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1

ABSTRACT

Biological approaches have been explored as environmentally friendly alternatives to engineered soil methods in geotechnical engineering practices, recently. The use of microbial induced calcite precipitation, reactive enzymes, and microbial polymers, such as biopolymers, in soil improvement has been studied by researchers around the world. In the present study, gellan gum, a microbial polysaccharide generally used in the food industry due to its hydrogel rheology, was used to strengthen sand. The effects of gellan gum on the geotechnical behaviors of cohesionless sand were evaluated through a series of experimental programs including an unconfined compression test, direct shear test, falling head permeability test, and scanning electron microscopy. The geotechnical properties (friction angle, cohesion, and unconfined compressive strength) of gellan gum-treated sands were determined based on varying moisture conditions: initial, dried, and re-submerged. Gellan gum has a distinct strengthening effect on cohesionless sands through artificial cohesion which varies with the moisture conditions. The strengthening effect of gellan gum on sand appears to be a result of the combination of enhanced bonding between unreactive sand particles and the agglomeration of sand particles through hydrogel condensation, in which the agglomerated sand particles behave as enlarged aggregates in soil.

Keywords: Gellan gum biopolymer, Unconfined compressive strength, Friction angle, Inter-particle cohesion, Hydraulic conductivity

1 **INTRODUCTION**

2   Biological approaches for soil improvement recently have been studied in an effort to address

3 environmental concerns related to improving existing soil materials (*e.g.,* an increased shear strength).

4 A number of studies have investigated the possibility of directly implementing microbial induced

5 calcite precipitation (MICP) using microbes in the soil with the goal of strengthening the soil using a

6 lower carbon footprint than that of ordinary engineering soil practices such as cement mixing (Cheng

7 et al. 2013; DeJong et al. 2006; DeJong et al. 2010; van Paassen et al. 2010; Whiffin et al. 2007).

8 Although MICP is an innovative and environmentally friendly alternative soil treatment method,

9 several drawbacks of this approach have been noted including difficulties in obtaining a uniformly

10 treated soil layer due to the uncertainty of the microbial activity and the secretion quality of the soil,

11 which restricts its application to loose and collapsible sands (Soon et al. 2013). Moreover, microbial

12 ureolysis in the ground releases 2 moles of ammonium ions, NH_4^+, for every 1 mole of calcite $CaCO_3$

13 precipitation, which increases the soil pH, forms toxic salts, and emits toxic gas into the atmosphere

14 (Akiyama and Kawasaki 2012; Ferris et al. 2004).

15   Meanwhile, several attempts to use biological organic matter, such as biopolymers, have been

16 reported to stabilize soil aggregates directly instead of activating microbial responses in the soil,

17 (Chang and Cho 2012, 2014; Chang et al. 2015d; Ferruzzi et al. 2000; Orts et al. 2007; Ringelberg et

18 al. 2014). For example, direct biopolymer mixing of a high-molecular chain structure glucan-type

19 biopolymer, β-1,3/1,6-glucan, results in sufficient workability due to the pseudoplastic rheology of the

20 biopolymer. This enables the formation of a uniform biopolymer-soil mixture providing high

21 strengthening with a low carbon dioxide footprint (Chang and Cho 2012). Specifically, the unconfined

22 compression strength provided by 0.25% β-1,3/1,6-glucan mixing (2.17 MPa) is similar to that

23 obtained with 10% cement mixing (2.65 MPa) showing that biopolymers are capable of achieving

24 similar strengths at significantly lower concentrations (1/10 or lower). Thus, biopolymer soil

25 treatment for geotechnical purposes has the potential to reduce the net amount of mixing materials,

26 particularly materials such as cement that involve a high amount of carbon dioxide emissions during

27 production (8 % of all global emissions) (Chang et al. 2015b; International Cement Review 2015).

28   In addition, recent studies have shown the possibility of using gel-type biopolymers, such as agar

29   gum, xanthan gum, and gellan gum, to enhance the strengthening efficiency in geotechnical soil

30   treatment (Chang et al. 2015a; Chang et al. 2015c; Khatami and O'Kelly 2012). Specifically, thermo-

31   gelation biopolymers such as agar gum and gellan gum provide significant strengthening. Thermo-

32   gelating biopolymers initially form a hydrocolloid phase when dissolved in water with a temperature

33   above 90 °C and then, transform to a hydrogel phase with significantly increased viscosity when

34   cooled to below 40 °C. The firm hydrogel of the thermo-gelation biopolymers forms matrices with

35   mediating particles and enhances the overall strength of the whole network (Huang et al. 2007;

36   McHugh 2003).

37   The use of gellan and agar biopolymers involves different strengthening mechanisms depending on

38   the soil type. Thermo-gelation biopolymers show stronger adhesion with clayey particles, making it

39   possible to achieve an unconfined compressive strength of 13 MPa for 3%, relative to the soil mass,

40   gellan gum biopolymer mixed kaolin under a dried condition (Chang et al. 2015c).

41   However, most previous studies have only provided preliminary work introducing biopolymers as a

42   possible new binder material for civil and geotechnical engineering purposes. Although gellan gum

43   forms stronger biopolymer-soil matrices with clayey particles than it does with cohesionless coarse

44   particles, in practical applications, sandy soils are generally preferred over clayey soils due to their

45   superior workability and drainage control. Moreover, sandy soils have a higher strengthening

46   efficiency than that of clayey soil based on the unconfined compressive strength (UCS) of untreated

47   natural soils   (Chang et al. 2015a), and many aspects of geotechnical engineering require

48   cohesionless soils with an improved shear strength behavior. For these reasons, sand was used as the

49   target soil for this study.

50   Using a series of experimental approaches, this study examined in detail the effect of gellan gum

51   biopolymer treatment on cohesionless sand focusing on geotechnical engineering design parameters,

52   specifically, the friction angle, cohesion, and hydraulic conductivity. Furthermore, possible

53   applictions of gellan gum biopolymers for geotechnical engineering purposes are recommended based

54   on the findings of these experimental studies and analyses.

55

56 **MATERIALS AND METHODS**

57 *Materials*

58 *Jumunjin sand*

59   Jumunjin sand is a typical standard sand in Korea that has been widely used in various studies

60 (Chang et al. 2015c; Min and Huy 2010; Park et al. 2008). Using the USCS classification, Jumunjin

61 sand is classified as a poorly graded sand (*SP*). Fig. 1 shows the particle size distribution of Jumunjin

62 sand. The coefficient of uniformity (C_u) and the coefficient of gradation (C_c) are 1.94 and 1.09,

63 respectively. It has particulate structures between a minimum void ratio of 0.64 and a maximum void

64 ratio of 0.89; its specific gravity (G_s) is 2.65.

65

66 *Biopolymer: Gellan Gum*

67   Gellan gum is a high molecular weight polysaccharide that is fermented from *Sphingomonas elodea*

68 microbes. Low acyl gellan gum biopolymer supplied by Sigma Aldrich (CAS No: 71010-52-1) was

69 used in this study. Low acyl gellan gum partially hydrates in cold water and fully dissolves at

70 temperatures above 90 °C forming a hydrocolloid phase solution. The viscosity of a gellan gum

71 solution increases with decreasing temperature, and finally, a highly viscous hydrogel is formed below

72 40°C (*i.e.*, thermo-gelation). Thus, hydrogel formation is expected to occur naturally during the

73 mixing process with soil through decreasing temperature.

74

75 *Sample preparation*

76   All gellan gum-sand samples were prepared by following the same sample preparation method.

77 First, a gellan solution was prepared by dissolving pure gellan gum into distilled water at 100°C

78 according to the target concentration (0.5, 1.0, 1.5, 2.0, and 5.0% relative to the mass of the soil) of

79 the gellan gum-sand mixtures. The initial water content for mixing was set at 30% of the dry sand

80 weight because it was found that an initial water content of 30 %is is sufficient to fully fill the inter-

81 granular voids with biopolymer gels, which leads to a fully saturated condition for gellan gum-sand

- 5 -

82   mixtures during sample preparation. Thus, 1.67%, 3.33%, 5.0%, and 6.67% gellan gum solutions

83   were prepared to finally deliver 0.5%, 1.0%, 1.5%, 2.0%, and 5.0% gellan gum-sand mixtures (by

84   weight), respectively.

85   To prevent immediate cooling and thus ensuring sufficient mixability, dry sand was heated in an

86   oven to 100 °C before mixing. The heated gellan gum solution and sand were mixed on a laboratory

87   hot plate to minimize temperature reduction during mixing and to form a uniform gellan gum-sand

88   mixture. After mixing, the hot gellan gum-sand mixtures were immediately placed into molds relevant

89   for tests before cooling. Cube samples (50 mm × 50 mm × 50 mm) were prepared for the unconfined

90   compression tests, while disk samples (diameter = 60 mm, height = 20 mm) were molded for use in

91   the direct shear tests.

92   The strength of biopolymer treated soils strongly depends on the water content of the biopolymer-

93   soil mixture (Chang et al. 2015c). Thus, three different moisture conditions were considered in the

94   experimental studies: 1) the initial condition, in which samples were tested immediately after cooling;

95   2) a dry condition, in which samples were fully dried for 28 days at room temperature (20°C) with a

96   final water content less than 1 %, and 3) a re-submerged condition, in which the dried samples were

97   submerged in water for 24 hours before testing. Due to the hydrophilic characteristic of the gellan

98   gum, dried gellan gum gels immediately adsorb water and re-hydrate within 2 hours while higher

99   concentrations require up to 6 hours. Full saturation of all the samples was obtained in less than 6

100   hours. However, the gellan gum-treated sands were submerged for 24 hours to provide sufficient time

101   for re-hydration and stabilization, in which the gellan gum gels inside the soil recover to the initial

102   concentration before drying. The water content (w / m_s) and biopolymer concentrations (biopolymer to

103   water ratios in mass; m_b / w) of the gellan gum-treated sands at different moisture conditions are

104   summarized in Table 1.

105   For the re-submerged condition, we think that the adsorbed water interacts with the hydrophilic

106   gellan gum biopolymers rather than with the electrically neutral sand particles. However, this

107   hypothesis is inappropriate for soils that contain clay particles which have hydrophilic double-layer

108   surfaces.

109

Unconfined compression tests

111   Cubic samples (50 mm × 50 mm × 50 mm) of the gellan gum treated sands were prepared at gellan

112   gum concentrations of 0.5%, 1.0%, and 2.0%. Unconfined uniaxial compressive testing was

113   performed with a Universal Testing Machine (UTM). The axial strain rate was controlled at a rate of

114   0.5 mm/min (1% strain/min). The maximum strength and the stress-strain behaviors were obtained by

115   averaging three different measurements for a single condition.

116

Direct shear tests

118   Disk shaped gellan gum-sand samples with a 50 mm diameter and 20 mm height were placed into a

119   direct shear apparatus with porous stones placed above and beneath them and confined at 50, 100,

120   200, and 400 kPa with a pneumatic actuator for 12 hours before applying horizontal shear. Direct

121   shear tests were performed for the three different moisture conditions mentioned in Section 2.2. The

122   samples for the initial and the re-submerged conditions were tested under a saturated condition by

123   filling the inside of the shear box with water before applying vertical confinement. Horizontal shear

124   with a shear rate of 2% strain/min was applied under a consolidated-drained condition following the

125   ASTM D 3080 standard (ASTM D3080 / D3080M-11 2011) with confining pressures of 50, 100, 200,

126   and 400 kPa. Horizontal shear was applied for 500 seconds to finally induce a 10 mm horizontal

127   displacement. Horizontal load, vertical strain, and horizontal displacements were obtained

128   automatically through load cell and linear variable differential transformer (LVDT) measurements, for

129   which the electronic measurements were attained with a computerized data acquisition system.

130

Hydraulic conductivity tests

132   The initial void ratios of the gellan gum-sand mixtures were within a relatively narrow range

133   between 0.82 and 0.87, regardless of the gellan gum content in the soil mass. Gellan gum-sand

134   mixtures were prepared and poured into a cylindrical cell with an inner diameter of 70 mm and a

135   height of 140 mm (height to diameter ratio = 2:1) before thermo-gelation. The cylindrical cells were

- 7 -

136   sealed with parafilm and left to cool for 7 days to prevent loss of moisture from the gellan gum-sand

137   mixtures and thereby preserving the initial mixing conditions.

138   Permeability tests were performed according to the ASTM D 5084 standard (ASTM D5084-10

139   2010) using a flexible wall permeameter at room temperature ($20\pm1°C$) while the inlet and outlet

140   flows were controlled and maintained using a pressure panel. A head difference of 150 mm was

141   applied to fully saturate the specimens, and a confining pressure of 30 kPa was applied. When the

142   Skempton B-values (Skempton 1954) of the samples exceeded 95%, the samples then were

143   considered as fully saturated. After saturation, falling-head permeability tests were performed with a

144   hydraulic gradient of 20 or less. Only the initial condition was considered for the laboratory

145   permeability tests to simulate the bio-clogging effect (Mitchell and Santamarina 2005) induced by

146   instant gellan gum-treatment (*e.g.*, mixing) for saturated (below ground water table) sand deposits *in*

147   *situ*.

148

149   *Scanning Electron Microscopy (SEM)*

150   SEM images were taken to observe the micro-scale direct interactions between the sand particles

151   and the gellan gum biopolymers. Undisturbed and disturbed 1% gellan gum-treated sand samples

152   were examined by collecting 0.5 cm^3 bulk cubic samples from dried gellan gum-sand mixtures that

153   had not been subjected to any testing, and disturbed samples were prepared by attaching the 1% gellan

154   gum-sand specimens that had been subjected to failure on an SEM mount (diameter 25 mm) using

155   carbon conductive tabs. Carbon paint was applied to the edges and bottoms of both the undisturbed

156   and disturbed sample to provide sufficient grounding. Specimens were coated for 20 seconds using an

157   Osmium Plasma Coater (OPC) with osmium tetroxide (OsO_4) as the source of osmium. An extreme

158   high-resolution scanning electron microscope was used to observe the surface of the gellan gum-sand

159   samples.

160

161   **RESULTS AND ANALYSIS**

162   *Unconfined Compressive Strength (UCS) of the gellan gum-treated sand samples*

163   The values for the unconfined compressive strengths and accompanying water contents of the

164   gellan gum biopolymer treated sands are summarized in Fig. 2 and Table 1. The strength of the gellan

165   gum-sand mixtures increased with increasing biopolymer content regardless of the moisture

166   conditions, while the fully dried condition resulted in the highest values for the unconfined

167   compressive strengths at 130.2, 241.9 and 434.6 kPa for the 0.5%, 1%, and 2% gellan gum,

168   respectively (Fig. 2a). In comparison with the cement treatment, even a small amount of gellan gum

169   treatment increased the dry strength of sand significantly. For instance, the UCS of the 2% gellan

170   gum-treated sand (434.6 kPa) is higher than that of 12% cement-treated sand (380 kPa), even though

171   the gellan gum-treated sand has a lower dry density (1,430 kg/m^3) than that of the cement treated sand

172   (1,770 kg/m^3) (Yoon and Abu-Farsakh 2009). Moreover, the UCS of 2 % gellan gum-treated sand is

173   identical to the UCS of densely compacted (1,930 kg/m^3) 8 % cement-sand mixtures after 7 days of

174   curing (470 kPa) (Cheng et al. 2013).

175   The UCS behavior of the gellan gum-treated sands is consistent with the varying concentrations of

176   gellan gum gels (m_b / w; gellan gum to water ratio, in mass) in the soil (Table 1). Dehydration of the

177   gellan gum gels from the initial to the dried state increases the m_b / w significantly, while a higher

178   gellan gum content (m_b / m_s) results in the retention of a higher final water content (w / m_s) after

179   drying due to the strong hydration characteristics of the gellan gum biopolymer (Nussinovitch 1997).

180   The increased m_b / w with drying is highly correlated to the strengthening efficiency of the gellan gum

181   treatment. Therefore, an increase in strength can be attributed to the densification of the gellan

182   biopolymers, which results in higher m_b / w ratios. Previous studies have been conducted on the

183   strength of gels based on the concentration of the biopolymers to the water mass (Banerjee and

184   Bhattacharya 2011). In an earlier study, it was found that the strength of the gels increased with higher

185   m_b / w ratios. Because the gellan gum biopolymer has strong hydration capabilities (Nussinovitch

186   1997) exceeding that of sand particles, the water content of the samples interacts mostly with the

187   gellan gum. Therefore, a change in the water content of the samples can be attributed to a similar

188   change in the m_b / w ratios.

189   For the re-submerged condition, although the m_b / w values were similar to that of the initial state

190   (≈30%), the UCS values of the re-submerged samples were lower than those of the initial condition

191   samples (17-23% lower). Therefore, not only the gellan gum gel concentration but also the hydro-

192   rheology (*e.g.,* swelling and disturbance) is an important factor regarding the strength behavior of

193   gellan gum-treated sands. This indicates that even though re-submergence recovers the specimen

194   water content and gellan gum concentration to levels similar to the initial state, subsequent re-wetting

195   after the primary dehydration process does not recover the strength of the gellan gum hydrogels up to

196   the initial condition. However, the strength of the gellan gum-treated sand without any variations in

197   the water content are known to remain stable regardless of the time (Chang et al. 2015c). Thus, it can

198   be concluded that the initial state and strength of gellan gum hydrogels is not recoverable once the

199   gellan gum gel is condensed via dehydration. Further studies are recommended to characterize the

200   strength and micro-rheology variations of the gellan gum-treated sands under subsequent repeated

201   drying and wetting cycles with additional analytical methods such as infrared or EDX (Energy

202   Dispersive X-ray) spectroscopy.

203   The stress-strain relationships of the gellan gum-treated sands (shown in Fig. 3) provide a better

204   explanation, showing that the dried gellan gum-treated sand had higher stiffness compared to the other

205   conditions (initial and re-submerged). This indicates that inter-particle strengthening results from the

206   formation of thick and high-tensile biopolymer dehydrates (*e.g.*, films) among the sand particles,

207   which has also been observed in previous studies (Chang and Cho 2012; Chang et al. 2015a).

208   However, once the primary dried samples are re-submerged, and the water content is recovered

209   through hydrophilic absorption, the strength of the soil greatly diminishes due to the swelling of the

210   biopolymer hydrogels and the weakening of the biopolymer bonds, while the ductility increases to a

211   level higher than that of the initial conditions. This appears to be the result of partial and sequential

212   swelling of the gellan gum hydrogels beginning at the outermost boundary of the dehydrated gellan

213   gum gels, which initially form thick layers on the sand particles during the primary drying process.

214

215   ***Direct shear strength of the gellan gum-treated sand***

216 *Peak shear behavior*

217   The peak strengths of the gellan gum-treated sands at different moisture content levels, obtained by

218 direct shear testing, are shown in Fig. 4. The peak shear strength (τ_{peak}) values of the gellan gum-sand

219 mixtures are increased with the higher biopolymer contents, regardless of the moisture conditions.

220 With the higher biopolymer contents, increased strengthening of the samples is clearly seen for both

221 the initial and dried conditions (Figs. 4a & b), while the strengthening effect appears to diminish for

222 the re-submerged samples (Fig. 4c).

223   Fig 5 shows the peak strength properties (cohesion and friction angle) of the gellan gum-treated

224 sand samples. Once the gellan gum hydrocolloids are mixed with the sand (initial condition), the

225 cohesion of the highly viscous hydrogels increases immediately while the peak friction angle remains

226 almost constant without any noticeable variation identical to the friction angle of 28º for the untreated

227 wet sand. As the gellan gum gels become denser with drying, both the peak interparticle cohesion

228 (c_{peak}) and the peak friction angle (Φ_{peak}) increase significantly with increasing gellan content.

229 Although the c_{peak} values show a continuous increment up to a 5% gellan gum content regardless of

230 the moisture condition, the Φ_{peak} increment appears to level off and converge after the 2% gellan gum

231 content. Meanwhile, dehydration of excess gellan gum gels above 2% does not affect the structural

232 composition of the sand particles. Instead, the particles exhibit enhanced interparticle cohesion due to

233 the higher strength of the highly concentrated gellan gum hydrogels which coat the particle surfaces,

234 enhance the inter-particle contacts and fill the void spaces.

235   A previous study showed that cement-treated sand, with a 10% cement to sand ratio by volume or

236 8% by mass, remarkably improves the friction angle (as great as 44°), while having a negligible effect

237 on the inter-particle cohesion (at a value of 0.42 kPa) (Krantz 1991). In comparison, the friction angle

238 of an 8% cement treatment sample is equivalent to that of a sample with a 5% gellan gum mixture;

239 however, the increase in the cohesion for the cement treatment sample cannot be compared to the

240 remarkable improvement in cohesion for the gellan gum-treated sands (as great as 166.17 kPa for 5 %

241 gellan gum in a dried state).

242   The strengthening during gellan gum treatment is due to the condensation and aggregation effects

243   of the high tensile gellan gum hydrogels among the sand particles; on the other hand, the

244   strengthening of the cement treated sand is dominated by the formation and pore filling of newly

245   formulated Calcium-Silicate-Hydrate (C-S-H) crystals inside the particle voids.

246   Re-submerged specimens experienced a remarkable reduction in c_{peak} (Fig. 5a) with values even

247   lower than the c_{peak} of the initial conditions, while Φ_{peak} (Fig. 5b) remains higher than that of the initial

248   condition. These results indicate that the re-wetting and swelling of the primary dried gellan gum

249   hydrogels is not reversible. In particular, the decrease in cohesion indicates the possibility that the

250   molecular structures and the bonding of the gellan gum biopolymers have been disturbed.

251   Overall, c_{peak} and Φ_{peak} increase with higher biopolymer contents; however, at around 2-5% of the

252   gellan gum biopolymer, the rate of increase in c_{peak} and Φ_{peak} decreases regardless of the moisture

253   conditions. This indicates that there likely is an optimal biopolymer concentration (≈2%) for the sand

254   treatment when the goal is effective shear strengthening.

255

Residual shear behavior

257   Fig. 6 shows the residual shear strengths of the gellan gum-treated sands under different moisture

258   conditions. The residual strength of the gellan gum-treated sand has a similar behavior regardless of

259   the moisture condition unlike the peak shear strength behavior described above.

260   The samples for the initial, dry, and re-submerged conditions all show an increase in the residual

261   friction angle (Φ_{res}) and residual cohesion (c_{res}) with higher gellan contents, while the c_{res} values of the

262   gellan gum-treated sands are almost equal for all of the gellan gum contents regardless of the soil

263   moisture conditions (Fig. 7a). This indicates that the strengthening effect induced by the gellan gum

264   hydrogels in sand persists at high levels of strain which implies persistent inter-particle interlocking

265   and adhesion induced by the hydrogen bonding and high tensile strength of the gellan gum hydrogels

266   (Lee et al. 2004).

267   However, the Φ_{res} values for both the initial and re-submerged gellan gum-sand mixtures are

268   similar at low gellan gum content*s*, 1.0% and lower, while the values at the initial state are slightly

269   higher than those of the re-submerged state for gellan gum contents higher than 2%. Meanwhile, the

270   dried condition sample shows a distinctly higher Φ_{res} than that of the moist condition sample (Fig.

271   7b). Of note, for the gellan gum content below 1%, the Φ_{peak} is clearly higher than the Φ_{res} , while the

272   difference between the Φ_{peak} and Φ_{res} decreases when the gellan gum content exceeds 1% (Figs. 5b

273   and 7b).

274   In general, the shear strength of coarse soil increases with greater dry density (Bolton 1986;

275   Terzaghi et al. 1996). Fig. 8a shows the relationship between the peak shear strengths and the dry

276   densities of the gellan gum-treated sand, for which the dry density reflects the actual particle

277   composition. As the dry density increases, the peak shear strength generally increases, regardless of

278   the biopolymer content and wet conditions.

279   Meanwhile, with an increase in the biopolymer content relative to the mass of soil, the shear

280   strength increment decreases, shown in Fig. 8b, due to a unit dry density (1 g/cm^3) increment (*i.e.*, the

281   ratio between the *change in shear strength* for a subjective *change in dry density*; $\Delta\tau_{DS}$ / $\Delta\gamma_d$ [MPa·cm^3

282   / g]). This indicates that the soil composition has less of an effect on the strengthening mechanism of

283   the gellan gum-treated sands at higher gellan gum contents, and instead, the majority of the

284   strengthening results from the interparticle bonding provided by the gellan gum hydrogels. The $\Delta\tau_{DS}$ /

285    $\Delta\gamma_d$ ratio shows a dramatic degradation with increased gellan gum content and finally levels off at the

286   2-5% gellan gum content. However, the peak interparticle cohesion (c_{peak}) increases inversely to the

287    $\Delta\tau_{DS}$ / $\Delta\gamma_d$ trend. Thus, the strengthening mechanism of the gellan gum-treated sands appears to be less

288   dependent on the structural composition of the sand particles, whereas it is strongly affected by the

289   rheology and strength of the gellan gum hydrogels existing inside.

290

291   *Shearing mechanism of the gellan-treated sand samples*

292   The condensation and aggregation mechanism of gellan gum-treated sands is schematically shown

293   in Fig. 9. Natural (untreated) sand has no chemical inter-particle bonding between sand particles, only

294   along the shear band (Fig. 9a), resulting in individual particle movement, rotation and overturning

295   (Fig. 9b). Meanwhile, gellan gum hydrogels provide artificial inter-particle cohesion which

- 13 -

296   strengthens with condensation and finally binds particles into agglomerates through increased particle

297   connections and inter-particle bridging. This causes several soil particles to act as a larger single

298   aggregate which requires a larger dilation angle and rotational frustration as well as an increased peak

299   friction angle shown in Figs. 4, 5b and 9c. As such, gellan treated sands have high interparticle

300   cohesion and friction angle values, especially in the dried state compared to those of untreated sand.

301   The values for the residual friction angle (Φ_{res}) of the dried gellan gum-treated sands are higher

302   than those of both moist (*i.e.*, initial and re-submerged states) gellan gum-sand mixtures in all cases

303   (Fig. 7b). This can be attributed to the microscopic structure of the crushed gellan gum-sand mixtures;

304   this microstructure is identical to that of the residual condition sample that was subjected to shearing.

305   Dried and condensed gellan gum gels break into fragments under large strains; these fragments are

306   expected to behave as frictional materials, improving interlocking between sand particles even in the

307   residual state. Although their viscosities are lower than those of the dried gels, the initial and re-

308   submerged gellan gum hydrogels also have higher Φ_{res} values than those of the untreated sand. Moist

309   hydrogels are expected to be partially crushed due to the motions of the sand particles; this can also

310   possibly enhance the friction characteristics of the sand particles at large strain shearing through

311   interface attractive adhesive forces between the sand surfaces and the viscous gels (Gong et al. 1999;

312   Tominaga et al. 2008). Thus, it becomes clear that the existence of gellan gum hydrogels inside soil

313   increases both the peak and residual friction properties through hydrogel condensation.

314

315   ***Scanning Electron Microscopy (SEM)***

316   SEM images of the 1% gellan gum-sand mixtures are shown in Fig. 10. Undisturbed gellan gum-

317   treated sand (Fig. 10a) shows that the gellan gum biopolymers coagulate into gellan gum films which

318   form connection bridges between adjacent particles (Fig. 10b). This results in an increase of both the

319   peak strength and the elastic modulus (Fig. 3) which was also observed in previous biopolymer

320   studies, including research on beta-glucan (Chang and Cho 2012) and xanthan gum (Chang et al.

321   2016; Chang et al. 2015a). Therefore, the gellan gum-sand matrix formation and hardening

- 14 -

322   mechanism through dehydration is expected to be governed by the densification (m_b / w increase;

323   Table 1) of the gellan gum hydrogels inside the sand pores.

324

Effect of water on the behavior of the gellan gum-treated sands

326   One drawback to the strengthening mechanism of gellan gum-treated sand is the decrease in

327   strength when the primary dried gellan gum-sand mixtures are re-submerged in water. With

328   dehydration, the condensed gellan gum hydrogels force sand particles to interact more directly and

329   tightly because the gellan biopolymers form increased particle interactions (Fig. 10a) and fibrous

330   clumps (Fig. 10c) which enhance the strength of the gellan gum-treated sand samples. When the dried

331   gellan gum-treated sands are subjected to water again, the hydrophilic gellan gum-water interaction

332   and accompanying swelling (Fig. 2b) appear to degrade the strength properties such as the strength

333   and stiffness (Fig. 3) as well as the inter-particle cohesion (Fig. 5a) of the gellan gum gels inside soil,

334   regardless of the gellan gum to sand ratio. This is consistent with the phenomenon of an

335   unrecoverable initial state of gellan gum hydrogels once they are condensed through dehydration

336   outlined in section 3.1. Thus, it can be concluded that the initial state and strength of the gellan gum

337   hydrogels is not recoverable once the gellan gum gel is condensed through dehydration.

338   The hypothesis that gellan gum-treated sand with varying water contents has different geotechnical

339   engineering behaviors is shown in Fig. 11. In the initial state, the gellan gum hydrocolloids uniformly

340   disperse between sand particles, and the viscosity increases by thermo-gelation (cooling) (Fig. 11a).

341   As the gellan gum hydrogels undergo dehydration, the thickened gels begin to coagulate around the

342   sand particles, while the decrease in volumetric moisture content leads to the formation of discrete air

343   voids along the gellan gum gels (Fig. 11b). Once the gellan gum gels are dried, condensed film-like

344   gellan gum gels enhance the inter-particle interaction (Figs. 3 and 5a) through biopolymer matrix

345   formation (Figs. 10a & b) among the sand particles (Fig. 11c). However, once the dried gellan gum-

346   treated sand is re-submerged, the dried gels are expected to absorb water due to their hydrophilicity.

347   Gradual swelling of the gellan gum gels from the outside rim results in weaker viscosity (or stiffness)

348   of the re-hydrated gellan gum gel relative to that of the initial / uniform hydrocolloid state (Fig. 11a).

- 15 -

349   The strength difference of the gellan gum gels between the initial and re-submerged conditions (Figs.

350   2 and 5) implies that a uniform initial gel matrix cannot be recovered by the re-wetting and

351   accompanying swelling of the dehydrated biopolymer gels without post thermal treatment.

352

353   ***Hydraulic conductivity of the gellan gum-treated sand samples***

354   The hydraulic conductivity of the gellan gum-treated sand in the initial state decreases

355   exponentially, from an untreated condition ($\sim 2.1 \times 10^{-4}$ cm/s) to 1% gellan gum content ($\sim 2.6 \times 10^{-8}$

356   cm/s) shown in Fig. 12. Then, for higher gellan gum contents, the hydraulic conductivity converges to

357   a very low value ($\sim 8.2 \times 10^{-9}$ cm/s) which correlates with an impermeable layer.

358   The convergence of gellan gum-treated sands above a 1% gellan gum content toward a stabilized

359   hydraulic conductivity behavior appears to be induced by a combination of the water holding capacity

360   and the pore-filling characteristic of the gellan gum hydrogels under a saturated condition. Within a

361   fixed pore size, the water content of a fully saturated specimen remains constant; as such, an increase

362   in the gellan gum content will decrease the permeability by reducing the number of easily moveable

363   water molecules inside the specimen. Moreover, thick gellan gum hydrogels are expected to fill the

364   pore spaces of the sands by creating multiple film-type gellan gum layers on the sand particle

365   surfaces, consequently reducing the pore spaces (bio-clogging). Thus, it can be concluded that gellan

366   gum shows promise in terms of controlling the hydraulic conductivity, a feature that can be applied to

367   various environmentally friendly water control purposes in geotechnical engineering.

368   The bio-clogging efficiency increased with greater gellan gum content; once the gellan gum

369   hydrogel reached a concentration for which the water molecules inside the pores completely

370   interacted with the biopolymer molecules, the hydraulic conductivity stabilized to a lower bound.

371   From this point, the excess amount of gellan gum no longer had a large effect on decreasing the

372   hydraulic conductivity. However, for higher hydraulic pressure gradient conditions ($i > 20$), it is

373   expected that a greater amount of gellan gum will be required to produce a similar hydraulic

374   conductivity control effect. It is hence recommended that further studies be performed to investigate

375   the hydraulic conductivity behavior of gellan gum treated soils under large hydraulic pressure

376   conditions and to evaluate the potential of gellan gum biopolymer use in deep grouting or mixing

377   purposes.

378

379   **DISCUSSION**

380   *Economic feasibility of gellan gum use for geotechnical engineering purposes*

381   Currently, the economic feasibility of using gellan gum to replace ordinary cement in geotechnical

382   engineering applications appears to be rather low. Specifically, the current material price of gellan

383   gum is roughly 20,000 USD/ton[1], while the price of ordinary cement is only 100 USD/ton[2] (Kelly

384   and Matos 2014; Morris et al. 2012).

385   Taking into account that lower concentrations of gellan gum are required compared with cement,

386   for instance, 1% gellan gum treatment has an equivalent strength to a 10% cement treatment, and the

387   material price to treat 1 ton of soil amounts to approximately 200 USD for gellan gum and 10 USD

388   for cement. This large difference in price makes the current use of gellan gum in soil treatment

389   impractical. However, the current market price is for food grade gellan gum, and the market for lower

390   grade gellan gum has not yet been developed.

391   If the market for gellan gum were to develop further, allowing for greater commercialization of

392   gellan gum, the prices could decrease significantly. This is what transpired with the

393   commercialization of xanthan gum, with the price dropping from 30,000 USD/ton to 3,000 USD/ton

394   within 20 years (Imeson 2010). With proper marketing and commercialization, it can be assumed that

395   the price of gellan gum could reach the current price of xanthan gum (3 USD/kg). Additionally, high

396   purity food grade gellan gum is unnecessary for construction purposes, and with a lower required

397   purity, production costs could be greatly reduced. Without the cost of additional purification, the total

398   cost could be significantly lower than the anticipated cost of 3 USD/kg, reaching 1.5 - 2 USD/kg. At

399   this point, the cost of treating 1 ton of soil with 1% gellan gum would be approximately 15 – 20 USD,

400   which would be comparable with that of cement (10 USD).

---

[1] Material trade price of bulk (1 ton) low acyl gellan gum (www.alibaba.com)

[2] Market price of cement in the United States in 2014.

401   For future implementation, a heated gellan gum solution (over 90 ºC) can be injected (*e.g.*,

402 grouting) or mixed (*e.g.*, deep auger mixing) directly with *in-situ* soil, to facilitate quick setting by

403 thermo-gelation due to a temperature decrease during implementation. With the cost for heating the

404 gellan solution for soil treatment, a rough calculation of the required energy was performed assuming

405 an overall 50% efficiency with gas heating equipment (Nakićenović et al. 1998). In detail,

406 approximately 588 MJ is required to treat 1 ton of soil with 300 kg of heated 3.3 % gellan gum

407 solution. Generally, gasoline has an energy density of 46.4 MJ/kg (Nakićenović et al. 1998), and thus,

408 the average fuel cost to treat 1 ton of soil with thermo-gelated gellan gum treatment becomes

409 approximately 9.63 USD/ton, considering the average market price of gasoline in the U.S. in 2014

410 (Kilian 2014). This value is only the cost for the required energy and is highly susceptible to change

411 depending on many variables such as heating mechanisms and methods.

412

413 ***Possible applications of the gellan gum treatment in geotechnical engineering practices***

414   Gellan gum is a biodegradable biopolymer with a high sensitivity to the presence of water.

415 Therefore, any permanent application using gellan gum-treated soils may be difficult. However,

416 because the material is non-toxic and ecofriendly, its use in temporary construction may be extremely

417 beneficial. As the results of the current study show, the use of gellan gum in sand provides a cohesive

418 strength not found in such sands.

419   The increased inter-particle cohesion and the friction angles of the gellan gum-treated sand are

420 expected to provide significant ground improvement in practical geotechnical applications, regardless

421 of exposure to water. To address considerations for foundation engineering, the ultimate bearing

422 capacities ($q_{ult(square)}$) at a 1 m depth of gellan gum-treated sand deposits for square-type (1 m × 1 m)

423 foundations are presented in Table 1. Table 1 is shown in accordance with Terzaghi's formulas on

424 ultimate bearing capacities (Das and Sobhan 2014). Table 1 shows a significant increase in the

425 $q_{ult(square)}$ values with higher gellan gum content. Although both the interparticle cohesion and the

426 friction angle values of gellan gum-treated sand vary with different moisture conditions, the

427 strengthening effects for all moisture conditions improve the $q_{ult(square)}$ of the sands remarkably. For

428   instance, even at the weakest gellan gum treatment condition (0.5%), $q_{ult(square)}$ increased by 266%

429   from that of the untreated sand, 269 kPa, reaching a final value of 983 kPa. The highest $q_{ult(square)}$ was

430   obtained from 5% gellan gum treated sand in a dry condition, reaching an extremely high value of 44

431   MPa. Thus, it can be cautiously concluded that the strengthening efficiency of the gellan gum

432   treatment on sandy soils provides effective and sufficient ground improvement for various

433   geotechnical engineering applications including foundation design.

434   In terms of temporary construction purposes, the proposed treatment may be valuable in

435   applications such as erosion resistance at construction sites, by reducing the amount of airborne dust

436   particles produced by the construction process and the problems that arise with such aeolian dust

437   particles (Pye 1987). It can also provide temporary strengthening of unpaved ground at construction

438   sites, including that used for temporary roads, allowing easier movement and use of heavy machinery.

439   This technology also shows promise as a practical solution for quick permeability control. Unlike

440   cement, because gellan gum does not require a long period of time to set, a quick injection of a gellan

441   gum solution may allow for rapid application of a permeability control, and such quick applications

442   can be helpful in reducing time and material costs (Nimah et al. 1983).

443   The major benefit of biopolymer soil treatment technology is that after the service period of a given

444   temporary support or ground improvement, the biodegradability of biopolymers will allow for natural

445   decomposition while simultaneously providing a carbon food source for local vegetation without any

446   demolition waste.

447

448   **CONCLUSIONS**

449   As an environmentally friendly biopolymer used for soil improvement, the gellan treated sands

450   showed relatively high strengthening even at low concentrations. The unconfined compression test

451   results for the gellan sands (434 kPa) were comparable to those of 12% cement treated sands (380

452   kPa). In terms of shear strength, the gellan sands showed large improvements in the cohesion and the

453   friction angle of the sands. With large improvements in the cohesion and the friction angle at low

454   concentrations, gellan gum can be easily implemented to improve the strength and quality of shallow

455   soils.

456   Additionally, due to the pore filling effects of the gellan hydrogels, the use of gellan has been

457   shown to be capable of decreasing the permeability of sands to 1×10^{-8} cm/s. The decrease in

458   permeability when applying gellan gum into the soil is almost immediate; therefore, when used as a

459   permeability-controlling barrier, gellan gum can provide a fast alternative to sufficiently reduce the

460   permeability of soils.

461   Although gellan gum is highly sensitive to water, the retention of the strength and friction angle

462   even in the re-submerged state indicates a certain degree of durability with drying and wetting cycles.

463   With sufficient durability, the working life of biopolymers will be adequate for numerous construction

464   purposes. In addition, because gellan gum is biodegradable and eco-friendly, its use will allow for

465   natural disposal of the biopolymer into the ecosystem without causing any harm to the environment.

466   Therefore, this method of soil improvement offers many benefits compared to current methods,

467   especially in applications involving temporary improvement. The use of cement may allow for strong

468   and durable engineered ground construction, but due to its overly high durability and difficulties in

469   disposal, its use in temporary applications is highly undesirable. The present results show that gellan

470   gum has various benefits for ground improvement including improvements in strength, friction angle,

471   and reduction of permeability.

472

482

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Table 1. Water content (w / m_s) and biopolymer concentration (biopolymer to water ratios in mass; m_b / w) of gellan gum-treated sands at initial, after drying, and after re-submergence.

Gellan gum content, m_b / m_s [%]	Water content [%]					
	Initial		Dry		Re-submerged	
	w / m_s	m_b / w	w / m_s	m_b / w	w / m_s	m_b / w^*
0.5	30.0	1.67	0.5	100	26.0	1.92
1.0	30.0	3.33	0.7	143	28.0	3.57
1.5	30.0	5.00	0.8	187	28.3	5.30
2.0	30.0	6.67	0.9	222	29.5	6.78

*Maximum possible biopolymer to water ratios for the re-submerged condition

Table 2. Bearing capacities of gellan treated soils

Gellan gum content [%]		0 (untreated)	0.5	1	2	5
c [kPa]	Initial	0.0	24.7	33.2	85.6	103.5
	Dry	0.0	32.6	67.8	114.3	166.2
	Submerged	0.0	18.0	23.3	40.6	44.7
ϕ [°]	Initial	27.9	28.3	29.77	29.7	29.5
	Dry	27.9	34.6	36.85	42.7	47.3
	Submerged	27.9	29.3	30.35	33.4	36.0
$q_{ult(square)}$ [kPa]	Initial	269	1,127	1,618	3,612	4,243
	Dry	269	2,536	5,726	17,340	44,166
	Submerged	269	983	1,304	2,611	3,713
Improvement efficiency [%]	Initial	-	319	502	1,245	1,480
	Dry	-	844	2,032	6,356	16,343
	Submerged	-	266	385	872	1,282
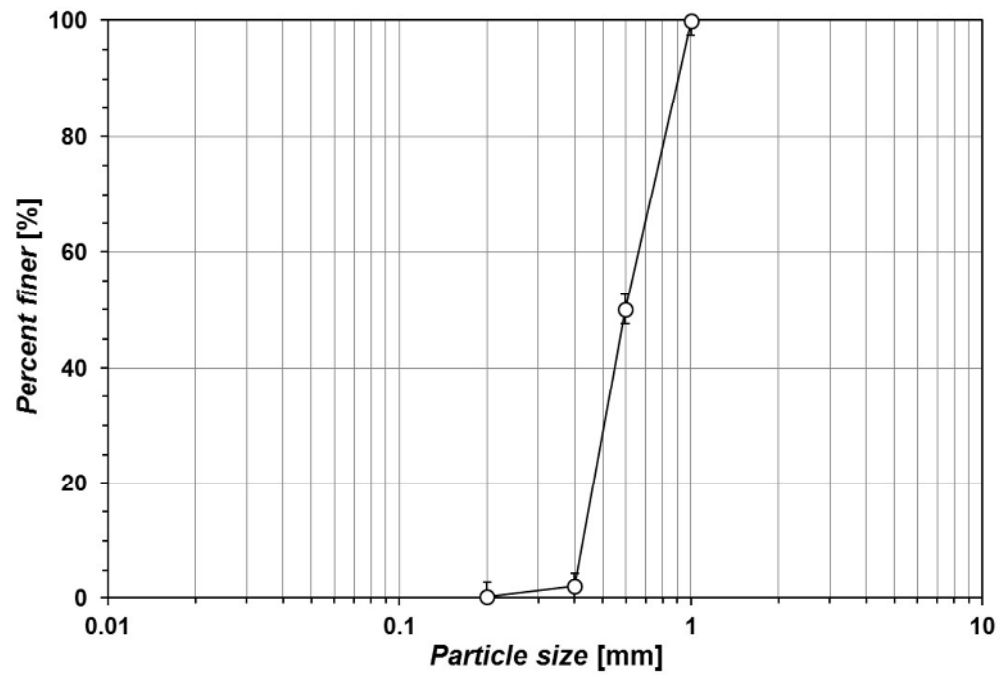
Figure 1. Particle size distribution of Jumunjin sand.
180x120mm (150 x 150 DPI)
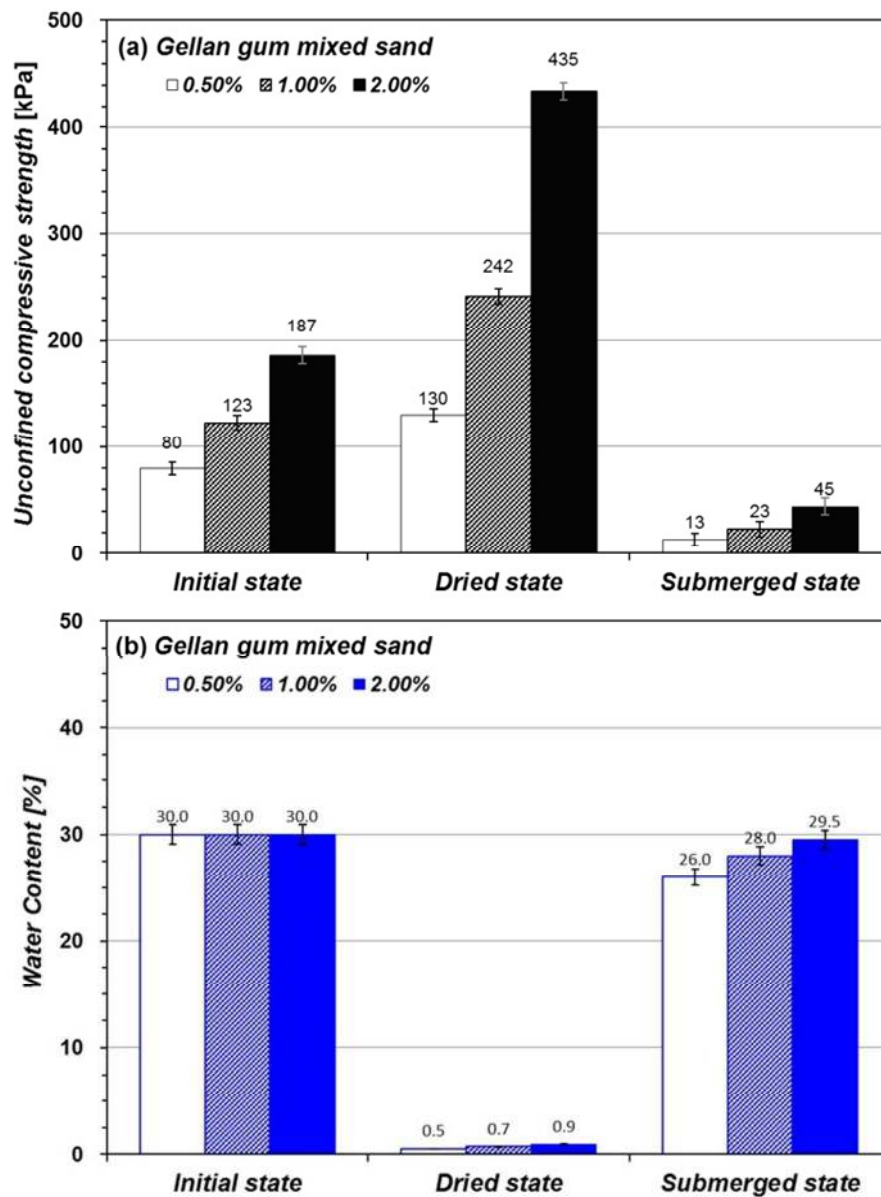
Figure 2. Unconfined compressive strength (a) and water content (b) of gellan gum-treated sand.
126x168mm (150 x 150 DPI)
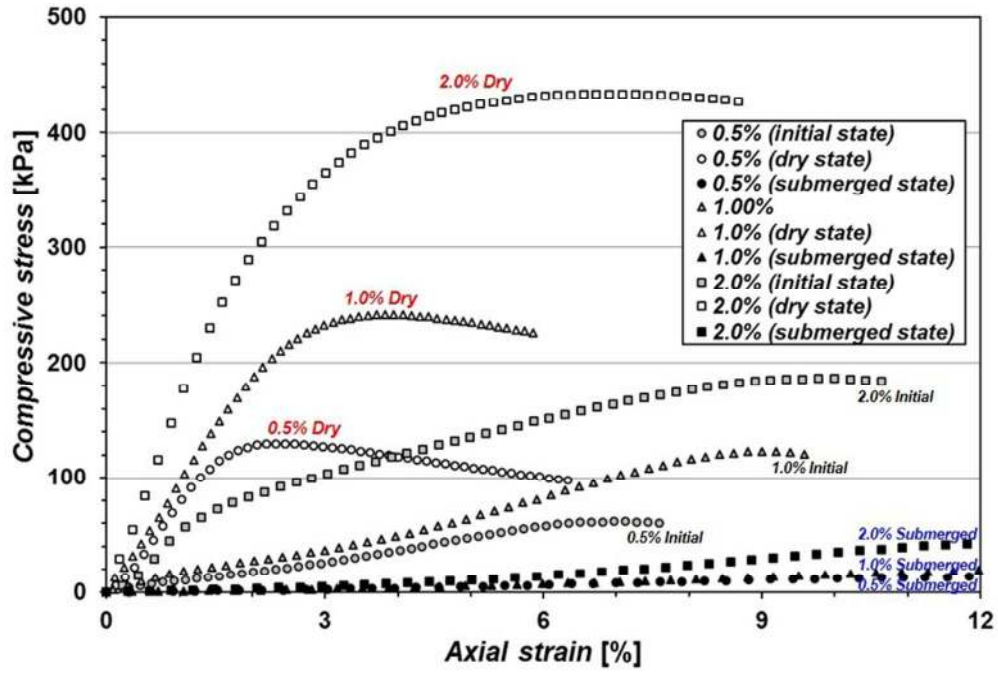
Figure 3. Stress-strain relationships of gellan gum-treated sand under unconfined compression.
144x96mm (150 x 150 DPI)
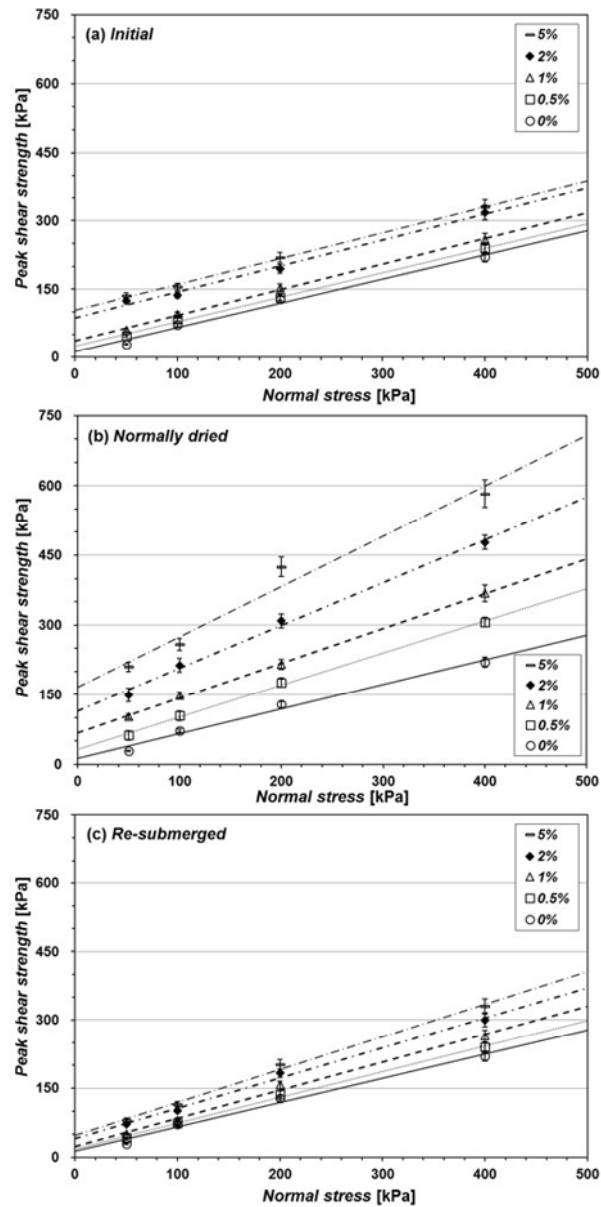
Figure 4. Peak strength of gellan gum-treated sand. (a) Initial condition. (b) Dried condition (28 days). (c) Re-submerged condition (24 hours before testing).
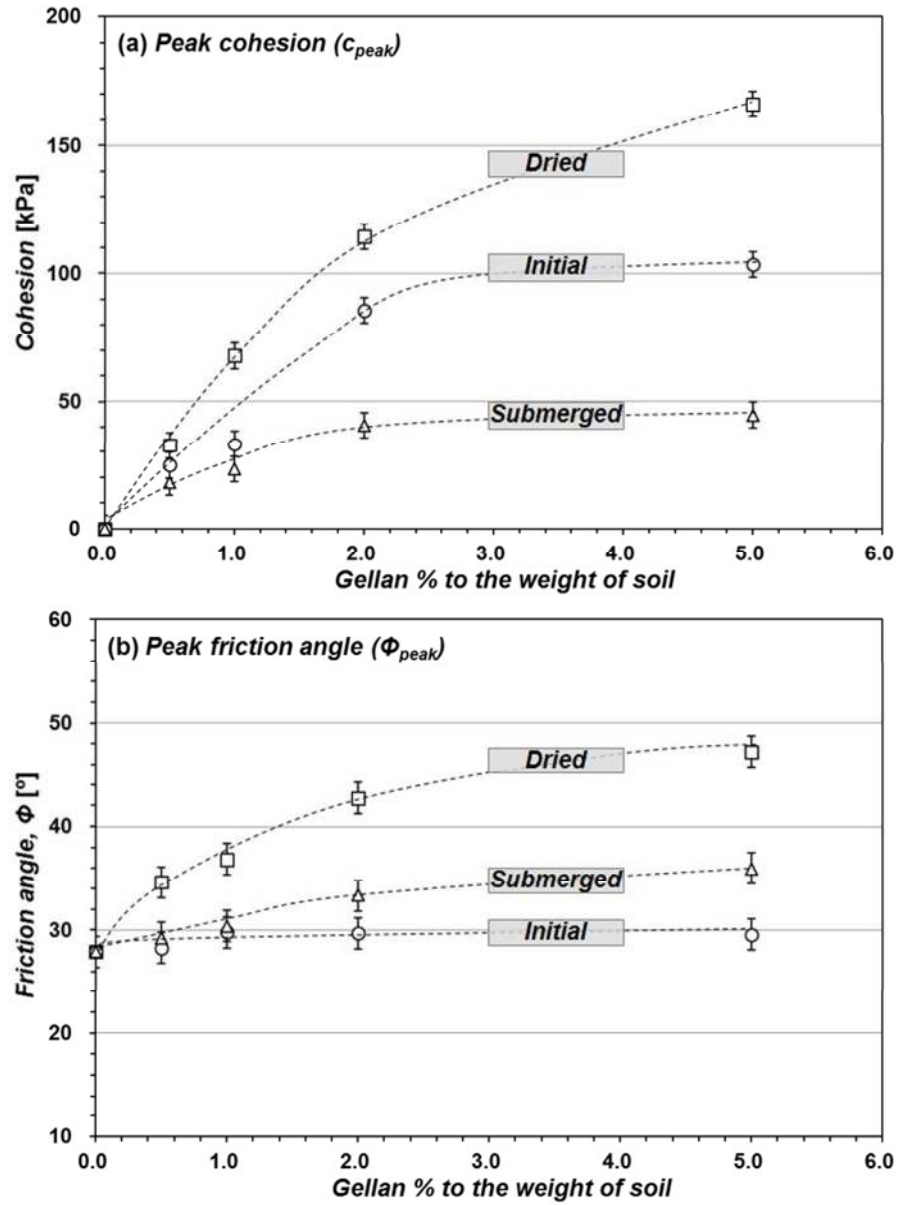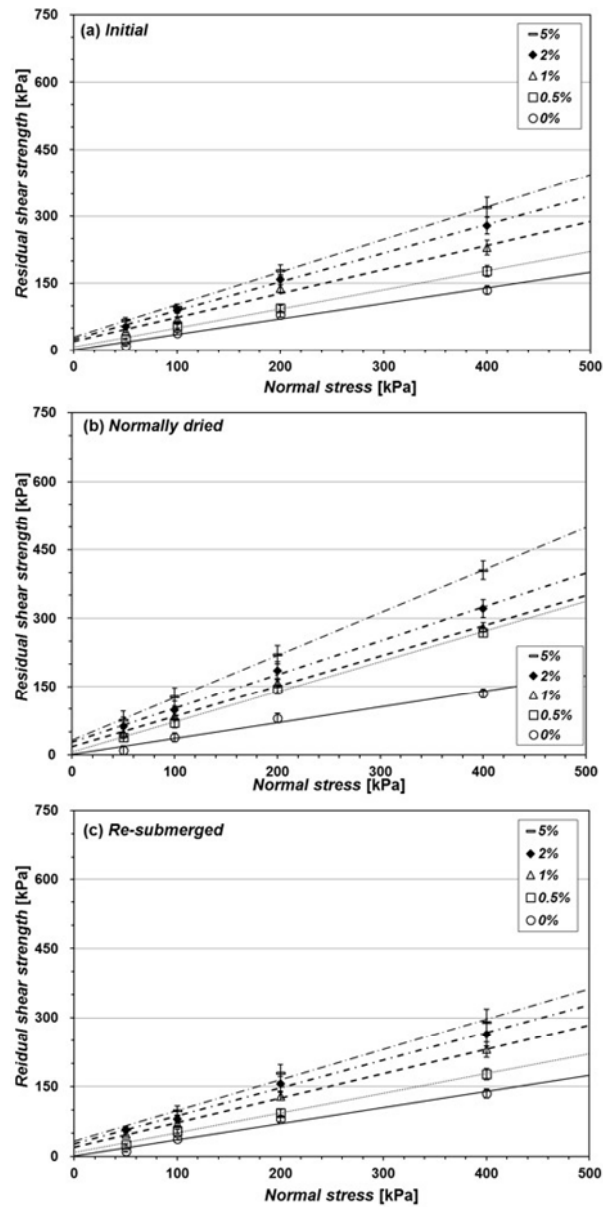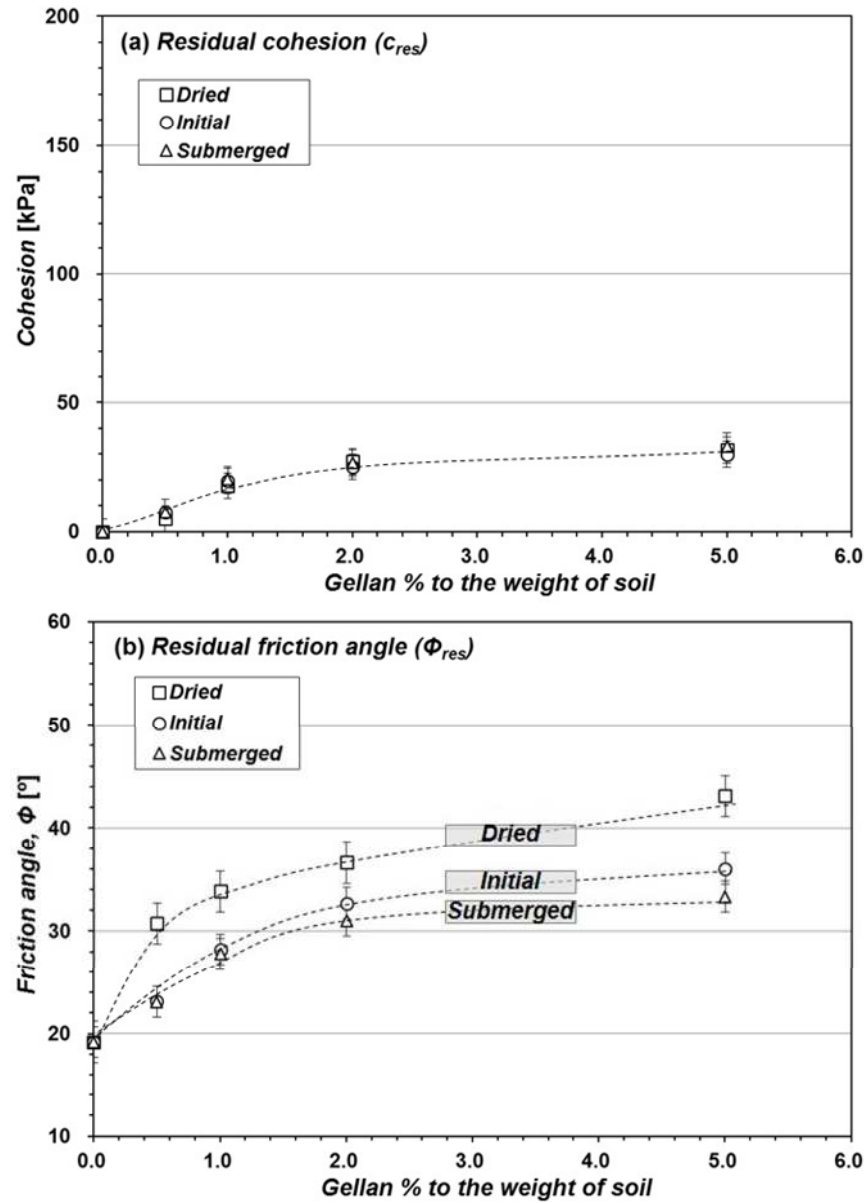85x171mm (150 x 150 DPI)

Figure 5. Peak strength properties of gellan gum-treated sand. (a) Cohesion. (b) Friction angle.
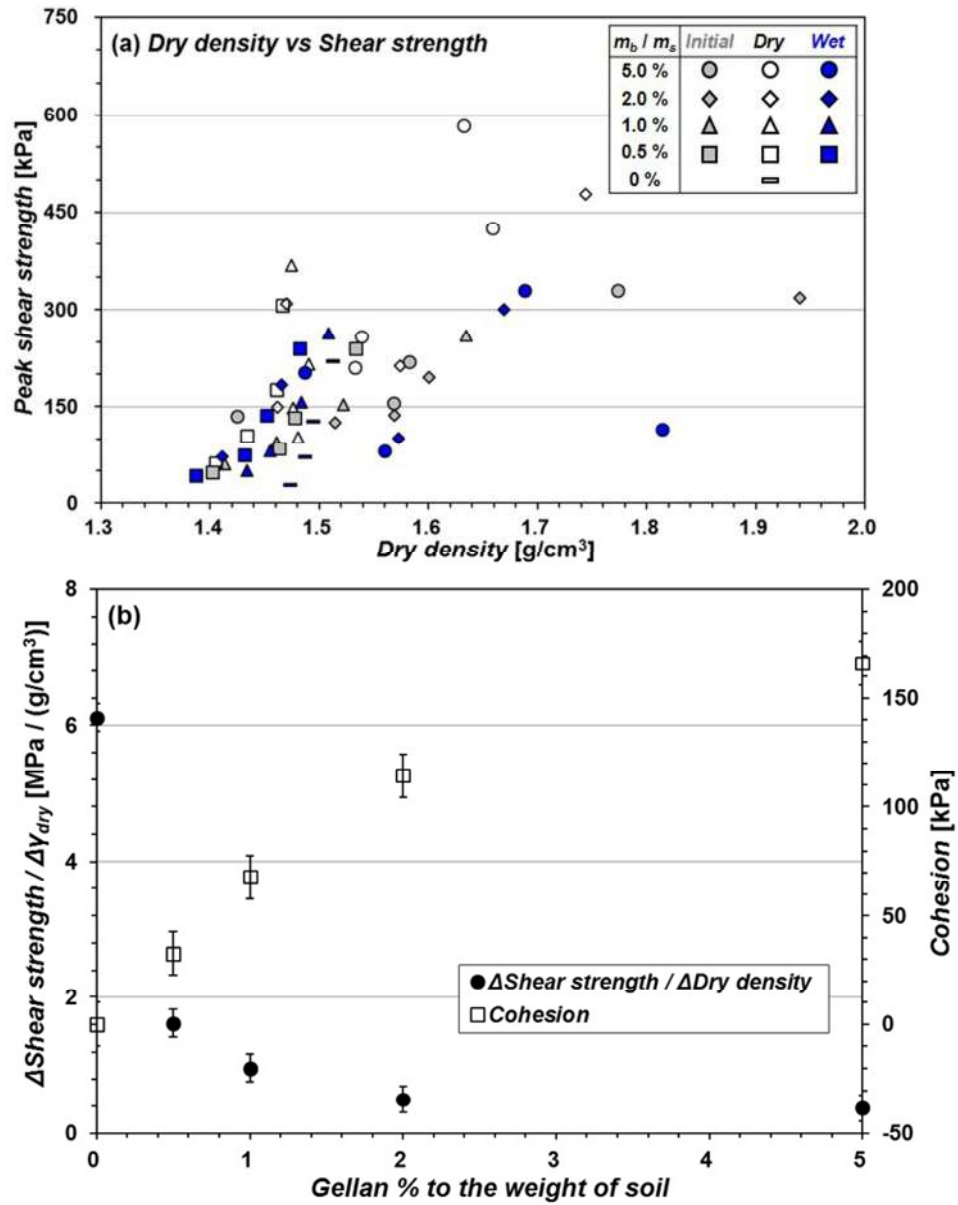120x159mm (150 x 150 DPI)

Figure 6. Residual strength of gellan gum-treated sand. (a) Initial condition. (b) Dried condition (28 days).
(c) Re-submerged condition (24 hours before testing).
85x171mm (150 x 150 DPI)

Figure 7. Residual strength properties of gellan gum-treated sand. (a) Cohesion. (b) Friction angle.
120x165mm (150 x 150 DPI)

Figure 8. Peak shear strength of gellan gum-treated sand (a) versus dry density (b) versus gellan gum content.
129x162mm (150 x 150 DPI)
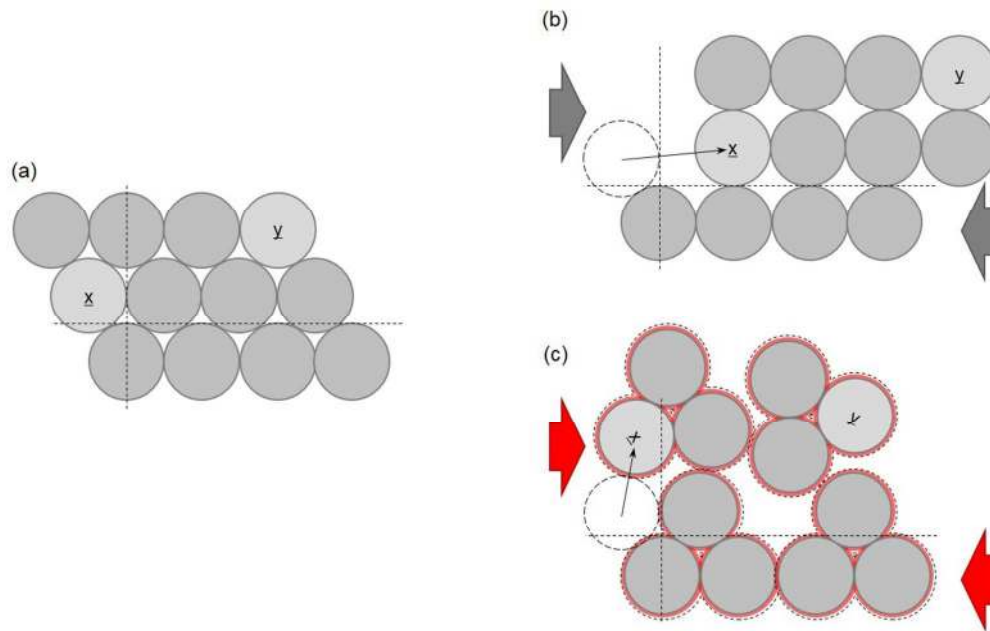
Figure 9. Schematic diagram of sands under shearing. (a) Natural (untreated) sand before shear. (b) Untreated sand under shear in which particles rotate along the shear plane. (c) Aggregation effect of gellan gum-sand mixtures, which induces a higher friction angle (ϕpeak) in a dried condition.
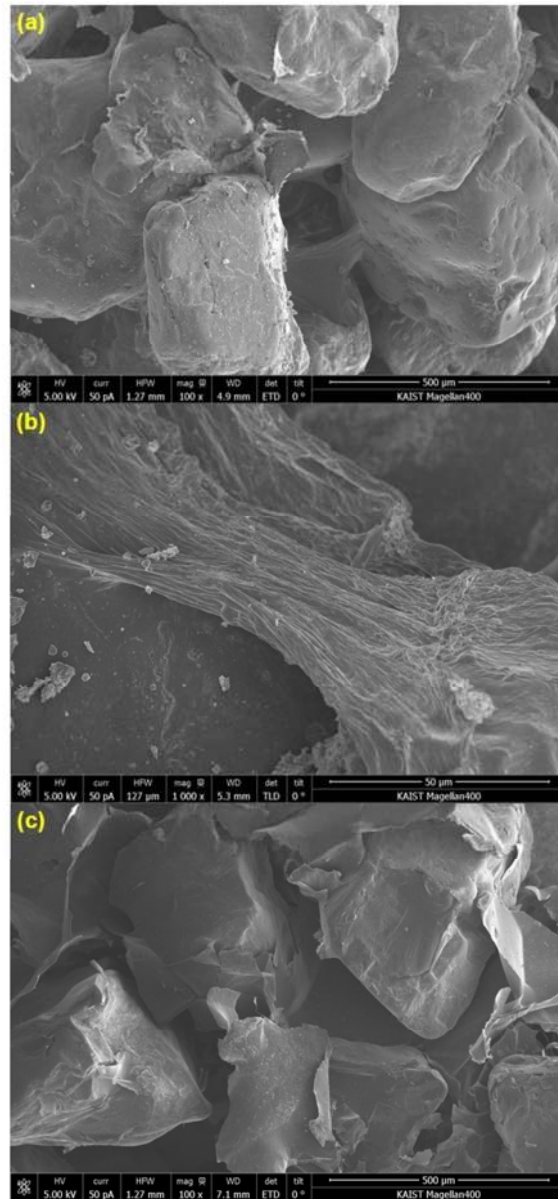199x126mm (150 x 150 DPI)

Figure 10. SEM images of 1% gellan gum-treated sand. (a) Before UTM testing (undisturbed). (b) Gellan gum films accumulated between particles (undisturbed). (c) After UTM testing (crushed)
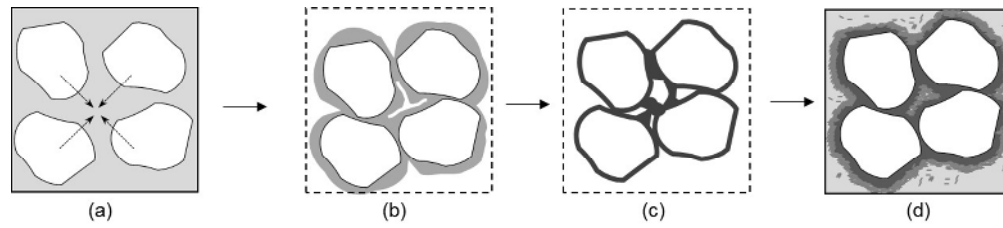86x183mm (150 x 150 DPI)

Figure 11. Drying and wetting mechanisms of gellan treated sands (a) Initial condition. (b) Propagation of cracks in gel with drying. (c) Dried condition. (d) Re-submerged condition.
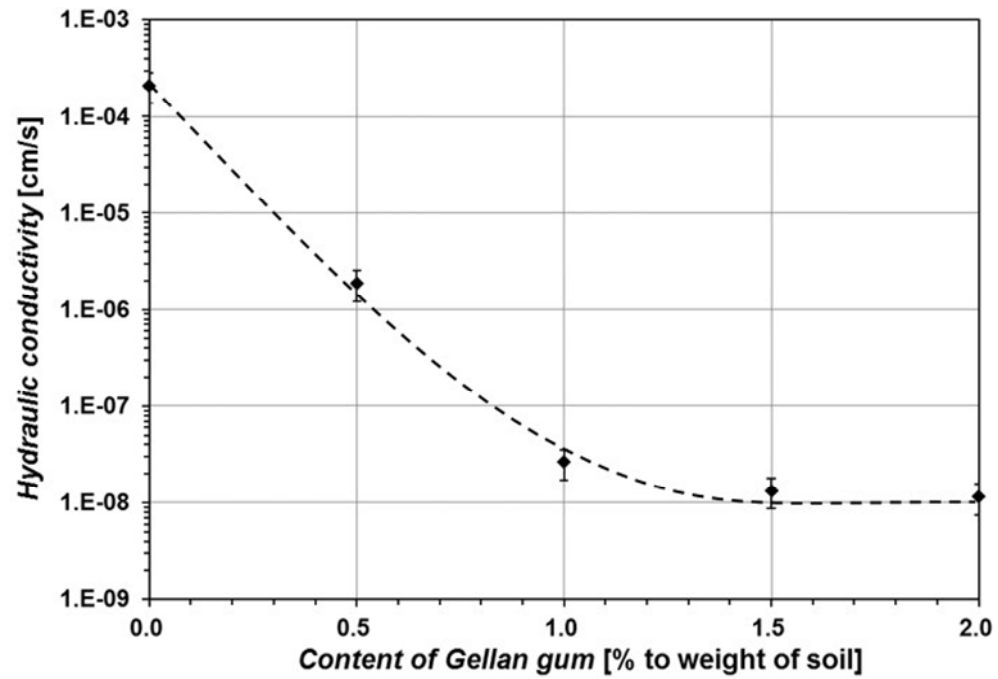211x48mm (150 x 150 DPI)

Figure 12. Hydraulic conductivity of gellan gum-treated sand in the initial state (without drying) with biopolymer content (by weight).
120x80mm (150 x 150 DPI)