

25 **Keywords:** Collapsible soils; Soil improvement; Clay nanoparticles; Grouting; Solid bridges

26 INTRODUCTION

27 More than 10% of the land worldwide is composed of collapsible soils, such as loess
28 deposits, mainly in arid and semiarid regions (Gaaver 2012; Assadi-Langroudi et al.
29 2018). The primary geotechnical concern of the unsaturated, metastable-structured
30 loess deposits is their significant volume change when subject to increasing mechanical
31 stress or decreasing matric suction (e.g., during (re)wetting), or combination of both
32 (Popescu 1986; Jiang et al. 2014; Haeri et al. 2014; Boixadera et al. 2015). Loess is
33 an aeolian sediment formed by aggregation of predominantly silt-sized particles (mode
34 20–60 μm) with often a small fraction of clays, typically in the range of 15–20 %
35 wt (Mitchell et al. 2005; Indraratna et al. 2015). The aggregates have a relatively
36 loose, open structure often featured by weak interparticle cementation bonds (Smalley
37 and Vita-Finzi 1968; Coudé-Gaussen 1987; Mitchell et al. 2005; Assadi-Langroudi
38 et al. 2018). Upon hydration, bonds are reduced and the inter-aggregate contacts fail
39 during shear, leading them to collapse under the applied load or even the self-weight
40 (Dudley 1970; Barden et al. 1973; Pereira and Fredlund 2000). Such wetting-induced
41 collapse mechanism can cause a reduction of the total soil volume by up to 15%
42 (Waltham 2002). Mechanical loading followed by wetting can be problematic when
43 building civil engineering structures on loess deposits, where the water content varies
44 due to intermittent precipitation events, irrigation, or change in the ground water level
45 (Clevenger 1956; Handy 1973).

46 Various techniques have been used to improve the mechanical behaviour of col-
47 lapsible soils including compaction and replacement (Mechanical methods), and sta-
48 bilisation (Chemical method). Chemical stabilisers include, but are not limited to,
49 cements (Horpibulsuk et al. 2010; Mohamed and El Gamal 2012), polymers (Arulrajah
50 et al. 2016; Latifi et al. 2016; Ayeldeen et al. 2017), fly ash (Arulrajah et al. 2016),

51 inorganic salts (Abbeche et al. 2010) and bituminous materials (Hoy et al. 2016). The
52 treatment often involves mixing soil with stabilisers (Ghadir and Ranjbar 2018), or
53 grouting the soil with solutions containing reactive particles such as cement, resin or
54 lime (Ibragimov 2005; Gallagher et al. 2007). Due to the low permeability of loess
55 deposits, the *in site* treatment methods using cements are limited to mainly mixing, soil
56 piles, and compaction grouting.

57 While ordinary Portland cement and lime are the most favoured materials in soil
58 stabilisation, chemical degradation under for instance internal sulfate attack (Schmidt
59 et al. 2009; Neville 2004) impose a threat for the long-term stability and functionality
60 of such soil binders. Furthermore, the production of cements raises several environ-
61 mental concerns, including high carbon dioxide emission, dust generation, and source
62 material depletion (Bosoaga et al. 2009; Fatehi et al. 2018). In recent years, application
63 of nanomaterials to enhance the hydromechanical behaviour of fine-grained soils with
64 less environmental drawbacks has received increasing attention (Luo et al. 2012; Taha
65 and Taha 2012; Iranpour et al. 2016; Bahmani et al. 2014; Latifi et al. 2015; Latifi
66 et al. 2016). Among various nano-sized additives such as copper, alumina, and silica
67 particles, mixing clay nanoparticles with soils is reported to decrease the soil collapse
68 potential, giving rise to a sustainable and environmentally friendly soil stabiliser (Iran-
69 pour et al. 2016; Latifi et al. 2016; Latifi et al. 2017). Clay minerals are one of the most
70 stable and abundant materials on the earth surface with less processing efforts required
71 compared to other synthesised stabilisers such as cements.

72 Less attention has been paid to permeation grouting to enhance the hydromechanical
73 behaviour of low permeable loess deposits mainly due to their low hydraulic conductiv-
74 ity. Permeation grouting involves injection of a solution or slurry containing stabilising
75 material in soil porous structure, where the grout will eventually turn into soil binders.
76 The binders increase the mechanical strength of the soil structure by typically forming
77 chemically-induced interparticle bonds such as calcium silicate hydrate (C-S-H). A uni-

78 form distribution of the grout in soil medium increases grouting efficiency, and is thus
79 economically favourable. Grout typically has large water content and will be subject to
80 evaporation immediately after injection. Aside from the chemical processes involved in
81 most grouts such as cement slurry, the variation of capillary suction upon evaporation
82 may play a significant role in the grout-soil interactions and the final formation of soil
83 binders. Seiphoori et al. (2020) experimentally showed that when suspensions con-
84 taining polydisperse particles are subject to evaporation, capillary suction condenses
85 small particles ($<5 \mu\text{m}$ size) in the capillary bridges formed between larger grains.
86 After evaporation of the solvent (e.g., water), small particles turn into solid bridges
87 that significantly increase the interparticle strength, giving rise to an effective cohe-
88 sion (Seiphoori et al. 2020). Smectite-based clay minerals such as montmorillonite
89 are characterised by nano-sized particles and large specific surface area. Evaporation-
90 induced bonds formed by montmorillonite nanoparticles might improve the collapsible
91 soil behaviour; however, to the authors' knowledge, application of clay nanoparticles
92 as a grout in stabilising collapsible formations is not yet investigated.

93 Here we use montmorillonite nanoparticles for grouting a collapsible loess deposit.
94 Large undisturbed loess samples were injected using solutions with different clay con-
95 tents, and after a certain curing time, the 1-D collapse potential of the grouted materials
96 were determined in the laboratory. Ordinary cement slurry was also used to represent
97 the most favoured grout in practice and to compare with clay. Furthermore, recon-
98 stituted specimens were prepared by mixing clay or cement particles with the loess
99 material at similar additive contents and curing times to allow a comparison with the
100 grouting results.

101 MATERIALS AND METHOD

102 Materials

103 *Natural collapsible soil*

104 The collapsible soil material was collected from Semnan Province located in the
105 subtropical areas of Iran. After removing about the top soil (~50 cm) from the original
106 ground surface, thin-walled cylindrical samplers were used to acquire undisturbed
107 material with dimensions of 35cm×35cm. The material was then sealed using paraffin
108 and transported to the laboratory. The basic physical and engineering properties of the
109 tested soil material are listed in Table 1. The Atterberg limits and specific gravity of
110 the soil were measured for three samples using the ASTM D4318 and ASTM D854
111 methods, respectively. The unit weight of the soil was determined at the site using
112 the sand-cone method (ASTM D1556-07). The index properties indicate that the fines
113 content primarily consist of silt. Furthermore, the soil particle size distribution was
114 determined using dry sieving and hydrometer tests (for $d < 75 \mu m$) as shown in Fig.
115 1-A. The soil is thus classified as low plasticity silt. Fig. 1-B presents scanning electron
116 photomicrographs of the soil aggregates, where the porous microfabric is observed to
117 consist of fine sand grains bonded by silt/clay-sized particles (see the inset image).
118 The scanning electron microscope (Hitachi SU3500; Japan) was operated under high
119 vacuum conditions at 15 keV of accelerating voltage in a backscattered electron mode.
120 The hydraulic conductivity of the soil was evaluated using the falling head method
121 (ASTM D5084, initial water height of 100 cm) with an average value of $7.7 \times 10^{-6} m/s$
122 which implies a low permeable silt-based soil. The collapse index of the undisturbed
123 and reconstituted specimens from the untreated soil studied here was evaluated to be
124 9.1% and 11.3%, respectively, with *moderately sever* to *sever* degree of collapsibility
125 based on ASTM D5333 (see Figure 3-C).

126 *Soil stabilisers*

127 In this study, Na-montmorillonite clay (NC) and ordinary Portland cement (OPC)
128 were used as soil stabilisers. Montmorillonite is a naturally occurring and reactive clay
129 mineral which belongs to the smectite mineral group. Montmorillonite k10 (Sigma-
130 Aldrich, USA) was used in this research, which is a highly porous substance with a
131 larger surface area and nanopores. It is chemically modified by the cation-exchange
132 method which results in reducing its swelling potential (Maiti et al. 2016; Alekseeva
133 et al. 2019). When dry montmorillonite particles are mixed with water to establish
134 the grouting solution, particles disperse upon further hydration. As time progresses,
135 the dispersed montmorillonite particles may aggregate and form larger clusters. The
136 particle size distribution of aqueous suspension of montmorillonite particles in distilled
137 water is presented in Figure 2 (Alekseeva et al. 2019), with an effective particle size
138 of 246 nm. The montmorillonite clay features a large specific surface area, $S_{BET}=195$
139 m^2/g (Alekseeva et al. 2019), and ion exchange capacity of about 48 meq/100 g (Sigma-
140 Aldrich, USA).

141 The cement used in this study was type II Portland (OPC) according to ASTM
142 C150, obtained from Gharb Cement Manufacturing in Iran. The specific surface area
143 of the OPC was evaluated to be $0.32 m^2/g$, significantly lower than that of the NC
144 material. The particle size distribution of the cement is also presented in Figure 2-B
145 with a dominant particle size of $\sim 13.3 \mu m$, 1-2 orders of magnitude larger than that of
146 the NC effective particle size.

147 **Sample preparation**

148 *Soil grouting setup*

149 A schematic view of the grouting system designed in this research is presented in
150 Figure 3-A. The system comprised a fluid tank positioned within an adjustable frame
151 to inject slurry under a constant initial pressure head (h_0) by changing the elevation

152 of the fluid tank. The scaling factor of the injection setup was about 1/15 of a typical
153 full-scale injection system (Nichols and Goodings 2000; Iai et al. 2005). The grout
154 solid content was considered based on the weight of the dry stabiliser particles per
155 total weight of the cylindrical soil sample (~30 kg). Solid contents of 0.5, 1, and 2.5
156 wt.% for OPC, and 0.05, 0.1, and 0.25 wt.% for NC were selected. It is noted that
157 the OPC contents are one order of magnitude larger than NC contents for grouting
158 similar soil volume to achieve comparable impacts on the soil collapse index. The
159 grout was prepared by mixing dry particles with 1L of deionized water. As a result, the
160 water content of OPC and NC grouts varies in the range of (57–87%) and (93–98%),
161 respectively. Mixing was conducted using a lab mixer for a short period of 2 min.
162 Prior to injection the surface of the samples was flattened and an injection hole was
163 drilled by rotating an open-ended tube with an outer diameter of 7 mm down into the
164 undisturbed sample along its height. The slurry was transmitted from the fluid tank into
165 the soil through a connection tube, where it was injected uniformly using a perforated
166 rod (holes of 0.6 mm diameter and spacing of 2.0 cm along the rod). The injection
167 pressure which is a function of soil overburden pressure is a key parameter of grouting
168 performance. Different grouting heads were applied to determine an optimum initial
169 grouting pressure head. Surface rupture was observed at high injection pressures (>14
170 kPa) and low grout penetration at low hydraulic pressure (<4 kPa). The optimum
171 injection pressure was thus obtained to be about 7 kPa, approximately 1.5 times greater
172 than the soil maximum *in-situ* overburden pressure. As the injection proceeds, the
173 pressure head drops and after a given period of time the tank supply is exhausted (*i.e.*,
174 injection under a falling head). We note that, under the above-described conditions, the
175 NC grout solution takes up to about 2 min to be injected through the soil matrix, while
176 cement grout taking up to 20 min at higher concentration likely due to the difference
177 in their effective aggregate size and thus their mobility in soil porous structure (Figure
178 2-B) and the soil pore size distribution. After grouting, the samples were cured for

179 7, 14 and 28 days under open air condition with $T = 30 \pm 6^\circ\text{C}$ and $\text{RH} = 21 \pm 11\%$ (with
 180 average annual variations of $T = 22 \pm 14^\circ\text{C}$ and $\text{RH} = 43 \pm 33\%$ recorded for the loess site
 181 location). Undisturbed specimens were then taken from the centre and corners of the
 182 grouted sample (see Figure 3-A) and trimmed to evaluate their collapse potential in an
 183 oedometer cell (Figure 3-B).

184 *Soil mixing*

185 A number of specimens were reconstituted to compare with the grouted materials.
 186 Oven-dried soil was homogeneously mixed with dry cement or clay particles and then
 187 reconstituted in an oedometer mould at dry unit weight and water contents consistent
 188 with that of the *in situ* material (*i.e.*, $\gamma_d = 14 \text{ kN/m}^3$ and $w = 5\%$). The additive
 189 contents of 0.5, 1, and 2.5 wt.% for OPC, and 0.05, 0.1, and 0.25 wt.% for NC were
 190 selected. A wet under-compaction method (Ladd 1978) was used to prepare specimens
 191 with uniform density along the specimen height. Similar to the grouted specimens, the
 192 collapse potential of the reconstituted specimens was determined after 7, 14, and 28
 193 days.

194 **Collapse potential**

195 The 1-D soil collapse potential was determined through inundating the unsaturated
 196 soil specimens under a constant vertical stress in an oedometer device according to
 197 ASTM D5333. First, the specimens were incrementally ($\frac{\sigma_v[i+1]}{\sigma_v[i]} \sim 2$, $\sigma_v[0] = 5 \text{ kPa}$)
 198 loaded up to $\sigma_v[j] = 200 \text{ kPa}$ under their initial water content with 1h time intervals
 199 between each successive loading step. The specimens were inundated with distilled
 200 water after 1h under $\sigma_v[j] = 200 \text{ kPa}$ and the load was kept constant for 24 h, and
 201 the specimen deformation was recorded continuously. The 1-D collapse potential
 202 at the onset of the sudden deformation upon inundation is defined by the collapse
 203 index, $I_c = \frac{\Delta h[j]}{h[j]} = \frac{\Delta e[j]}{1+e[j]}$, where $\Delta h[j]$ and $\Delta e[j]$ are the specimen settlement and
 204 variation of void ratio due to inundation at load increment j , and $h[j]$ and $e[j]$ are

205 the specimen height and void ratio prior to collapse, respectively. The initial void
206 ratios of the grouted specimens were calculated using the weight volume relationship
207 by measuring the specimen's weight and water content. The collapse behaviour of
208 the undisturbed and reconstituted specimens from the untreated loess studied here is
209 presented in Figure 3-C. The undisturbed sample display larger compressibility and
210 collapse behaviour which is likely associate with sensitivity of the natural interparticle
211 bonds to the applied hydromechanical stress path. This indicates that the compaction
212 during reconstituting the material slightly improved the mechanical behaviour of the
213 loess.

214 **RESULTS**

215 **Stabilisation with active clay nanoparticles (NC)**

216 *Grouting*

217 The results of the compressibility behaviour of specimens acquired from the centre
218 and side of the grouted column (see Figure 3-A) at 0.25 wt.% NC and different times
219 after injection is presented in Figure 4-A and B, respectively. The collapse behaviour of
220 the untreated soil is also shown in these plots. The collapse index of the centre and side
221 specimens with different percentages of NC at 7, 14, and 28 days after treatment is also
222 presented in Figure 4-C and D. The collapse index of the untreated soil decreased from
223 11.3% to less than 0.7, 0.4, and 0.3 for centre specimens grouted with 0.05%, 0.1%
224 and 0.25wt.% NC, respectively, after 28 days. The optimum performance appears to
225 be achieved at 0.1 wt.% NC content for both centre and side specimens. This indicates
226 the high-mobility of the NC grout which was able to be uniformly distributed within
227 the soil matrix increasing the interparticle strength. Furthermore, the gain of strength
228 is likely completed in the early stages after grouting. The collapse index of the side
229 specimens slightly increased when increasing NC content to 0.25wt.%, indicating that
230 the efficiency decreased at higher grout concentration. Formation of the clay clusters

231 within the soil matrix can be observed in the SEM photomicrographs taken at various
232 NC contents after 28 days curing (Figure 5). The NC agglomerates are observed to
233 likely bind the soil grains/aggregates by forming interparticle bridges as well as filling
234 the inter-aggregate pore spaces.

235 *Mixing*

236 The compressibility and collapse behaviour of the specimens prepared by mixing
237 NC particles with soil is presented in Figure 6-A and B. Unlike the grouting, mixing
238 appears to be much less effective in reducing soil collapse, especially at higher clay
239 contents. Although mixing soil with NC particles is expected to result in a more
240 homogeneous distribution of clay particles within the soil matrix, it appears that the
241 sole presence of clay particles in the soil matrix does not result in the formation of
242 interparticle bonds (see Figure 6-C and D) as it spontaneously does during grouting.
243 Instead, coating particle surfaces likely contributes in reducing the soil strength by
244 decreasing the internal friction (Taha and Taha 2012). These experiments suggest that
245 in order to achieve a maximum performance in improving the mechanical behaviour of
246 collapsible soils using clay nanoparticles, the clay must be grouted than mixed.

247 **Stabilisation with cement (OPC)**

248 *Grouting*

249 The compressibility behaviours of specimens acquired from the centre and side
250 locations of the grouted material with 2.5 wt.% OPC after different curing times are
251 presented in Figure 7-A and B, respectively. The collapse index of the centre and side
252 specimens with different percentages of OPC after 7, 14, and 28 days is also presented
253 in Figure 7-C and D. While OPC has remarkably decreased the collapse index of the
254 soil around the grouting hole, its performance on side specimens is not acceptable,
255 especially at higher cement contents. The poor performance of cement at 2.5 wt.%
256 content is likely associated with a reduced hydraulic conductivity of the soil due to

257 flocculation of cement particles and clogging the pores. The soil strength appears to
258 be achieved during the first two weeks with an optimum performance for specimens
259 grouted with 1 wt.% OPC content, where the collapse index decreased to 0.2% and
260 1.6% for the centre and side specimens, respectively. SEM photomicrographs of the
261 materials from the centre and side specimens grouted with different OPC contents
262 are presented in Figure 8. While the soil inter-grain/aggregate pores in the centre
263 specimens are observed to be nearly filled with OPC material, the side specimens likely
264 remained more porous. The lower porosity of the centre specimens is also discernible
265 in consolidation graphs (Figure 7-A), where the initial void ratio has decreased from
266 0.9 to 0.75. The change of initial void ratio of the side material is negligible (Figure
267 7-B) which implies that the OPC slurry was not able to penetrate the soil and fill the
268 pores to decrease the porosity. This behaviour is consistent with the observation of the
269 microfabric shown in Figure 8. The specimens grouted with 1 wt.% OPC exhibits an
270 optimum performance.

271 *Mixing*

272 The compressibility and collapse behaviour of the specimens prepared by mixing
273 OPC material with soil is presented in Figure 9-A and B. Unlike the NC grout, an
274 increase in OPC content improves mechanical behaviour of the studied soil by reducing
275 the collapse index; however, the performance of mixing is significantly lower than
276 that achieved by grouting, especially at lower cement contents. It is noted that the
277 higher water content in grouting (57–86%) compared to mixing (~5%) results in
278 further hydration of the cement, where calcium silicate hydrate (C-S-H) bonds form
279 and increase the soil strength. SEM photomicrographs of the OPC-treated specimens at
280 different cement contents 28 days after mixing process are shown in Figure 9-C and D,
281 where the inter-particle bonds formed by OPC material can be observed.

DISCUSSION

The variation of the collapse index of the loess deposit 14 days after it was treated by mixing or grouting with the NC or OPC particles is presented in Figure 10. The collapse level of the untreated soil is also marked in the plot. Mixing NC at low content (0.05 wt.%) is more effective than the best performance achieved by OPC (at content of 2.5 wt.%), showing the efficiency of mixing NC particles with soil to improve mechanical behaviour with significantly less used material (i.e., 50 times less mass). The best performance of using NC particles as soil stabiliser was achieved through grouting small quantity (0.1 wt.%), which resulted in reducing the collapse index up to 96% (Figure 10). Although OPC grouting effectively reduces the collapse index of the material around the injection hole, its performance is largely impacted by the distance from the injection centre indicating its limited mobility. More importantly, the NC content is 1/10 of the OPC used to achieve comparable results. It is worth noting from the OPC grout trends that there might be an optimum OPC percentage (< 0.5 wt.%), where the collapse index of the side and centre specimens of the studied loess reaches about 1%; nevertheless, the application of NC is economically favourable and ecologically compatible.

Now the question is why grouting NC particles is remarkably effective (and efficient) in improving the the mechanical behaviour. The answer lies in two distinct features involved in the grouts of montmorillonite clay nanoparticles (NC) and the subsequent evaporation of the grout as time proceeds: (1) montmorillonite particles are remarkably smaller ($\sim 0.25\mu\text{m}$ size) than cement particles ($\sim 13\mu\text{m}$ size) (see Figure2-B), which facilitates their transport in soil porous skeleton; (2) subsequent evaporation of the grouted solution results in the formation of strong solid bridges which significantly increase the soil strength. Grout mobility in soil depends on a number of physical and chemical factors including the grout particle size and the pore size distribution of the soil medium as well as the interfacial interaction of the grout particles and soil grains

309 (Semmler et al. 2000; Auset and Keller 2004; Bradford et al. 2002). Larger particles or
310 aggregates are more likely to be trapped in narrow soil pore throats and thus prevent a
311 uniform distribution of the grout in the soil matrix during injection. Modified Kozeny-
312 Carman equation, $K = 0.0898 \frac{D^2}{\mu} \gamma_w \phi^{3.4}$ (Lala 2018), results in an effective pore size,
313 $D \sim 3 \mu\text{m}$, where K is the hydraulic conductivity of the tested soil (See Table 1), ϕ is the
314 porosity (0.48), μ is the water viscosity, and γ_w is the unit weight of water. It is noted
315 that the soil permeability to NC grout is expected to be larger than the permeability
316 obtained from falling head method which can potentially modify the pore structure;
317 nevertheless, this equation provides an estimate of the effective pore size of the loess
318 deposit studied here. This explains why the NC grout is more effective than OPC for
319 the side specimens that are far from the injection hole. An increase in the clay content
320 however encourages the formation of clay gel or weakly agglomerated particles that
321 may clog the soil pore throats, and thus decrease the effective permeability of the soil
322 matrix as seen for side specimen of NC grout at 0.25 wt. %.

323 Immediately after injecting the grout, the solution will be subject to evaporation.
324 The NC grout featuring a large initial water content (93–98%) will follow a drying path
325 similar to that described in Figure 11 (water retention data modified from (Seiphoori
326 et al. 2014)). As evaporation proceeds, capillary bridges form between adjacent soil
327 grains/aggregates, while the diminishing volume of the bridge leads NC particles to
328 form a clay gel. Further evaporation under the relatively low humidity of the site
329 (Average RH~21%) will turn the NC gel into solid structures that bind the particles
330 and reinforce the interparticle bonds, giving rise to an effective cohesion (Figure 11).
331 These bonds referred to as "solid bridges" can increase the interparticle strength by
332 orders of magnitude (Seiphoori et al. 2020) depending on the grout effective particle
333 size and its solid fraction in the capillary bridges. Seiphoori et al. (2020) show that
334 this interparticle cohesive force originates from the sum of the van der Waals bonds
335 within the solid bridges.

336 Furthermore, unlike the OPC-grouted material, in NC-injected specimens, the
337 mechanical improvement has been achieved in the early stages once the grout water is
338 further evaporated. The much less quantity of NC particles does not drastically change
339 the permeability of the soil, while the high content OPC grouting reduces the soil
340 porosity and thus permeability. The disturbance to soil fabric and chemistry would be
341 significantly minimised when NC particles are grouted as soil stabilisers.

342 CONCLUSIONS

343 Permeation grouting relies on the injection of grouts into soil porous skeleton to im-
344 prove its mechanical properties through formation or reinforcement of soil interparticle
345 bonds. The grout high-mobility in soil is a key parameter to optimise the cost and to
346 predict the grouting end performance. The conventional permeation grouting is limited
347 to soils which contain small fraction of fine particles (<15%). On the other hand, the
348 geochemical stability of the grout is important for the long-term performance of the
349 grouted geomaterials. Here we presented a grouting approach based on injecting a low
350 permeable collapsible soil with solutions containing montmorillonite clay nanoparti-
351 cles. The clay particle size facilitates its mobility in soil, while its large specific surface
352 area results in strong capillary-driven interparticle bonds (i.e., solid bridges). The
353 solid bridges driven by evaporation form relatively fast, especially in the semi-arid/arid
354 regions with geological formations susceptible to collapse upon rewetting. We showed
355 that the formation of solid bridges is facilitated through grouting, and mixing same
356 amount of dry clay material with soil does not lead to same results; coating soil grain
357 surfaces with clay particles indeed reduces the soil strength by decreasing the internal
358 friction. The montmorillonite clay is likely an ideal material to form solid bridges
359 through providing a large number of microscopic contacts where the interfacial bonds
360 form, giving rise to an effective cohesion. In order for increasing the mobility of
361 conventional grouts, ultrafine cements are typically an option; however, the specific

362 surface area of the cement does not increase significantly by reducing the particle size.
363 For instance, an ultrafine cement with 90 *wt.%* below $7.32\mu\text{m}$, the specific surface
364 area is $0.725\text{ m}^2/\text{g}$ (Sarkar and Wheeler 2001), orders of magnitude smaller than that of
365 montmorillonite clay. Typical cement materials are susceptible to chemical degradation
366 which impacts the long-term functionality of the grout, while clays typically exhibit
367 high stability to chemical variation of the pore water. In addition, the production of
368 cement raises several environmental concerns, such as the emission of carbon dioxide
369 and depletion of the source material, encouraging the application of soil stabilisers
370 with less processing efforts such as clays. Our results thus suggest the application of
371 clay nanoparticles solutions for a fast, economical and more environmentally-friendly
372 grouting approach compared to cements, which likely results in formation of chemically
373 more stable interparticle bonds, and hence an improved performance of the grouted
374 soils. The proposed approach may help to improve the mechanical behaviour of geo-
375 materials susceptible to creep, liquefaction, or erosion which involve the disintegration
376 of metastable soil aggregates.

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380 **NOTATION**

381 d soil particle diameter

382 D_x particle size for the finer material at which x percent of the material is finer

383 NC Na-montmorillonite/Nano-sized clay

384 OPC Ordinary Portland cement

385 S_{BET} BET specific surface area

386 h_0 initial grouting head

387 *wt.* weight ratio

388	RH	relative humidity
389	V	volume
390	σ_v	vertical stress
391	I_c	collapse index
392	h	specimen height
393	e	void ratio
394	Δh	height increment
395	Δe	void ratio increment
396	K	hydraulic conductivity
397	D	effective pore size of soil
398	ϕ	soil porosity
399	μ	viscosity of water
400	γ_w	unit weight of water
401	γ_d	dry unit weight of soil
402	w_0	initial water content
403	w_l	liquid limit water content
404	w_p	plastic limit water content
405	PI	Plastic Index
406	G_s	solid particles specific gravity

407

408 REFERENCES

- 409 Abbeche, K., Bahloul, O., Ayadat, T., and Bahloul, A. (2010). "Treatment of collapsible
410 soils by salts using the double consolidation method." *Experimental and Applied
411 Modeling of Unsaturated Soils*, 69–78.
- 412 Alekseeva, O., Noskov, A., Grishina, E., Ramenskaya, L., Kudryakova, N., Ivanov, V.,

- 413 and Agafonov, A. (2019). “Structural and thermal properties of montmorillonite/ionic
414 liquid composites.” *Materials*, 12(16), 2578.
- 415 Arulrajah, A., Mohammadinia, A., Phummiphan, I., Horpibulsuk, S., and Samingthong,
416 W. (2016). “Stabilization of recycled demolition aggregates by geopolymers
417 comprising calcium carbide residue, fly ash and slag precursors.” *Construction and
418 Building Materials*, 114, 864–873.
- 419 Assadi-Langroudi, A., Ng’ambi, S., and Smalley, I. (2018). “Loess as a collapsible
420 soil: Some basic particle packing aspects.” *Quaternary International*, 469, 20–29.
- 421 Auset, M. and Keller, A. A. (2004). “Pore-scale processes that control dispersion of
422 colloids in saturated porous media.” *Water Resources Research*, 40(3).
- 423 Ayeldeen, M., Negm, A., El-Sawwaf, M., and Kitazume, M. (2017). “Enhancing
424 mechanical behaviors of collapsible soil using two biopolymers.” *Journal of Rock
425 Mechanics and Geotechnical Engineering*, 9(2), 329–339.
- 426 Bahmani, S. H., Huat, B. B., Asadi, A., and Farzadnia, N. (2014). “Stabilization
427 of residual soil using sio2 nanoparticles and cement.” *Construction and Building
428 Materials*, 64, 350–359.
- 429 Barden, L., McGown, A., and Collins, K. (1973). “The collapse mechanism in partly
430 saturated soil.” *Engineering Geology*, 7(1), 49–60.
- 431 Boixadera, J., Poch, R. M., Lowick, S. E., and Balasch, J. C. (2015). “Loess and soils
432 in the eastern ebro basin.” *Quaternary international*, 376, 114–133.
- 433 Bosoaga, A., Masek, O., and Oakey, J. E. (2009). “Co2 capture technologies for cement
434 industry.” *Energy procedia*, 1(1), 133–140.
- 435 Bradford, S. A., Yates, S. R., Bettahar, M., and Simunek, J. (2002). “Physical factors
436 affecting the transport and fate of colloids in saturated porous media.” *Water resources
437 research*, 38(12), 63–1.
- 438 Clevenger, W. A. (1956). “Experiences with loess as foundation materials.” *Journal of
439 the Soil Mechanics and Foundations Division*, 82(3), 1–26.

- 440 Coudé-Gaussen, G. (1987). “The perisaharan loess: sedimentological characterization
441 and paleoclimatical significance.” *GeoJournal*, 15(2), 177–183.
- 442 Dudley, J. H. (1970). “Review of collapsing soils.” *Journal of the Soil Mechanics and*
443 *Foundations Division*, 96(3), 925–947.
- 444 Fatehi, H., Abtahi, S. M., Hashemolhosseini, H., and Hejazi, S. M. (2018). “A novel
445 study on using protein based biopolymers in soil strengthening.” *Construction and*
446 *Building Materials*, 167, 813–821.
- 447 Gaaver, K. E. (2012). “Geotechnical properties of egyptian collapsible soils.” *Alexan-*
448 *dria Engineering Journal*, 51(3), 205–210.
- 449 Gallagher, P. M., Pamuk, A., and Abdoun, T. (2007). “Stabilization of liquefiable soils
450 using colloidal silica grout.” *Journal of Materials in Civil Engineering*, 19(1), 33–40.
- 451 Ghadir, P. and Ranjbar, N. (2018). “Clayey soil stabilization using geopolymer and
452 portland cement.” *Construction and Building Materials*, 188, 361–371.
- 453 Haeri, S. M., Garakani, A. A., Khosravi, A., and Meehan, C. L. (2014). “Assessing the
454 hydro-mechanical behavior of collapsible soils using a modified triaxial test device.”
455 *Geotechnical Testing Journal*, 37(2), 190–204.
- 456 Handy, R. L. (1973). “Collapsible loess in iowa 1.” *Soil Science Society of America*
457 *Journal*, 37(2), 281–284.
- 458 Horpibulsuk, S., Rachan, R., Chinkulkijniwat, A., Raksachon, Y., and Suddeepong,
459 A. (2010). “Analysis of strength development in cement-stabilized silty clay from
460 microstructural considerations.” *Construction and building materials*, 24(10), 2011–
461 2021.
- 462 Hoy, M., Horpibulsuk, S., and Arulrajah, A. (2016). “Strength development of recycled
463 asphalt pavement–fly ash geopolymer as a road construction material.” *Construction*
464 *and Building Materials*, 117, 209–219.
- 465 Iai, S., Tobita, T., and Nakahara, T. (2005). “Generalised scaling relations for dynamic
466 centrifuge tests.” *Geotechnique*, 55(5), 355–362.

- 467 Ibragimov, M. (2005). “Soil stabilization with cement grouts.” *Soil mechanics and*
468 *foundation engineering*, 42(2), 67–72.
- 469 Indraratna, B., Chu, J., and Rujikiatkamjorn, C. (2015). *Ground improvement case*
470 *histories: compaction, grouting and geosynthetics*. Butterworth-Heinemann.
- 471 Iranpour, B. et al. (2016). “The influence of nanomaterials on collapsible soil treatment.”
472 *Engineering geology*, 205, 40–53.
- 473 Jiang, M., Li, T., Hu, H., and Thornton, C. (2014). “Dem analyses of one-dimensional
474 compression and collapse behaviour of unsaturated structural loess.” *Computers and*
475 *Geotechnics*, 60, 47–60.
- 476 Ladd, R. (1978). “Preparing test specimens using undercompaction.” *Geotechnical*
477 *Testing Journal*, 1(1), 16–23.
- 478 Lala, A. M. S. (2018). “Modifications to the kozeny–carman model to enhance petro-
479 physical relationships.” *Exploration Geophysics*, 49(4), 553–558.
- 480 Latifi, N., Eisazadeh, A., Marto, A., and Meehan, C. L. (2017). “Tropical residual soil
481 stabilization: A powder form material for increasing soil strength.” *Construction and*
482 *Building Materials*, 147, 827–836.
- 483 Latifi, N., Horpibulsuk, S., Meehan, C. L., Abd Majid, M. Z., Tahir, M. M., and
484 Mohamad, E. T. (2016). “Improvement of problematic soils with biopolymer—an
485 environmentally friendly soil stabilizer.” *Journal of Materials in Civil Engineering*,
486 29(2), 04016204.
- 487 Latifi, N., Rashid, A. S. A., Siddiqua, S., and Horpibulsuk, S. (2015). “Micro-structural
488 analysis of strength development in low-and high swelling clays stabilized with
489 magnesium chloride solution—a green soil stabilizer.” *Applied Clay Science*, 118,
490 195–206.
- 491 Luo, H.-L., Hsiao, D.-H., Lin, D.-F., and Lin, C.-K. (2012). “Cohesive soil stabilized
492 using sewage sludge ash/cement and nano aluminum oxide.” *International Journal*
493 *of Transportation Science and Technology*, 1(1), 83–99.

- 494 Maiti, S., Pramanik, A., Chattopadhyay, S., De, G., and Mahanty, S. (2016). “Electro-
495 chemical energy storage in montmorillonite k10 clay based composite as superca-
496 pacitor using ionic liquid electrolyte.” *Journal of colloid and interface science*, 464,
497 73–82.
- 498 Mitchell, J. K., Soga, K., et al. (2005). *Fundamentals of soil behavior*, Vol. 3. John
499 Wiley & Sons Hoboken, NJ.
- 500 Mohamed, A.-M. and El Gamal, M. (2012). “Treatment of collapsible soils using sulfur
501 cement.” *International Journal of Geotechnical Engineering*, 6(1), 65–77.
- 502 Neville, A. (2004). “The confused world of sulfate attack on concrete.” *Cement and*
503 *Concrete research*, 34(8), 1275–1296.
- 504 Nichols, S. C. and Goodings, D. J. (2000). “Physical model testing of compaction
505 grouting in cohesionless soil.” *Journal of Geotechnical and Geoenvironmental En-*
506 *gineering*, 126(9), 848–852.
- 507 Pereira, J. H. and Fredlund, D. G. (2000). “Volume change behavior of collapsible
508 compacted gneiss soil.” *Journal of geotechnical and geoenvironmental engineering*,
509 126(10), 907–916.
- 510 Popescu, M. E. (1986). “A comparison between the behaviour of swelling and of
511 collapsing soils.” *Engineering Geology*, 23(2), 145–163.
- 512 Sarkar, S. L. and Wheeler, J. (2001). “Important properties of an ultrafine cement—part
513 i.” *Cement and Concrete Research*, 31(1), 119–123.
- 514 Schmidt, T., Lothenbach, B., Romer, M., Neuenschwander, J., and Scrivener, K. (2009).
515 “Physical and microstructural aspects of sulfate attack on ordinary and limestone
516 blended portland cements.” *Cement and Concrete Research*, 39(12), 1111–1121.
- 517 Seiphoori, A., Ferrari, A., and Laloui, L. (2014). “Water retention behaviour and
518 microstructural evolution of mx-80 bentonite during wetting and drying cycles.”
519 *Géotechnique*, 64(9), 721–734.
- 520 Seiphoori, A., Ma, X.-g., Arratia, P. E., and Jerolmack, D. J. (2020). “Formation of

- 521 stable aggregates by fluid-assembled solid bridges.” *Proceedings of the National*
522 *Academy of Sciences*, 117(7), 3375–3381.
- 523 Semmler, M., Rička, J., and Borkovec, M. (2000). “Diffusional deposition of colloidal
524 particles: electrostatic interaction and size polydispersity effects.” *Colloids and*
525 *Surfaces A: Physicochemical and Engineering Aspects*, 165(1-3), 79–93.
- 526 Smalley, I. and Vita-Finzi, C. (1968). “The formation of fine particles in sandy deserts
527 and the nature of” desert’loess.” *Journal of Sedimentary Research*, 38(3), 766–774.
- 528 Taha, M. R. and Taha, O. M. E. (2012). “Influence of nano-material on the expansive
529 and shrinkage soil behavior.” *Journal of Nanoparticle Research*, 14(10), 1190.
- 530 Waltham, T. (2002). *Foundations of engineering geology*. CRC Press.

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Table 1. Physical and geomechanical properties of untreated collapsible soil

e_0	$\gamma_d(\frac{kN}{m^3})$	w_0	w_l	w_p	PI	G_s	$K(m/s)$
0.95±0.03	13.9±0.16	4.67±0.47	22.33±2.05	19.67±1.24	3.66±0.94	2.69±0.01	$(7.7±0.2) \times 10^{-6}$

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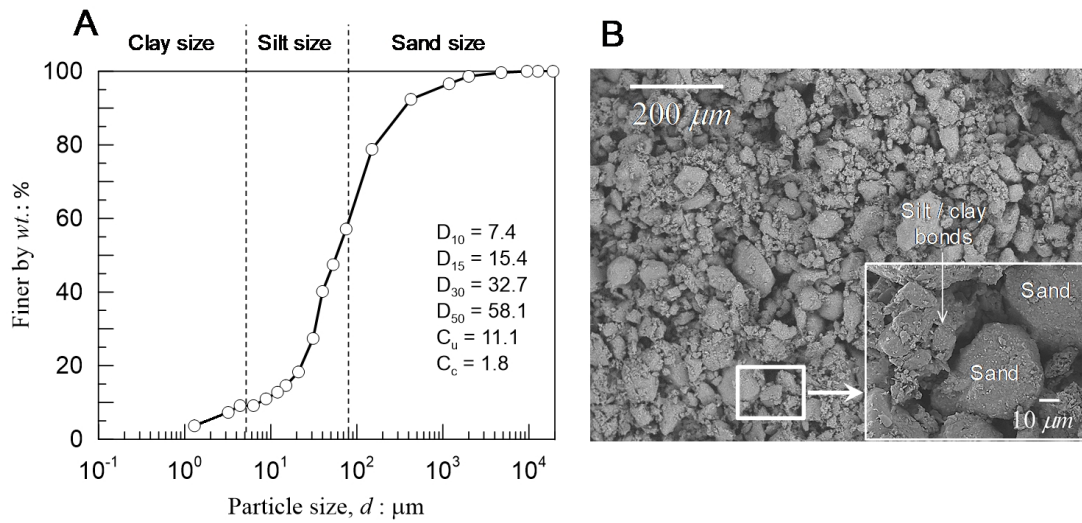


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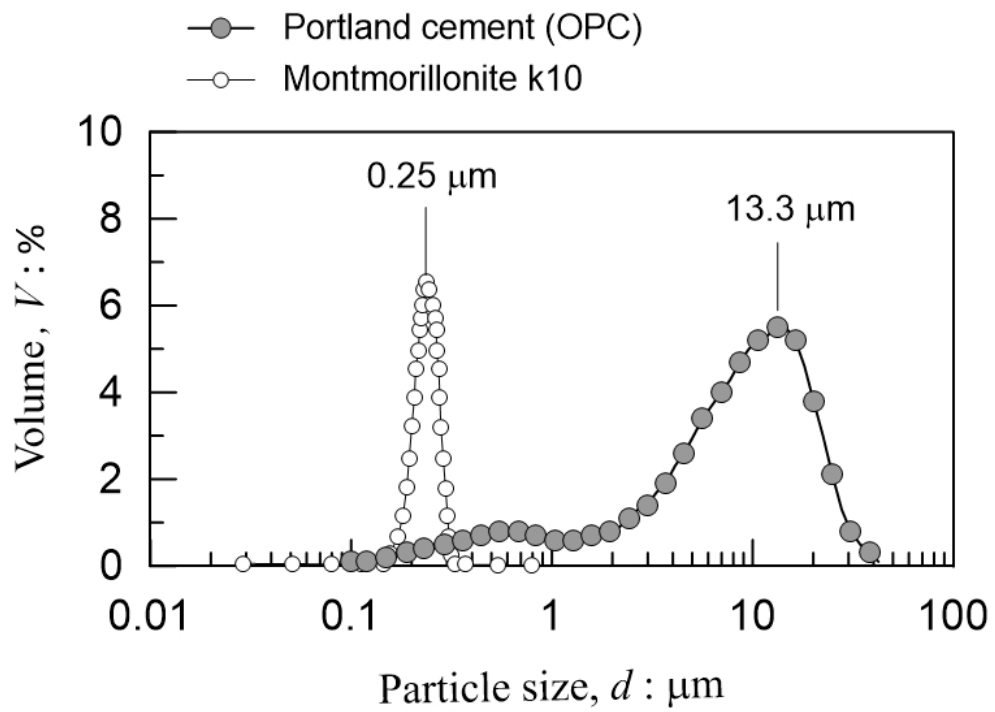


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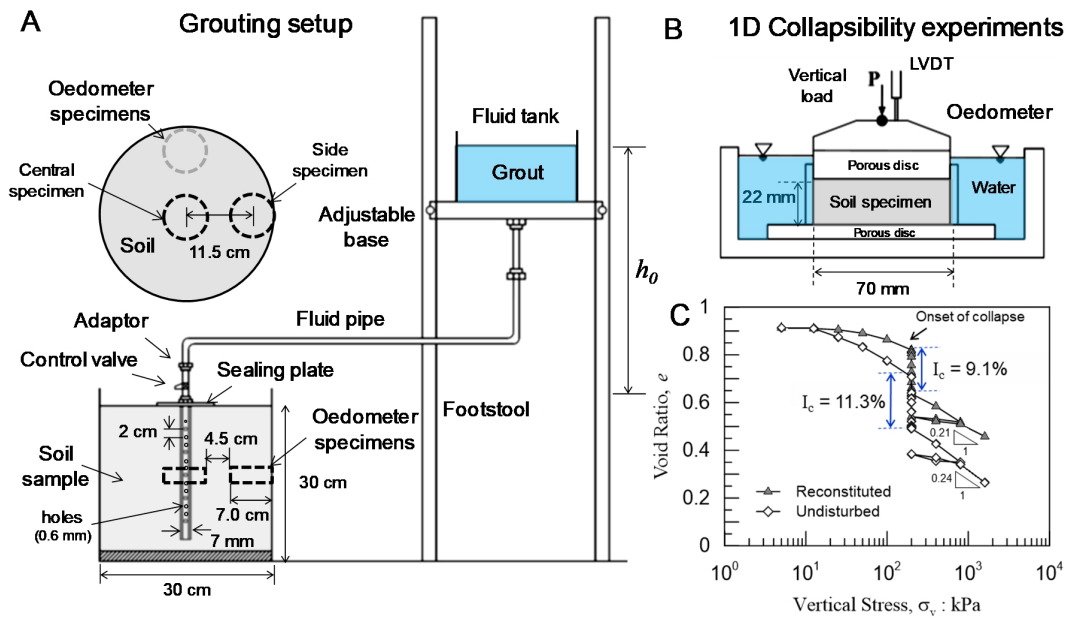


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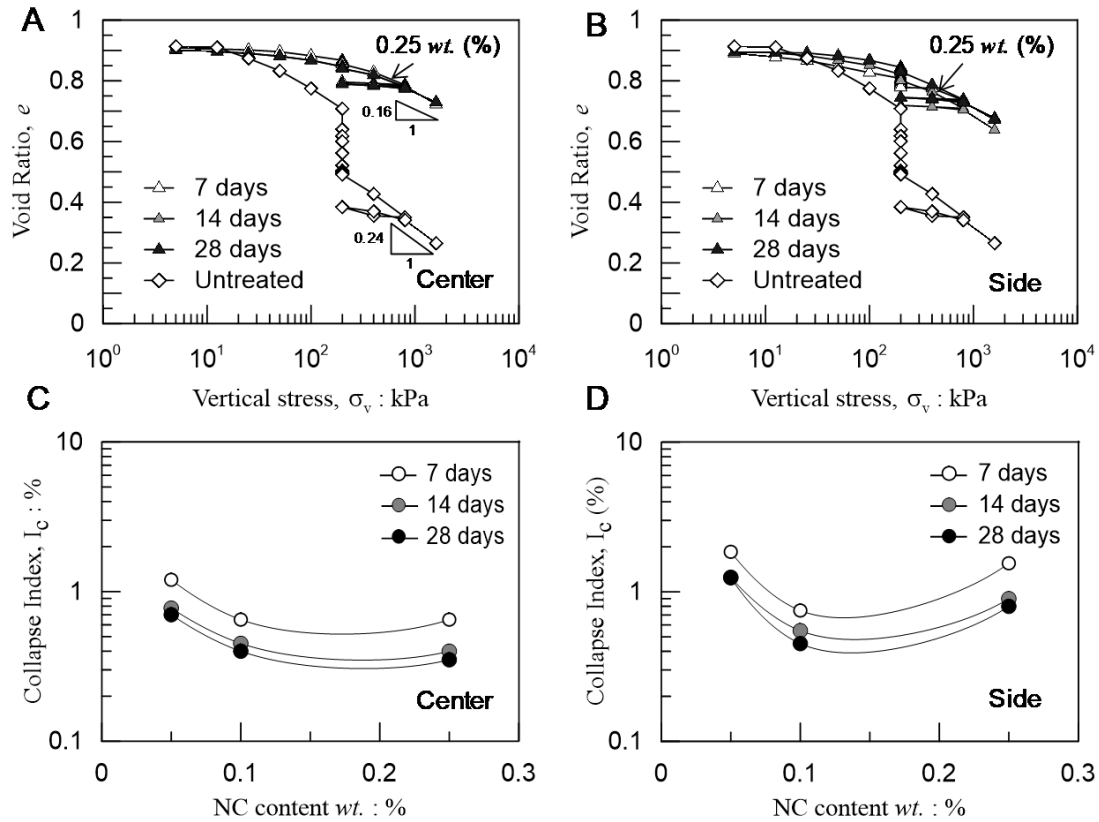


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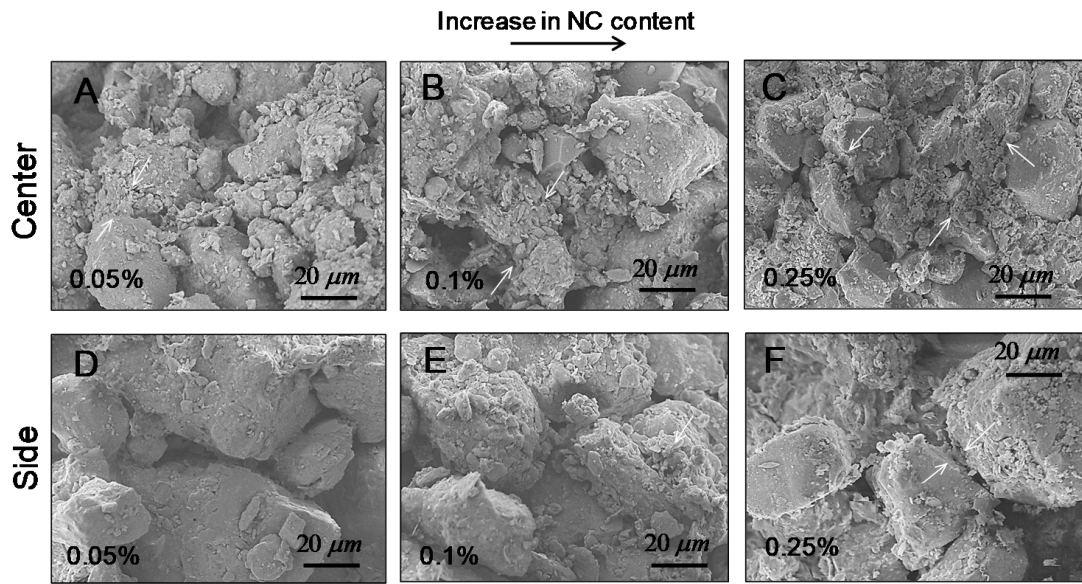


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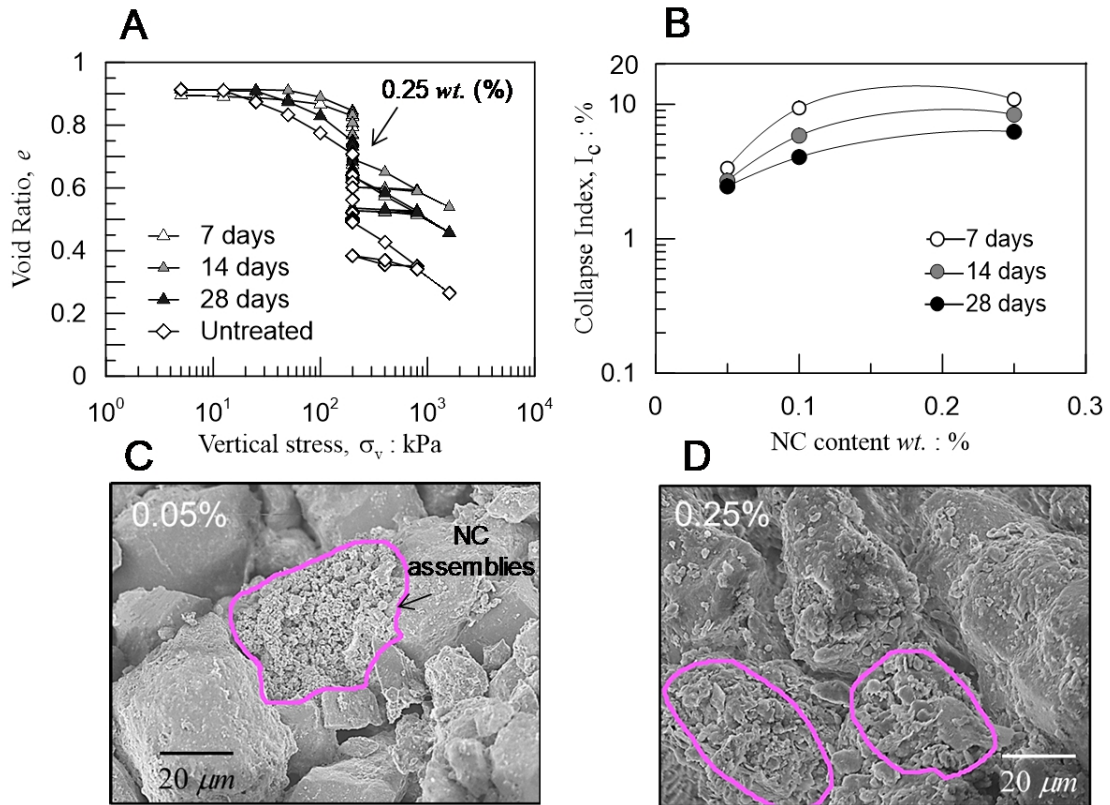


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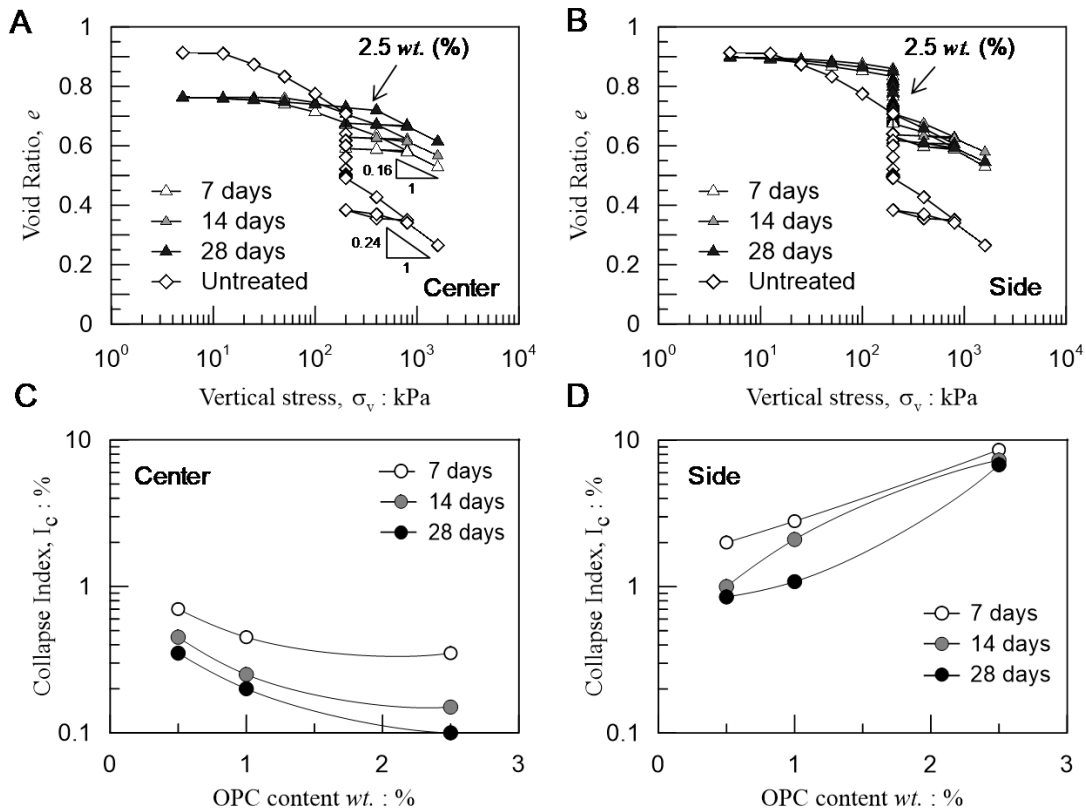


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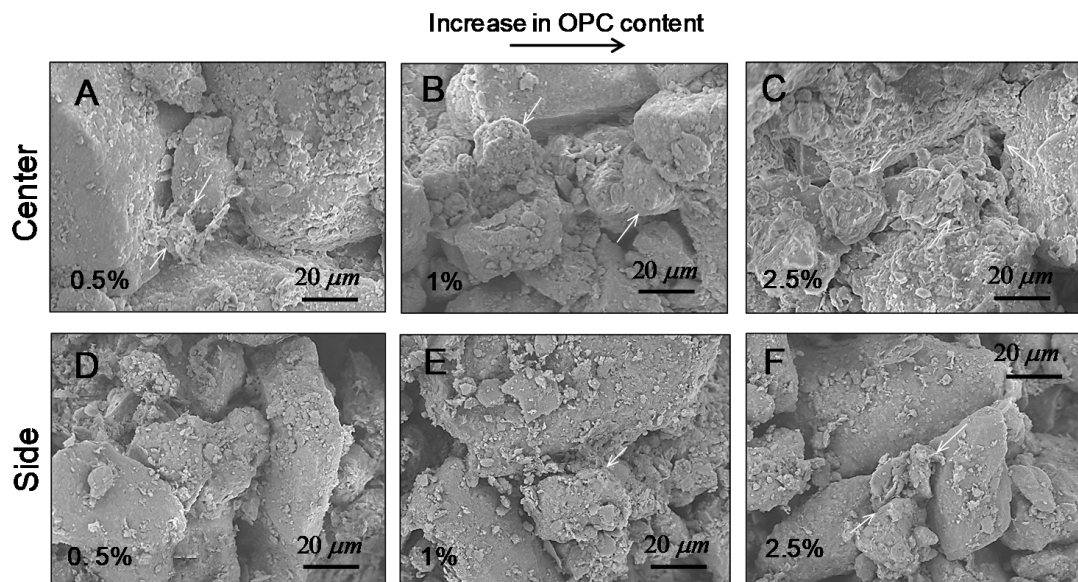


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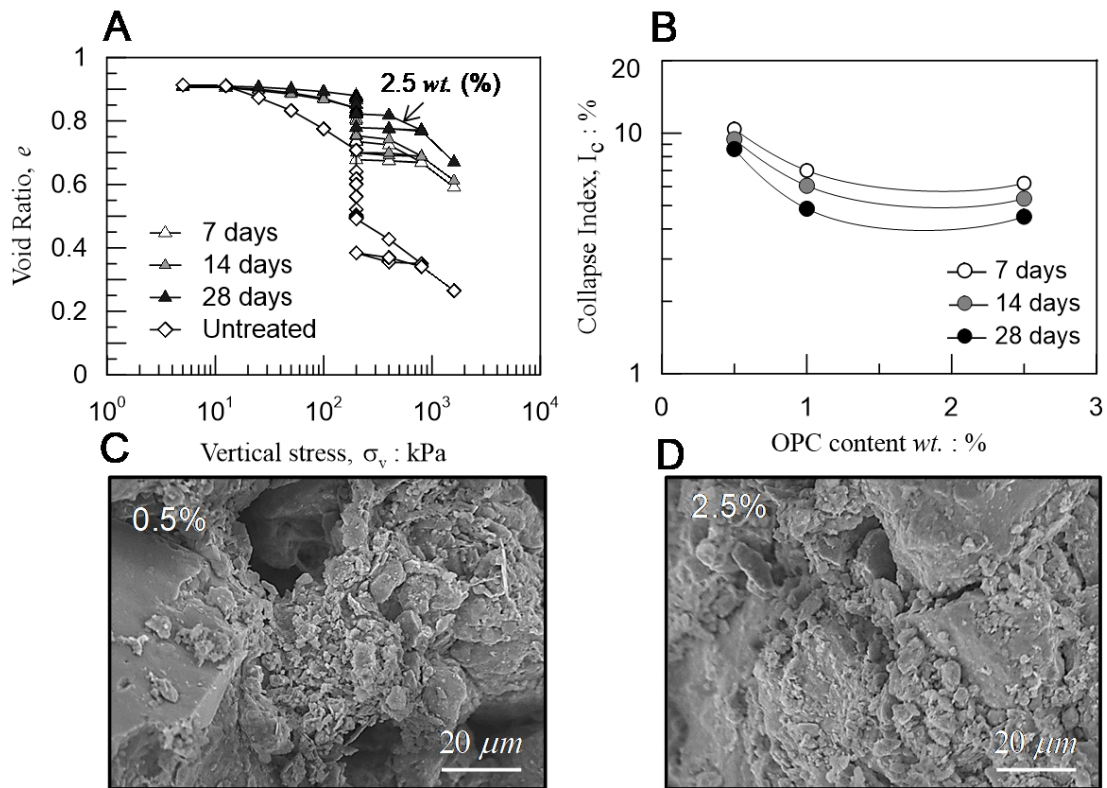


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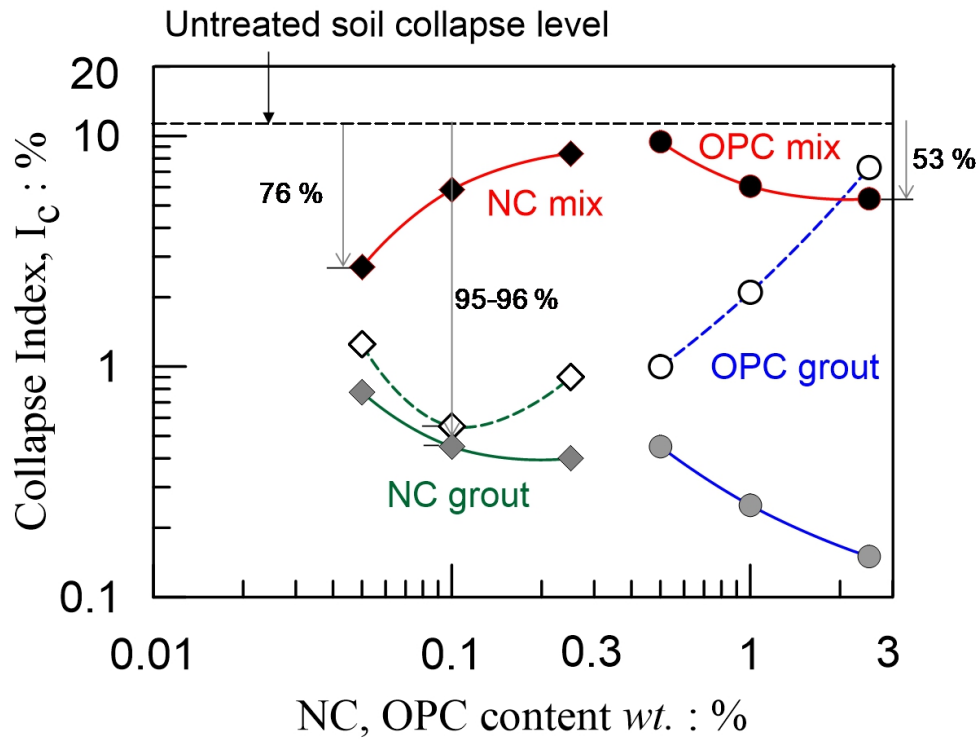


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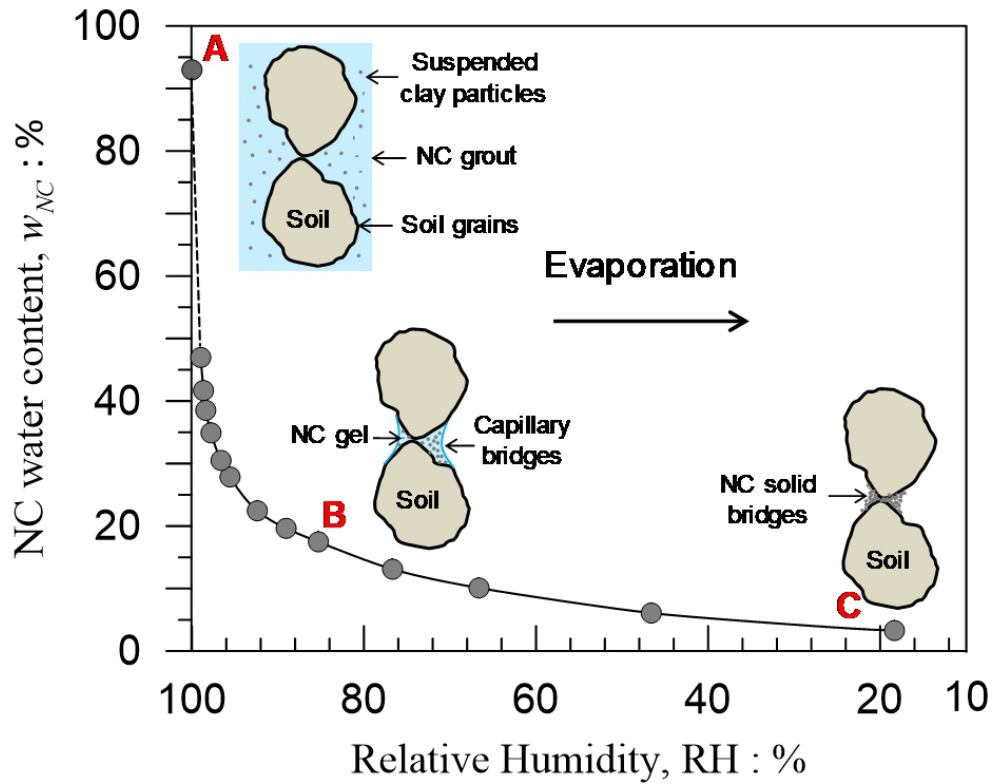


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