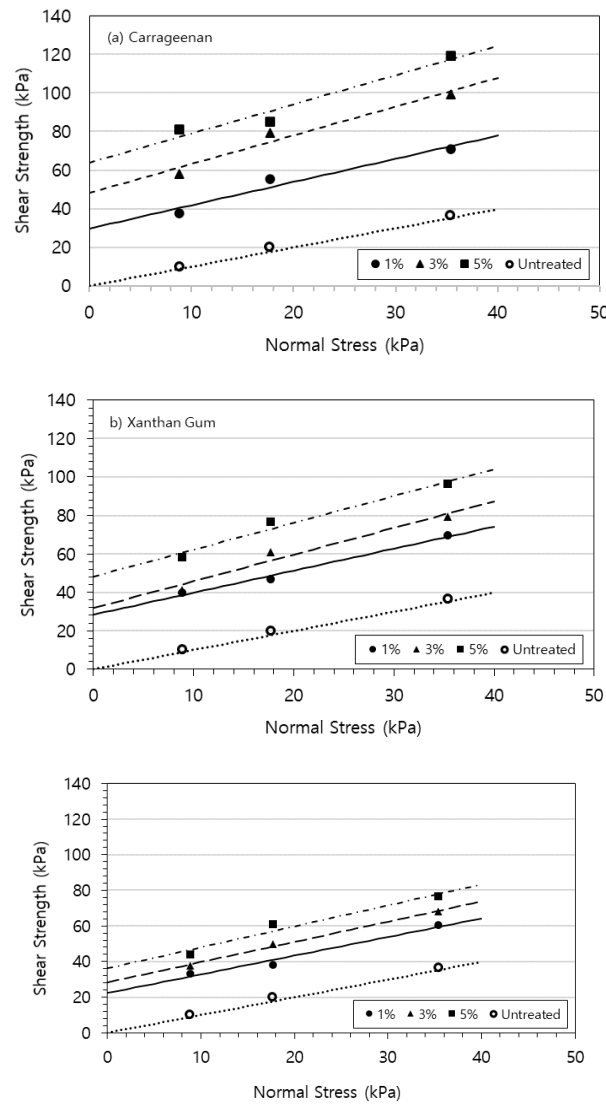


Fig. 5 Results for shear strength of biopolymer-treated soil: (a) carrageenan, (b) xanthan gum, and (c) guar gum

friction angle increase because the treatment mainly effective on inter-particle cohesion and improve the partial contact (Lee *et al.* 2017).

The compressive stress-axial strain curves of each biopolymer-treated soil (carrageenan as an alternative, xanthan gum, and guar gum for comparison) with different biopolymer contents (i.e., 1%, 3%, and 5%) on the seventh day of curing are shown in Fig. 7. The early behaviour does not show a nearly linear trend. Still, the elastic region boundaries are well defined in the carrageenan and xanthan gum-treated soils. The carrageenan and xanthan gum-treated sand initially showed drastic change in axial stress may occur due to the nonuniformity of the sample caused by the curing process. If the sample has a shallow depth, such as the DS sample ($h=2$ cm), the sample's S value decreases, or it dries out (the S ranges between 25%-27%). In contrast, the degree of saturation is not consistent across the sample for the UCS test, which has a higher depth ($h=10$ cm). The condition of the sample 2 cm-3 cm from the surface is fairly dry (the S content is only about 25%-27%), however in the

3 cm-10 cm section from the surface, the saturation level of the sample is approximately 84%-86%. This indicates that the initial deformation happened in the more rigid portion of the sample, whereas the slope of the graph indicates that further deformation happens in the more elastic portion. Therefore, the uniformity of the sample needs to be a concern in further research because uniformity will determine the actual strength.

The values for E_{50} and the ultimate strength (UCS) are shown in Table 2. The difference between E_{50} and the UCS is substantial; moreover, it appears that there are two peaks or that the material is ductile. It is understood that the additional material used was a biopolymer, which is essentially a gel. Therefore, after it passes the elastic limit, the break process does not occur directly. Rather, the length of the sample decreases and the compaction process occurs, which results in a very high ultimate strength (Alshami *et al.* 2022).

The E_{50} values for the carrageenan- and xanthan gum-treated soil increase with the increase in biopolymer

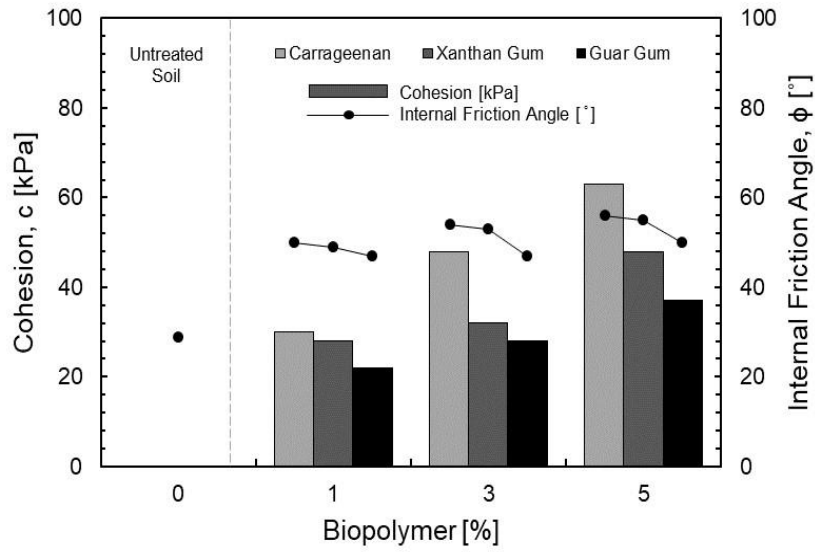


Fig. 6 Shear strength properties of biopolymer-treated soil

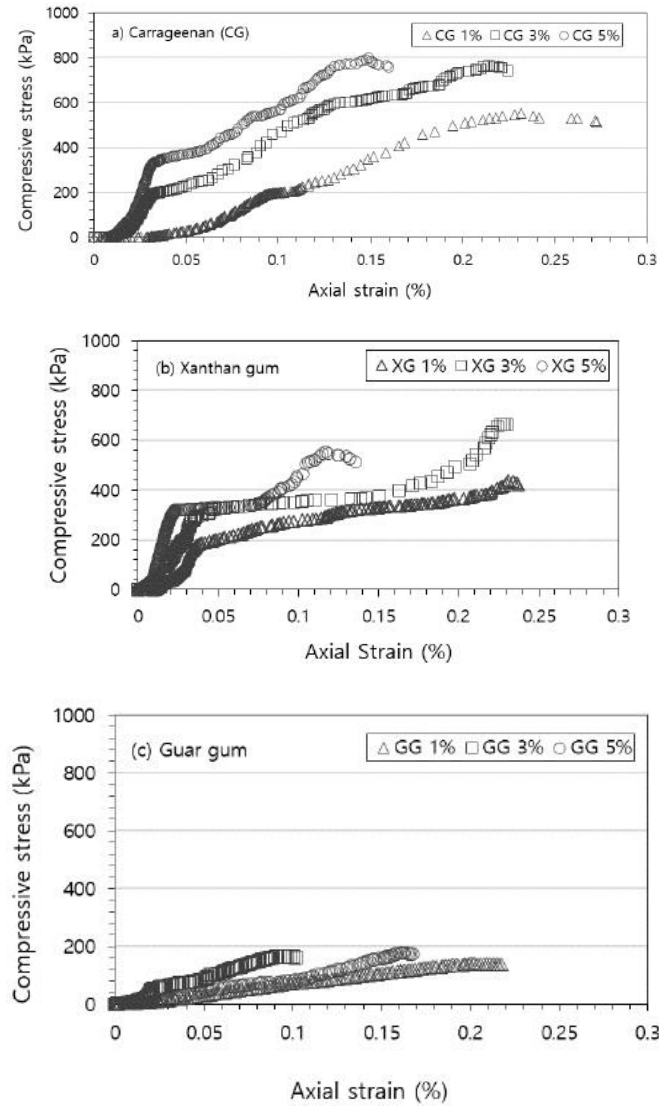


Fig. 7 Results for relation between compressive stress and axial strain: (a) carrageenan, (b) xanthan gum, and (c) guar gum

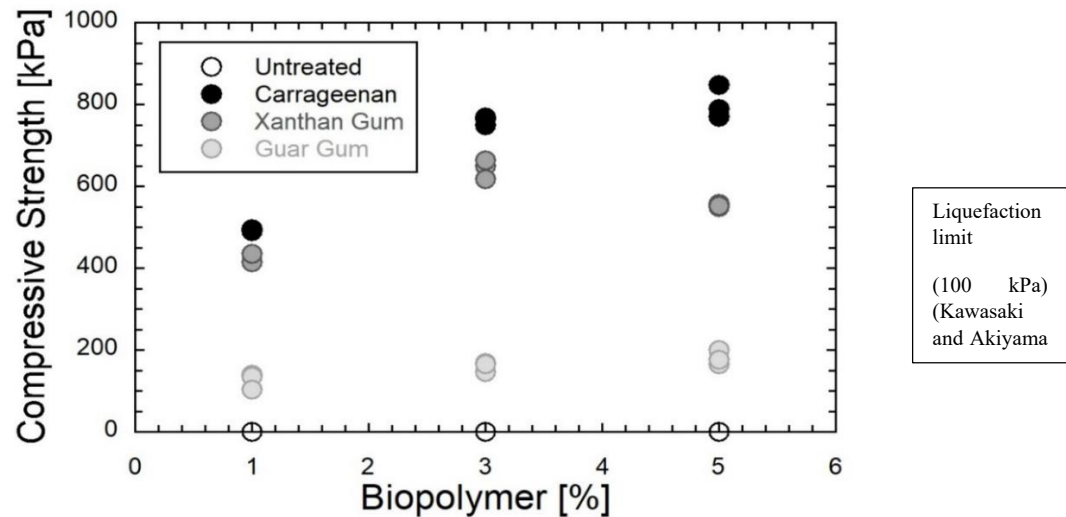


Fig. 8 Unconfined compressive strength (UCS) test result

Table 2 Evaluated E_{50} and UCS from unconfined compression experiments

Sample case	Sample	E_{50} [MPa]	UCS [kPa]
Carrageenan	CG 1%	2.8	551
	CG 3%	7.7	763
	CG 5%	13.6	799
Xanthan gum	XG 1%	5.1	440
	XG 3%	9.8	664
	XG 5%	15.1	553
Guar gum	GG 1%	0.4	140
	GG 3%	1.6	165
	GG 5%	0.9	177

contents. However, the E_{50} values for the xanthan gum-treated soil are higher for all concentrations than the values of carrageenan-treated soil. This demonstrates that the xanthan gum-treated soil has a greater stiffness characteristic, while the carrageenan-treated soil is more flexible (Leckie and Bello 2009). Xanthan gum-treated sample has a greater stiffness characteristic because its viscosity value is greater than that of carrageenan; this is also demonstrated by the results of the viscosity test (Fig. 9), where the viscosity value of xanthan gum at normal temperature is 30% greater than that of carrageenan. However, the UCS values for the carrageenan-treated soil are higher than those for the xanthan gum-treated soil. This is due to the possibility that the fracture process happens first in xanthan gum, which has a greater stiffness than carrageenan, which is more flexible, causing the compaction process, mentioned previously, to be more pronounced in this sample. To further reinforce this analysis, however, more tests, such as consolidation tests, will have to be performed.

The relation between the compressive strength and the concentration of biopolymers is depicted in Fig. 8. The figure shows that all the treated samples exceed the liquefaction limit based on Kawasaki and Akiyama (2013).

The carrageenan-treated soil has the highest compressive strength compared with the others. The compressive strength values of the soils treated with carrageenan tend to increase as the carrageenan concentration rises. In this liquefiable soil, a higher carrageenan concentration is predicted to result in a larger and denser carrageenan matrix. On the other hand, xanthan gum makes a difference. The compressive strength of xanthan gum-treated soil decreased with the 5% concentration of xanthan gum, whereas it increased with the 1% and 3% concentrations.

The 5% samples were less workable because the viscosity of the soil-xanthan gum-water mixture significantly increased (Chang *et al.* 2015). This means that the 5% of xanthan gum has reached the viscosity limit of the treatment, and that the most effective amount of xanthan gum appears to be approximately less than 3%. As can be seen in the results of the viscosity test in Fig. 9, the viscosity value of xanthan gum is greater than the others, so that xanthan gum is more likely to approach the viscosity limit first. Thus far, the concentration of 5% carrageenan is still within an acceptable viscosity range. However, additional research must be conducted on carrageenan's viscosity limit, which can make it less effective as a treatment.

Subsequently, the E_{50} and compressive strength values of the guar gum-treated soil could be better. It is expected that this circumstance is due to the temperature. Guar gum may be even more effective in a high-temperature environment. This is shown in the results of the viscosity test in Fig. 9 that temperature considerably affects the viscosity value of guar gum. The same thing was also shown from several previous studies regarding using guar gum in soil reinforcement. Previous studies (Ayeldeen *et al.* 2017, Sujatha and Saisree 2019) used thermal treatment by making soil samples under oven-dried conditions at 100°C to 105°C before treatment.

Fig. 9 shows the viscosity test result of carrageenan (CG), xanthan gum (XG) and guar gum (GG). The graph indicates that the viscosity value of xanthan gum and guar gum at normal temperature is greater than the viscosity

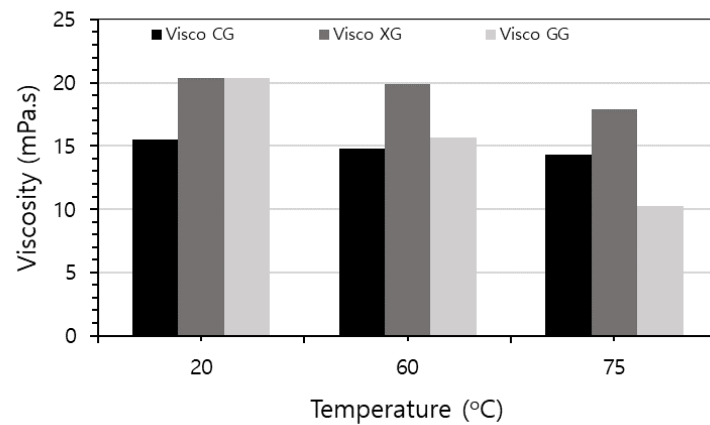


Fig. 9 Viscosity test result of biopolymer

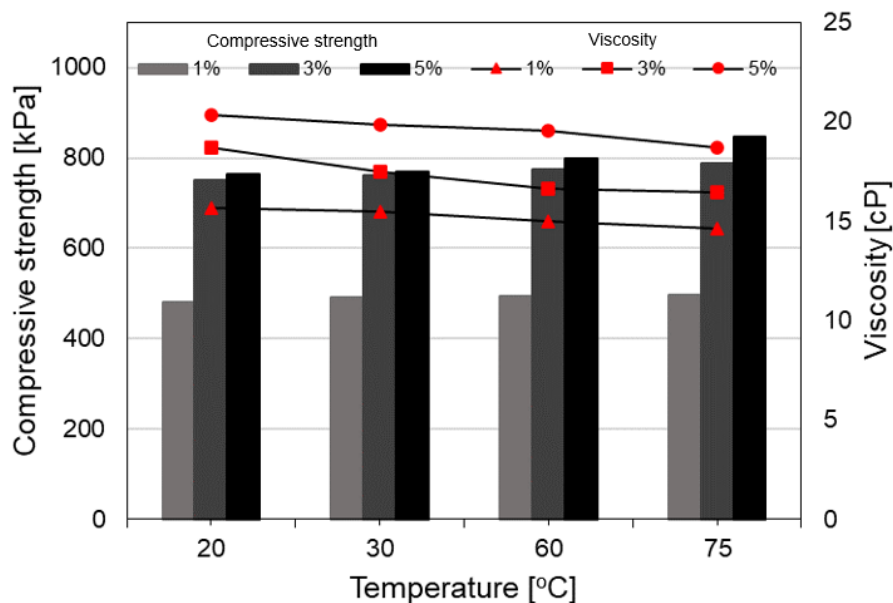


Fig. 10 Results for observing the effect of temperature and viscosity

value of carrageenan. It also shows that temperature can affect viscosity, the higher the temperature, the lower the viscosity value, which means the solution is getting closer to the viscosity of water. The effect of temperature was not significant for carrageenan and xanthan gum because the difference in viscosity between temperatures was less than 5%. This means that carrageenan is a biopolymer that has a stable viscosity value at a wide range of temperature. Temperature simply impacts the ease of the carrageenan and xanthan gum to dissolve in water. While in guar gum, temperature greatly affects the viscosity value. At a temperature of 20°C, the viscosity value reaches 20cP and continues to decrease by about 25% at 60°C and 75°C, respectively.

In terms of observing the effect of temperature on the carrageenan-treated soil in this study, several samples with thermal treatment were prepared. The dry liquefiable soil was firstly heated in an oven at several temperatures (i.e.,

20°C to 75°C) before the mixing process. The molding and mixing processes were the same as in the dry mixing method, but a laboratory hot plate was used for mixing in order to minimize the reduction in temperature. On the other hand, the viscosity test under a range of temperatures was also conducted to evaluate the effect of heat on the viscosity of carrageenan. The compressive strength (column) and viscosity (line) results for the range in temperature are given in Fig. 10. The results show that the compressive strength values increase with the increase in concentration and temperature. However, the difference in compressive strength between each concentration in the range of temperatures is less than 5%, which is a quite small. Moreover, carrageenan also shows small differences in the viscosity values, but an increase in concentration will also increase the viscosity. It is concluded that the compressive strength and viscosity values of carrageenan are relatively stable over a wide range of temperatures;

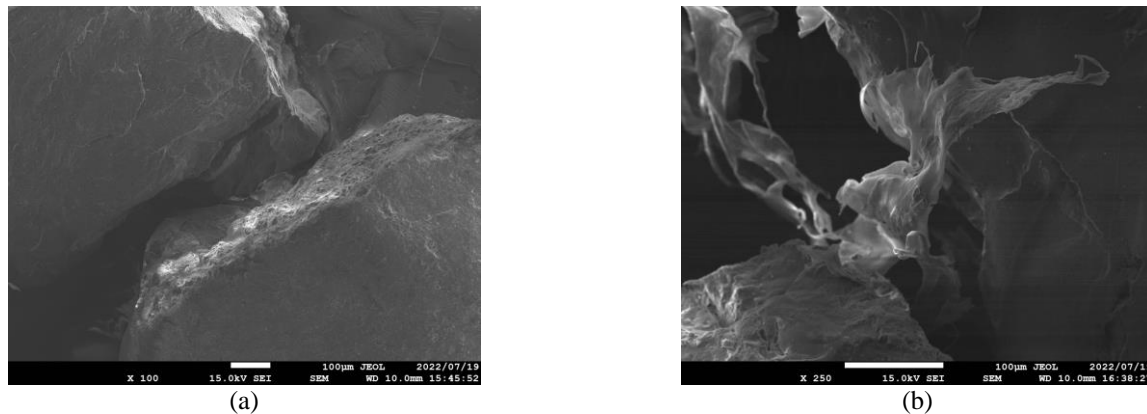


Fig. 11 SEM images of (a) untreated soil and (b) carrageenan-treated soil

these features are similar to those of the xanthan gum biopolymer based on Chang *et al.* (2015). Temperature simply impacts the ease of the biopolymer to dissolve in water.

3.3 Microscopic behavior

The SEM images of the untreated soil and carrageenan-treated soil are shown in Fig. 11. The untreated soil (Fig. 11(a)) shows pure sandy soil with no bonds between the sand particles. Fig. 11(b) demonstrates that the carrageenan treatment for liquefiable soil covered the grain surfaces and enhanced the contact area among the soil particles, while also building connection bridges between particles that are not directly in contact. This can happen because the dry mixing method, which firstly mixes the soil with the carrageenan, allows the carrageenan grains to fill in the pores between the soil particles before the reaction occurs. This is because biopolymers do not interact with the soil itself (Chang *et al.* 2015). The reaction will occur when water is added; it accumulates and aligns as yarn and textiles. This demonstrates how strongly the strength of the carrageenan fiber matrix (in the form of threads or fabrics), present in the pore spaces, affects the strength values of soil that has been treated with carrageenan.

This is also consistent with study conducted by Lee *et al.* (2022), which indicates that the formation of intergranular matrices between sand grains significantly contributed to the overall rise in q_u and E_{50} of biopolymer-treated soil. In cohesionless soil, the dried biopolymer-treated soil displayed equivalent strength to cement-treated soil. In addition, a higher biopolymer concentration was coupled by a more widespread and thicker biopolymer matrix after dehydration, resulting in greater q_u and E_{50} values. Even though biopolymer treatment of sand results in a higher q_u than cement treatment, the biopolymer-treated sand exhibits more flexibility and less rigidity.

4. Conclusions

The study of carrageenan as an alternative biopolymer for soil strengthening, and xanthan gum and guar gum for comparison, has been conducted through several

experimental tests. The results indicate that carrageenan is a tremendous potential alternative biopolymer for improving the strength of liquefiable soil. It can increase the cohesion value of liquefiable soil by 22% to 60% compared to untreated soil. It causes an increase of 60% to 92% and a change from very loose sandy soil (i.e., $\phi=29^\circ$) to very dense sandy soil.

Carrageenan-treated soil seems more flexible because it has fairly small E_{50} value, although due to the compaction process, the ultimate strength value is higher. Carrageenan was also shown to have a significant effect on the compressive strength and to exceed the liquefaction limit.

The compressive strength values of soils treated with carrageenan tend to increase as the carrageenan concentration rises. Carrageenan treatment for liquefied soil covers the grain surface and increases the contact area between soil particles, while building bridges between particles that are not in direct contact. The strength of the carrageenan fiber matrix (in the form of yarn or cloth) in the pore spaces affects the strength of the soil that has been treated with carrageenan.

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