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Volume change behaviour of swelling and non-swelling clays upon inundation with water and a low dielectric constant fluid

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1   **Volume change behaviour of swelling and non-swelling clays upon inundation with**

2   **water and a low dielectric constant fluid**

3   **Tom Schanz [†], Snehasis Tripathy, and Asuri Sridharan**

4

5   **Abstract:** Studies of the volume change behaviour of saturated and unsaturated fine-grained

6   soils upon exposure to various types of fluid are of significant interest while dealing with

7   contaminated soils. In this study, the one-dimensional volume change behaviour of two clays

8   (Spergau kaolin and Calcigel bentonite) with dominant minerals as kaolinite and

9   montmorillonite was studied by inundating several initially unsaturated and saturated clay

10   specimens with water and a non-polar organic liquid (heptane). For both cases, applied

11   vertical pressures of 25 and 100 kPa were considered under oedometric conditions. The

12   magnitude of compression and swelling deformations of the clays were found to be strongly

13   dependent upon the mineralogy of clays, dielectric properties of the pore-fluid, initial

14   compaction conditions, and applied stress during the wetting process. The test results

15   indicated distinct interactions between unsaturated clays and water. Specimens of Spergau

16   kaolin exhibited compression with water, whereas specimens of Calcigel bentonite exhibited

17   swelling. Compression and swelling deformations of the clays with heptane as the inundating

18   liquid remained within about ± 1.0% for both clays indicating a very minor interaction

19   between unsaturated clays and molecules of this non-polar organic liquid. The test results

20   emphasized the significance of attractive and repulsive forces and their impact on the volume

21   change behaviour of clays of different mineralogy.

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25   † Deceased 12 October 2017

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1. Introduction

Studies of the volume change behaviour of fine-grained soils upon exposure to various types of fluid are of great interest, particularly while dealing with contaminated soils. Contamination of soils in vadose zone may occur due to industrial activity, agricultural chemicals, gasoline spillage, and improper disposal of waste. Similarly, mitigation strategies adopted for soils contaminated with various organic contaminants demands understanding of the soil-contaminant interaction at various stress states of soils.

The physico-chemical interaction between soil particles and their influence on the volume change behaviour of fine grained soils depends upon the clay mineralogy and the pore fluid characteristics (e.g., Sridharan and Rao, 1973; Lagaly 1989; Mitchell and Soga 2005). Organic fluids, chemicals, and petroleum products usually possess dielectric constant values far lower than that of water. A review of the literature suggests that the influence of organic fluids on the plasticity properties, compressibility, shear strength, and hydraulic conductivity of saturated clays has been studied in detail by several researchers in the past (Mesri and Olson 1971; Sridharan and Rao 1973; Sridharan and Rao 1979; Fernandez and Quigley 1985; Lagaly 1989; Shang *et al.* 1994; Kaya and de Brito Galvao 1998; Kaya and Fang 2005). However, studies concerning the volume change behaviour of compacted unsaturated clays upon exposure to organic fluids, particularly in the context of the influence of mineralogy, compaction conditions, and applied external stress are scarce in the literature. In this study, laboratory one-dimensional swell or compression tests were carried out on two clays with two types of fluid and at two different stress levels for studying the impacts of polar and non-polar fluids, the magnitude of an applied stress, and initial compaction conditions on the volume change behaviour of clays.

53 **2. Background**

54

55   During the wetting process and under a constant applied stress, the volume change of

56 unsaturated fine-grained soils is accompanied by a decrease in the shearing resistance at

57 inter-particle level brought about by a reduction in the soil suction, and by the changes in the

58 magnitude of the physico-chemical forces. As a result, a soil may exhibit swelling or

59 compression, or even the volume change may not occur under specific stress and hydraulic

60 boundary conditions. The physico-chemical forces are comprised of the electrical attractive

61 and the repulsive forces (Verwey and Overbeek 1948; Bolt 1956; van Olphen 1977;

62 Sridharan and Jayadeva 1982; Heuckel 1992; Mitchell and Soga 2005). The repulsive forces

63 that may get generated within clay-fluid systems are due to the interaction of electrical double

64 layers around the clay particles and hydration of ions and surfaces of clay particles (Madsen

65 and Müller-Vonmoos 1989; Laird 2006; Baille et al. 2010). These forces tend to cause an

66 increase in the volume of clays. The interaction between the edges of clay particles with the

67 negatively charged surfaces are electrostatic (Columbic) in nature. The primary and

68 secondary bonding (hydrogen bond and van der Waals bond) are the other possible sources of

69 attractive force that govern the shear resistance at inter-particle level. The short range inter-

70 particle force exists at very close spacing, whereas the long-range attractive forces (van der

71 Waals force) are considered to be sensitive to applied force and pore fluid properties.

72

73   The mobilised physico-chemical forces within clay systems to a great extent depend

74 upon the physical and chemical properties of the clays (i.e., specific surface area and type and

75 amount of exchangeable cations present in the clay), the bulk fluid properties (i.e., solvent

76 type, concentration, dielectric properties, and pH), the applied external stress, the soil suction,

77 the initial compaction conditions of the clay (i.e., dry density and water content), and the

78   temperature (Sridharan and Jayadeva 1982). The physico-chemical forces influence the fabric

79   in fine-grained soils, which in turn regulates the hydraulic conductivity, compressibility,

80   stiffness, and shear strength (Sridharan, 1991, Sridharan and Rao 1979; Fernandez and

81   Quigley 1985; Chen et al. 2000; Santamarina et al. 2001; Mitchell and Soga 2005; Wang and

82   Siu 2006). The fine-grained soil system is complex and the individual effects of various

83   parameters influencing the repulsive and attractive pressures cannot be readily separated.

84   However, it has been brought out in the literature that the dielectric constant of the pore

85   medium plays an important role in that, the attractive force varies inversely and the repulsive

86   force varies directly with the dielectric constant of the pore medium (Verwey and Overbeek

87   1948; van Olphen 1977; and Sridharan and Rao 1973 and 1979).

88

89   Adsorption of water molecules in the silicate layers of clay minerals tends to

90   eliminate the attractive pressure at the edge-face contacts in flocculated clay structure.

91   Development of the repulsive pressure during the wetting process causes further decrease in

92   the attractive pressure between the clay particles. A decrease in the attractive pressure gets

93   manifested on a reduction in the shearing resistance due to a reduction in effective contact

94   stress. Both the mobilized repulsive and attractive pressures are strongly dependent upon the

95   characteristics of the hydrating fluid. Partially saturated and fully saturated clay systems

96   possess attractive pressures lower than that occur in dry clays due to a partial or completion

97   of the surface and ion hydration processes (Sridharan 1968).

98

99   The exchangeable cations in clay are soluble in polar fluids (e.g., water and polar

100   organic liquids); however, are insoluble in non-polar organic liquids (e.g., heptane, toluene

101   etc.). The adsorption of polar organic liquid molecules in the interlayers occurs either as

102   replacement to water molecules in partially saturated clay systems or via the exchangeable

103 cations in fully dry clays (Grim 1968; Theng 1974; and van Olphen 1977). Sorption of non-

104 polar organic molecules in the interlayers of dry clays is usually insignificant, whereas the

105 process can be very slow or only limited adsorption may take place in the interlayers of

106 partially saturated clays (Grim 1968; Theng 1974; and Amarashinghe et al. 2009).

107

108 **3. Materials and methods**

109

110 Spergau kaolin and Calcigel benonite, both clays from Germany, were used in this

111 investigation. The properties of the clays are given in Table 1. The chosen clays possess a

112 wide range of physical and chemical properties. X-ray diffraction studies indicated that about

113 60% of the minerals in Calcigel bentonite were montmorillonite, whereas the remaining were

114 quartz, feldspar, dolomite, and calcite. Spergau kaolin was found to contain about 72%

115 kaolinite, 16% quartz, and 10% muscovite. Studies of the cation exchange complexes of the

116 clays indicated that more than 85% of the exchangeable cations in Calcigel bentonite were

117 Ca^{+2} and Mg^{+2} with small fractions of Na^+ and other ions (Schanz and Tripathy 2009).

118 Spergau kaolin was found to contain Ca^{+2}, Mg^{+2}, and K^+.

119

120 One-dimensional swell and compression tests (ASTM 1986) were carried out in

121 conventional oedometers. All tests were carried out at a temperature of 20 ± 2 °C. Compacted

122 clay specimens were prepared at various water contents including from oven-dried clay

123 powder. Distilled water was used for preparing the clay-water mixtures. The clay-water

124 mixtures were stored in air-tight plastic bags for about a week in the laboratory for moisture

125 equilibration to take place. Compacted clay specimens were prepared corresponding to

126 predetermined dry densities with gentle tamping on the clay-water mixtures within the

127 oedometer rings. Extreme precautions were exercised in order to minimize the errors due to

128   side friction by lubricating the specimen rings with technical grade silicon grease. Filter

129   papers were used at the top and bottom of the specimens.

130

131   The details of the specimens tested for Spergau kaolin and Calcigel bentonite are

132   given in Tables 2 and 3, respectively. For both clays and for each combination of water

133   content-dry density, two different vertical pressures levels were considered, such as 25 and

134   100 kPa. For the clay specimens that were prepared from oven dried clay powder and at low

135   initial water contents (i.e., 0 and 12% for Spergau kaolin and 0 and 9.0% for Calcigel

136   bentonite), the targeted external loads were applied in a single step loading process, whereas

137   for the specimens that were prepared at higher water contents (61.5% for Spergau kaolin and

138   55.2 and 64.3% for Calcigel bentonite), a load increment ratio of one was adopted to attain

139   the required vertical pressure levels. The incremental loads were applied after an elapsed time

140   of more than three hours or upon completion of the primary consolidation phase, which ever

141   occurred earlier. Following the loading process, the specimens were inundated either with

142   water or heptane (C_7H_{16}). For every applied vertical stress, time-vertical deformation

143   readings were monitored using dial gauges of sensitivity of 0.001 mm and a total run of 25

144   mm. Similarly, the time-deformation readings (compression or swelling) were monitored

145   until the equilibrium conditions were attained. At each compaction conditions (see Tables 2

146   and 3), four specimens were tested. In total, thirty-six clay specimens were tested, sixteen for

147   Spergau kaolin and twenty for Calcigel bentonite.

148

149   The dielectric constant of water is 80.4, whereas that of heptane is 1.92 as against the

150   dielectric constant of air of 1.0. Additionally, water is a polar liquid, whereas heptane is a

151   non-polar organic liquid. Heptane is a highly flammable solvent. Extreme care was exercised

152   while handling the solvent by wearing gloves and a medical mask. The oedometer tests were

153 carried out in an isolated room with appropriate ventilation system. Similarly, appropriate

154 disposal methods were undertaken after completion of the laboratory tests.

155

156   Prior to testing for the clay specimens, the pressure-deformation characteristics of the

157 oedometers used were studied using a steel dummy following the procedure suggested by

158 ASTM (1998). Based on the pressure-deformation characteristics of the oedometers used, the

159 measured heights of the specimens at each loading steps were corrected for calculating the

160 actual vertical deformations. The vertical deformation at equilibrium under any given applied

161 vertical pressure is defined as the ratio of the change in height of a specimen upon either

162 swelling or compression to the height of the specimens prior to inundation and is expressed

163 as a percentage. The void ratio changes during swelling or compression of the specimens

164 were calculated based on height of solids method.

165

166 **4. Presentation of experimental results**

167

168 4.1 Volume change behaviour of Spergau kaolin

169

170   The elapsed time versus void ratio plots for Spergau kaolin specimens at four

171 different initial placement conditions, two different applied vertical pressures (i.e., 25 and

172 100 kPa), and with water and heptane as the inundating fluids are shown in Figs. 1 and 2. The

173 vertical deformations of the specimens at equilibrium are shown in Table 2.

174

175   Following inundation of the specimens either with water or heptane, except two

176 specimens that exhibited a small volume increase upon inundated with heptane (water

177 contents of 0 and 23.3%, dry density 1.11 Mg/m^3), all other specimens exhibited compression

178 (Table 2, Figs. 1 and 2). The compression deformation of the specimens occurred within

179 about 15 to 25 minutes following the inundation process in all cases.

180

181   The test results of the initially unsaturated specimens showed three distinct trends in

182 terms of the volume change (Table 2, Figs. 1a, 1b and 2a), such as (*i*) at any applied vertical

183 pressure (25 or 100 kPa), the compression deformation was significantly smaller for the

184 specimens that were inundated with heptane than that occurred with water, (*ii*) the vertical

185 deformation decreased with an increase in the initial compaction dry density and with an

186 increase in the water content for both inundating fluid types, and (*iii*) the vertical deformation

187 increased with an increase in the applied stress for both fluid types.

188

189   For the saturated clay specimens with an initial water content of 61.5% (Fig. 2b), the

190 vertical deformations at any given applied pressure and with both water and heptane as the

191 inundating fluids were very nearly similar clearly indicating that the pore-fluid type has

192 limited impact on the volume change behaviour of saturated kaolinites.

193

194 4.2 Volume change behaviour of Calcigel bentonite

195

196   The elapsed time versus void ratio plots for the specimens of Calcigel bentonite at

197 five different initial placement conditions, two different applied vertical pressures (i.e., 25

198 and 100 kPa), and with water and heptane as the inundating fluids are shown in Figs. 3, 4,

199 and 5. The initial compaction conditions of the specimens, the inundation fluids used, and the

200 applied vertical pressures are shown in Figs. 3 to 5. The vertical deformations of the

201 specimens at equilibrium are given in Table 3.

202

203   Following inundation either with water or heptane, except four specimens with high

204   initial void ratios of 1.94 and 1.8 that underwent slight compression at an applied stress of

205   100 kPa, all other specimens (Table 3) exhibited swelling. The test results of the initially

206   unsaturated specimens showed three distinct trends in terms of the volume change (Figs. 3 to

207   5 and Table 3), such as (*i*) the swelling deformation was significantly greater for the

208   specimens that were inundated with water as against their counterparts that were inundated

209   with heptane, (*ii*) with an increase in the applied stress during inundation with any fluid, the

210   deformation behaviour of the specimens was accompanied by either a decrease in the

211   swelling deformation or an increase in the compression deformation, or even specimens that

212   exhibited swelling at 25 kPa, underwent compression at 100 kPa, (*iii*) for the specimens with

213   an initial water content of 0%, an increase in the compaction dry density caused an increase

214   in swelling deformation, whereas swelling deformation decreased with an increase in the

215   initial water content for the same compaction dry density (see results for dry density of 1.10

216   Mg/m^3 in Table 3).

217

218   The test results presented in Figs. 3, 4, and 5 clearly indicated that unlike the

219   deformation behaviour of the unsaturated specimens of Spergau kaolin, in which case the

220   compression deformation invariably increased with an increase in applied vertical pressure

221   and for both inundation fluids used (Figs. 1 and 2, Table 2), the deformation behaviour was

222   found to be reversed in the case of Calcigel bentonite. In the latter case, the swelling

223   deformation decreased with an increase in the vertical pressure. Additionally, the influence of

224   fluid type and applied vertical pressure was found to be quite significant on the volume

225   change behaviour of the specimens that were initially saturated (Fig. 5 and Table 3). This was

226   not evident in the case of saturated Spergau kaolin specimens (Figs. 2*b* and Table 2).

227

5. Influence of physico-chemical forces on the volume change of clays

228

229

230   Table 4 summarises the influence of a decrease in the dielectric constant of the

231 inundating fluid, an increase in the applied stress during the inundation process, an increase

232 in the compaction water content, and an increase in the compaction dry density on the vertical

233 deformation of clay specimens observed in this study. The possible reasons for the volume

234 change of clays are presented in Table 4.

235   Further, the results can also be explained by the changes in the effective stress that has

236 been brought out by changes in normal stress, repulsive and attractive forces. It has been now

237 widely accepted that Terzaghi's concept of effective stress provides a satisfactory basis for

238 understanding the strength and deformation characteristics of saturated soils, which can be

239 stated as:

240

241 $$\sigma^{'} = \sigma - u \qquad\qquad (1)$$

242

243 where σ' = effective stress, σ = applied external stress or the total stress and u = pore water

244 pressure.

245

246   It may be noted that σ' is the contact stress at mineral to mineral contact zone, which

247 is also called inter-granular stress. While one can discuss at greater length the nature of this

248 contact, for purpose of brevity, it can be said that the role of contact is to transfer the stress. It

249 has been brought out earlier that both electrical attractive and repulsive forces exist between

250 clay particles. Since the fine grained soils are normally composed of clays, the existence of

251 attractive and repulsive forces in the soil-water system is inevitable. The studies of Sridharan

252   (1968) and Sridharan and Rao (1973, 1979) resulted in the proposition of modified effective

253   stress concept as given in eq. (2).

254

255 $$\bar{c} = \bar{\bar{\sigma}}a_m = \sigma - u - R + A \qquad (2)$$

256

257   For a saturated system, where $\bar{c}$ is the average contact stress, $\bar{\bar{\sigma}}$ is actual contact stress at

258   mineral - to - mineral level, a_m is area fraction over which $\bar{\bar{\sigma}}$ acts or percentage area through

259   which $\bar{\bar{\sigma}}$ acts (non-dimensional), σ = the external applied stress, u = pore water pressure, R =

260   average repulsive pressure acting throughout the area, A = average attractive pressure acting

261   throughout the area.

262

263   Equation (2) can be written as

264

265 $$\bar{c} = \bar{\bar{\sigma}}a_m = \sigma' + \sigma'' \qquad (3)$$

266

267   where σ" is the intrinsic effective stress and σ' is the conventional effective stress.

268

269   It may further be stated that the effect of σ and A is to bring the particles closer to each

270   other. The effect of positive pore water pressure and R is to keep the particles away from each

271   other. If the pore water pressure is negative (i.e., capillary pressure operative in partly

272   saturated soils), its role is to bring the particles closer to each other. The average contact

273   stress or the intergranular stress ($\bar{c}$) between particles, is defined as the modified effective

274   stress and it is hypothesised that $\bar{c}$ is the stress controlling the shearing resistance and

275   volume changes that take place in soil-water system. In fine-grained soils / clayey soils, the

276   attractive and repulsive forces cannot be neglected, especially when the water content and the

277   soil plasticity are high. Since the clay-water system is complex, quantitative determination of

278   *R* and *A* becomes difficult for real systems. However, qualitative evaluation could be done.

279   The validity of eqns. (2) and (3) have been qualitatively studied extensively considering the

280   volume change behaviour (Sridharan and Rao 1973; Sridharan et al. 1973; Sridharan 2003:),

281   the strength behaviour (Sridharan et al. 1971; Sridharan and Rao 1979; Allam and Sridharan

282   1981; Sridharan et al. 1983; Sridharan and Prakash, 1999b), the shrinkage phenomena

283   (Sridharan and Rao 1971), the secondary compression behaviour (Sridharan and Rao 1982)

284   and the sediment formation (Sridharan and Prakash 1999a).

285

286   The thickness of the electrical double layer that is formed around the clay particles

287   are influenced by several factors (Mitchell and Soga 2005), such as the characteristics of the

288   hydrating fluid, the type and amount of exchangeable cation present in the clay, the specific

289   surface area, the applied external stress, and the temperature (Verwey and Overbeek 1948;

290   van Olphen 1977; Sridharan and Rao 1979; and Sridharan and Jayadeva 1982). The thickness

291   of the electrical double layer controls the swelling, compressibility, shear strength, and

292   hydraulic conductivity in clays. The factors that cause an increase in the thickness of

293   electrical double layer are also responsible for a higher magnitude of swelling deformation in

294   expansive clays resulting in a decrease in the modified effective stress. Similarly, the factors

295   that cause a decrease in the thickness of electrical double layer aid in an increasing the

296   modified effective stress and increasing the shearing resistance and cause a decrease in the

297   compressibility and the hydraulic conductivity.

298

299   The attractive force varies inversely and the repulsive force varies directly with the

300   dielectric constant of the pore fluid (Sridharan and Rao 1973 and 1979). Therefore, a

301   decrease in the dielectric constant of the pore fluid caused two opposite effects for both clays

302   in this study when the inundating fluid was heptane, such as an increase in the attractive

303   pressure and a decrease in the repulsive pressure and thus an increase in the modified

304   effective stress. A greater mobilised attractive pressure with heptane as the inundating fluid

305   caused an increase in the modified effective stress and two distinct effects, such as that (*i*) the

306   interparticle shearing resistance did not decrease as that occurred with water as the inundating

307   fluid thereby causing a lesser compression of the specimens of Spergau kaolin and (*ii*) since

308   the electrical double layer thickness was reduced in case of Calcigel bentonite, the clay

309   exhibited a lesser swelling as compared to that occurred with water as the inundating fluid.

310   The magnitude of applied stress on the clays can be considered to have two effects, such as

311   an increase in the applied stress acts against the interparticle shearing resistance to cause a

312   greater deformation in case of Spergau kaolin and it acts as a restraint against the expansion

313   of the electrical double layer thereby causing a reduction in the swelling deformation in case

314   of Calcigel bentonite.

315

316   An increase in the initial compaction water content and its influence on the

317   deformation behaviour of the clays can be considered separately for the two inundating fluids

318   considered in this study (i.e., water and heptane). An increase in the compaction water

319   content causes a reduction in the inter-particle shearing resistance since the attractive pressure

320   is inversely proportional to the dielectric constant of the pore fluid. Additionally, an increase

321   in the compaction water content causes the hydration of the surfaces and the ions. A further

322   increase in the compaction water content above that is required for surface and ion hydration

323   aids in the formation of the electrical double layer. The increased volume of the clay due to

324   the expansion of the electrical double layer depends upon the volume of water available and

325   the type of clay mineral present. With the inundating fluid as water, the changes in the

326   vertical deformation in case Spergau kaolin was found to be minor since the pore fluid

327 remained unchanged. The swelling deformation in the case of Calcigel bentonite also reduced

328 since a part of the swelling process was already completed during the specimen preparation

329 stage. The clay specimens of Calcigel bentonite exhibited further swelling primarily due to

330 the available of water which caused a further expansion of the electrical double layer.

331

332   Replacement of water by heptane can be considered when the inundating fluid used

333 was heptane. This would tend to cause an increase in the inter-particle shearing resistance

334 primarily due to an increase in the attractive pressure and a decrease in thickness of the

335 electrical double layer. A replacement of molecules of water by heptane for kaolinite is

336 expected to cause no significant change in terms of the deformation, but due to a change in

337 the magnitude of the attractive pressure the clay may tend to flocculate causing an increase in

338 the volume (see test results for specimens at water content = 12% in Table 2). A decrease in

339 the thickness of electrical double layer in case of Calcigel bentonite reduced the swelling

340 deformation of the clay. Saturated specimens exhibited slight swelling deformation which

341 indicated that replacement of water with heptane will increase the electrical attractive forces

342 resulting in a relatively flocculent fabric which tend to get eliminated as the applied stress

343 increased (see test results of initially saturated specimens in Table 3).

344

345   An increase in the compaction dry density is associated with an increase in the applied

346 stress that overcomes the inter-particle shearing resistance and tends to produce a more

347 oriented fabric perpendicular to the direction of the applied load. A decrease in the void ratio

348 brings the clay particles closer towards each other which in turn, causes an increase in the

349 attractive pressure. Ignoring the orientation of clay particles and the difference in the

350 magnitude of the attractive pressure, under any given applied stress, clay specimens with

351 different initial compaction dry densities but with same initial water content may exhibit

352 different vertical deformations, but should attain the same void ratio upon saturation. A

353 decrease in the compression deformation in case of Spergau kaolin is attributed due to an

354 increase in the attractive pressure, whereas an increase in the swelling deformation due to an

355 increase in the compaction dry density in the case of Calcigel bentonite is attributed due to

356 expansion of the electrical double layer in response to the applied stress during the inundation

357 process.

358

359 Thus it can be seen that the volume change (compression or swelling) behaviour of

360 both the clays in this study can be explained by the changes in the intrinsic effective stress,

361 defined by equations (2) and (3), brought out by the changes in the electrical attractive and

362 repulsive pressures (influenced by the fluid types by their di-electric constant values), the

363 negative pore pressures and the externally applied stress.

364

365 **6. Conclusions**

366

367 The one-dimensional volume change behaviour of kaolinite and montmorillonite-rich

368 clays is presented in this paper. Several initially unsaturated and saturated clay specimens

369 were inundated either with water or heptane at applied vertical pressures of 25 and 100 kPa.

370

371 The test results clearly showed that the physico-chemical forces within the clay-fluid

372 systems are significantly influenced by the mineralogy of clays, the properties of the

373 hydrating and the inundating fluids, the compaction dry density and water content, and the

374 applied stress during the inundation process. The kaolinite-rich clay in this study exhibited

375 low compression with both water and heptane, whereas the montmorillonite clay exhibited

376 significant swelling (up to 23%) with water, whereas in the kaolinite clay exhibited swelling

377 up to about 0.9%. The changes in the volume of the clays were found to be much smaller

378 with heptane (attributed to strong electrical attractive forces) as compared that occurred with

379 water. The volume change behaviour of the clays are explained in the light of the modified

380 effective stress and the interplay of attractive and repulsive pressures.

381

387

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482   **Table 1** Properties of the clays used in the study

Properties	Spergau kaolin	Calcigel bentonite
Specific gravity of soil solids, G_s	2.62	2.80
Liquid limit, w_L (%)	53.4	178.0
Plastic limit, w_P (%)	30.1	56.0
Shrinkage limit, w_S (%)	22.3	8.8
Specific surface area, S (m^2/g)	14.0	650.0
Base exchange capacity, B (meq/100g)	8.0	74.0

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485   **Table 2** Details of Speregau kaolin specimens tested

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Initial compaction conditions				Inundation fluid	Vertical deformation (%) at applied vertical stress	
Water content (%)	Dry density (Mg/m^3)	Void ratio	Degree of saturation (%)		25 kPa	100 kPa
0	1.02	1.58	0	Water	-13.42	-17.63
0	1.02	1.58	0	Heptane	-0.23	-2.99
0	1.11	1.35	0	Water	-8.14	-11.87
0	1.11	1.35	0	Heptane	0.43	-0.13
12.0	1.11	1.35	23.3	Water	-7.57	-11.50
12.0	1.11	1.35	23.3	Heptane	0.34	-0.60
61.5	1.00	1.61	100	Water	-0.43	-0.47
61.5	1.00	1.61	100	Heptane	-0.44	-0.41

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489 **Table 3** Details of Calcigel bentonite specimens tested

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Initial compaction conditions				Inundation fluid	Vertical deformation (%) at applied vertical stress	
Water content (%)	Dry density (Mg/m^3)	Void ratio	Degree of saturation		25 kPa	100 kPa
0	0.95	1.94	0	Water	16.01	2.7
0	0.95	1.94	0	Heptane	-0.49	-0.55
0	1.10	1.54	0	Water	22.80	7.21
0	1.10	1.54	0	Heptane	0.91	0.37
9.0	1.10	1.54	16.4	Water	19.87	6.92
9.0	1.10	1.54	16.4	Heptane	0.83	0.28
55.2	1.10	1.54	100	Water	7.6	0.86
55.2	1.10	1.54	100	Heptane	0.16	0.00
64.3	1.00	1.80	100	Water	3.2	-0.70
64.3	1.00	1.80	100	Heptane	0.29	-0.22

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493 **Table 4** Influence of various factors on the vertical deformation of unsaturated clays upon

494 saturated under applied stress

Reference	Vertical deformation		Remarks
	Kaolinite	Montmorillonite	
Decrease in dielectric constant of inundating fluid	Compression decreases	Swelling decreases	Attractive pressure increases/Double layer thickness decreases
Increase in applied stress during inundation	Compression increases	Swelling decreases	Interparticle shearing resistance exceeded/Double layer expansion restricted
Increase in compaction water content	Compression decreases	Swelling decreases	Attractive pressure decreases/Surface and ion hydration processes and development of double layer
Increase in compaction dry density	Compression decreases	Swelling increases	Attractive pressure increases/Repulsive pressure increases

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497 **Figure captions**

498 **Fig. 1** Water content versus total suction plot for the clays used in this study

499 **Fig. 2** Time-deformation behaviour of Spergau kaolin specimens

500 **Fig. 3** Summary of vertical deformations of Spergau kaolin specimens

501 **Fig. 4** Time-deformation behaviour of Calcigel bentonite specimens

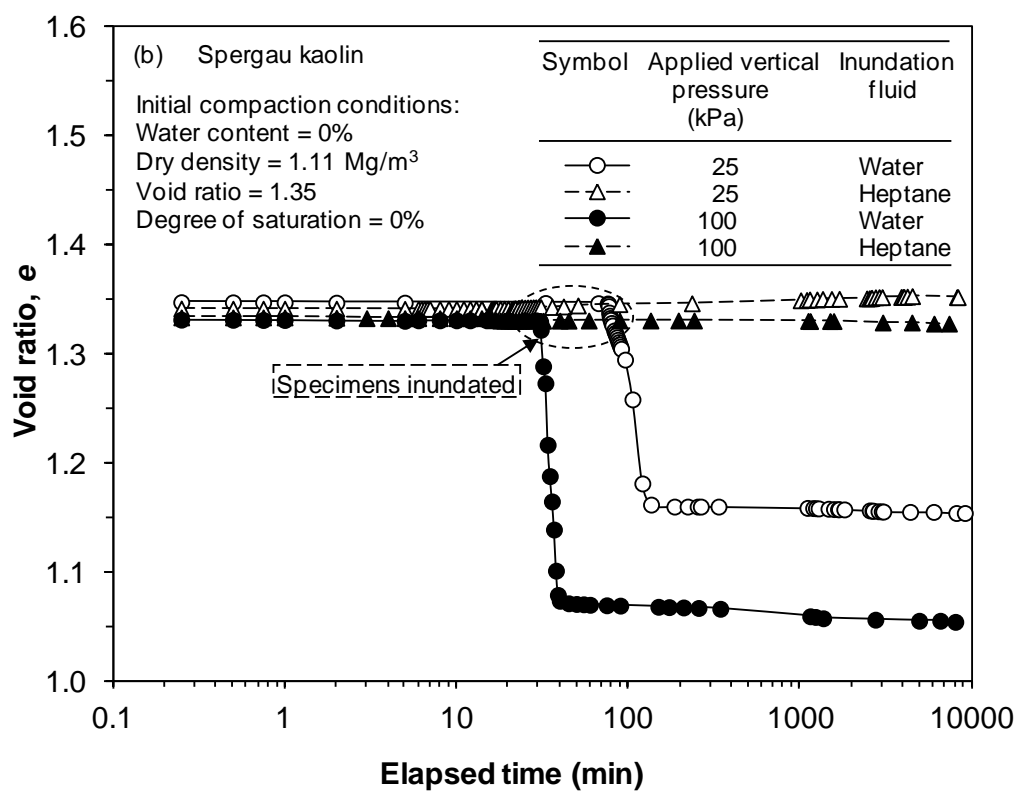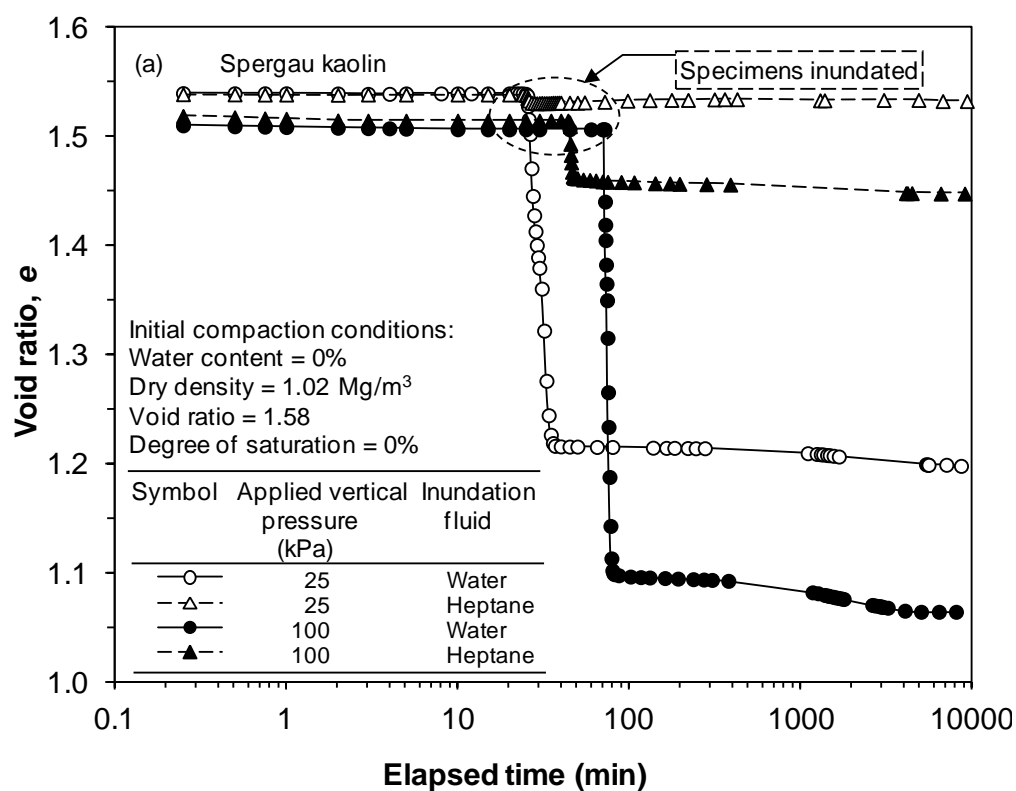502 **Fig. 5** Summary of vertical deformations of Calcigel bentonite specimens

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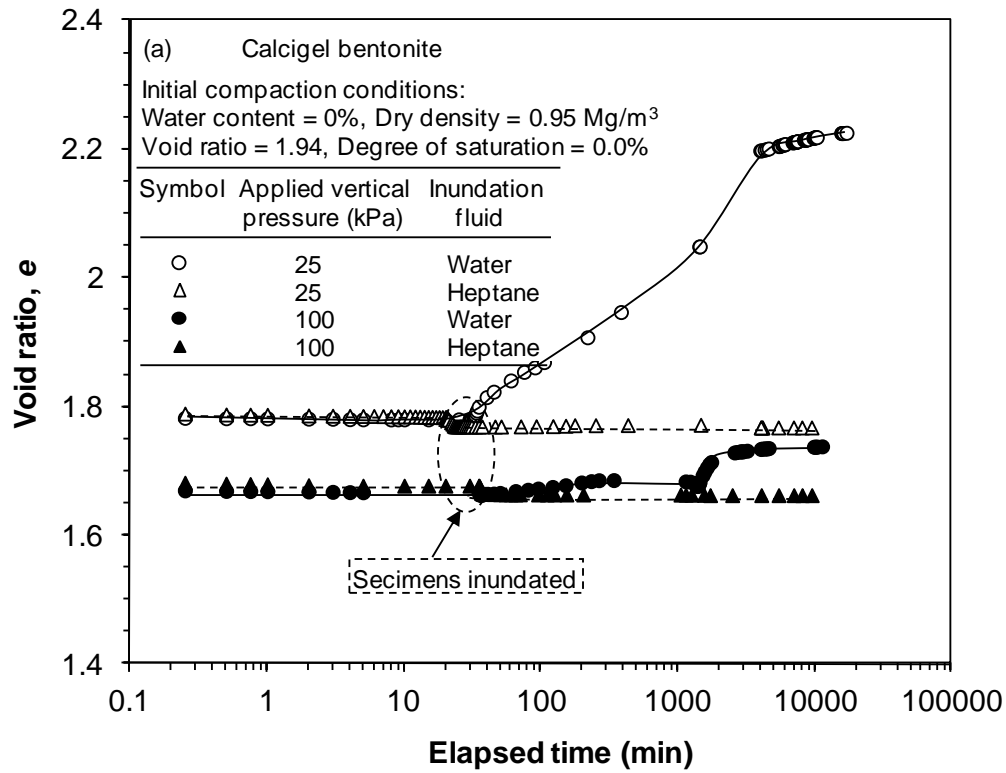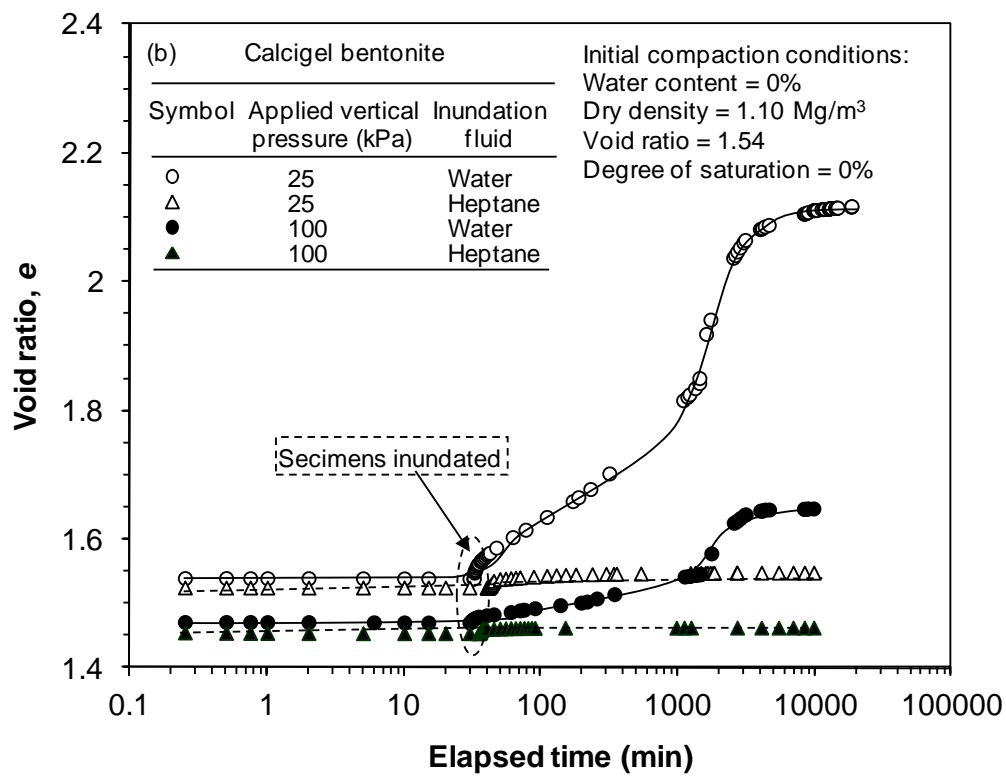508   **Fig. 1** Time-deformation behaviour of Spergau kaolin specimens (initial water content = 0%)
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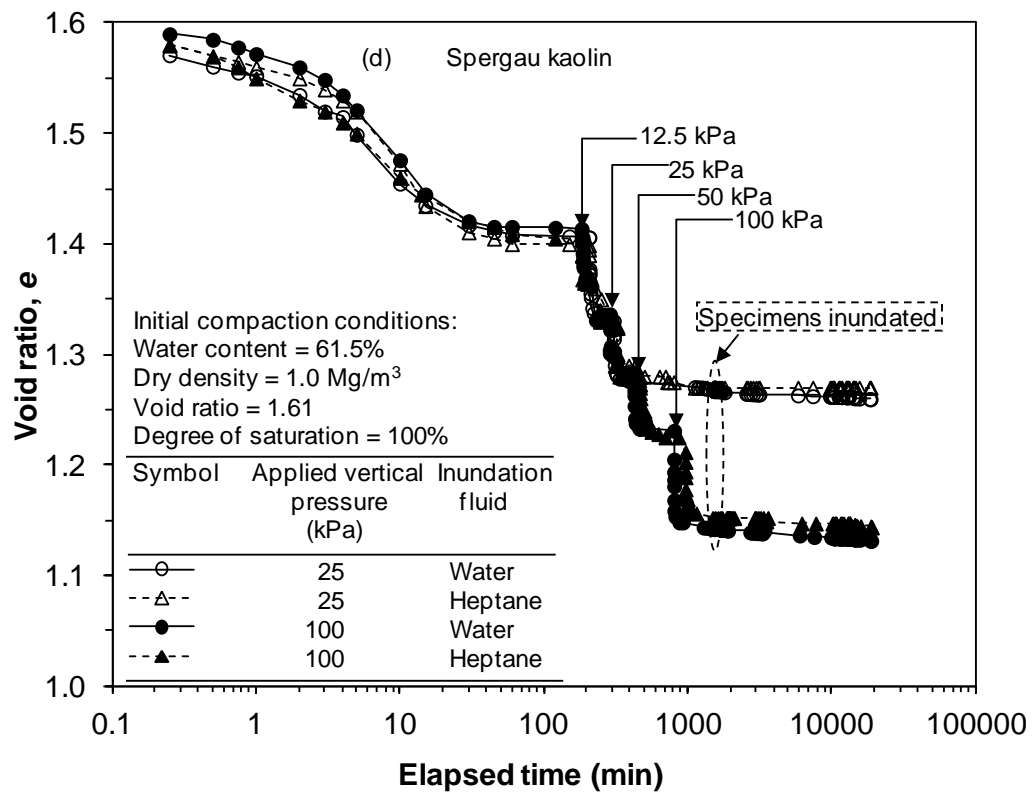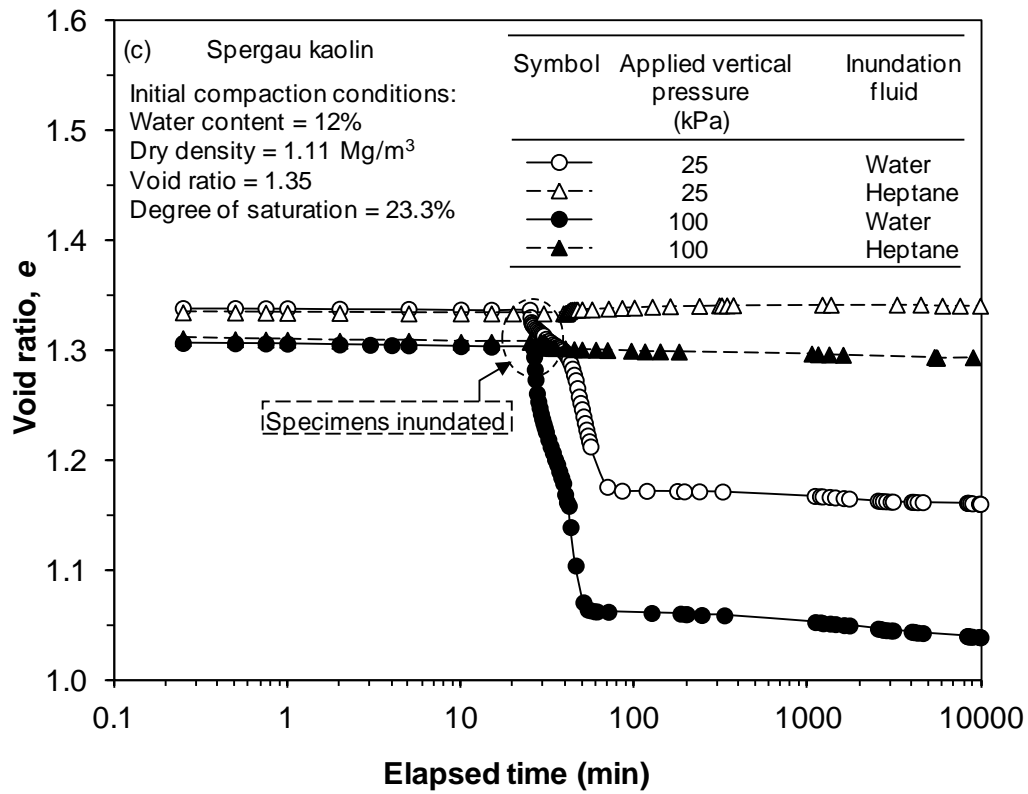
Fig. 2 Time-deformation behaviour of Spergau kaolin specimens with initial water contents of (a) 12% and (b) 61.5%

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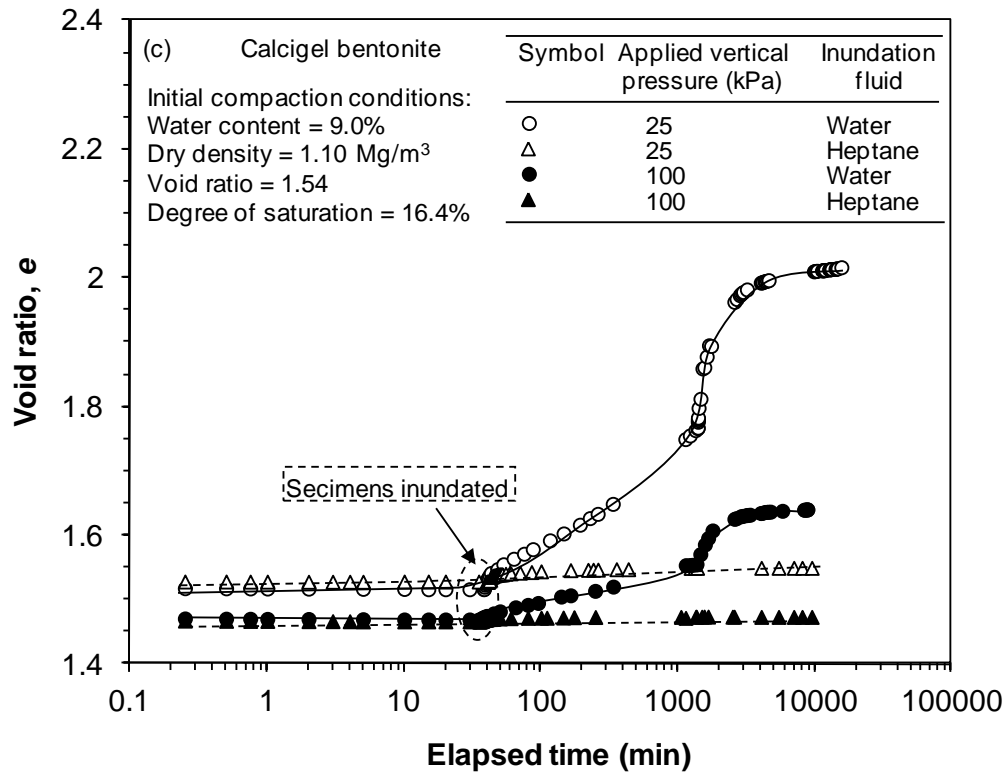517 **Fig. 3** Time-deformation behaviour of Calcigel bentonite specimens (initial water content =
518 0%)

The chart shows Void ratio, e (y-axis, from 1.4 to 2.4) versus Elapsed time (min) (x-axis, logarithmic from 0.1 to 100000).

Chart legend and conditions:

(c) Calcigel bentonite

Initial compaction conditions:
Water content = 9.0%
Dry density = 1.10 Mg/m³
Void ratio = 1.54
Degree of saturation = 16.4%

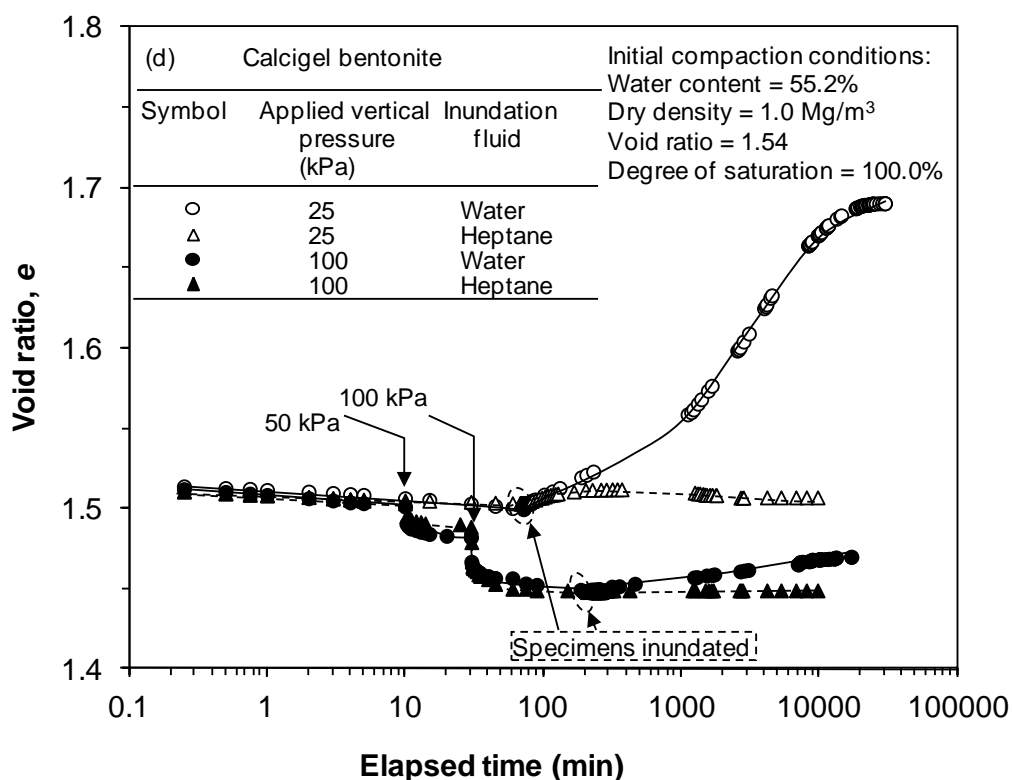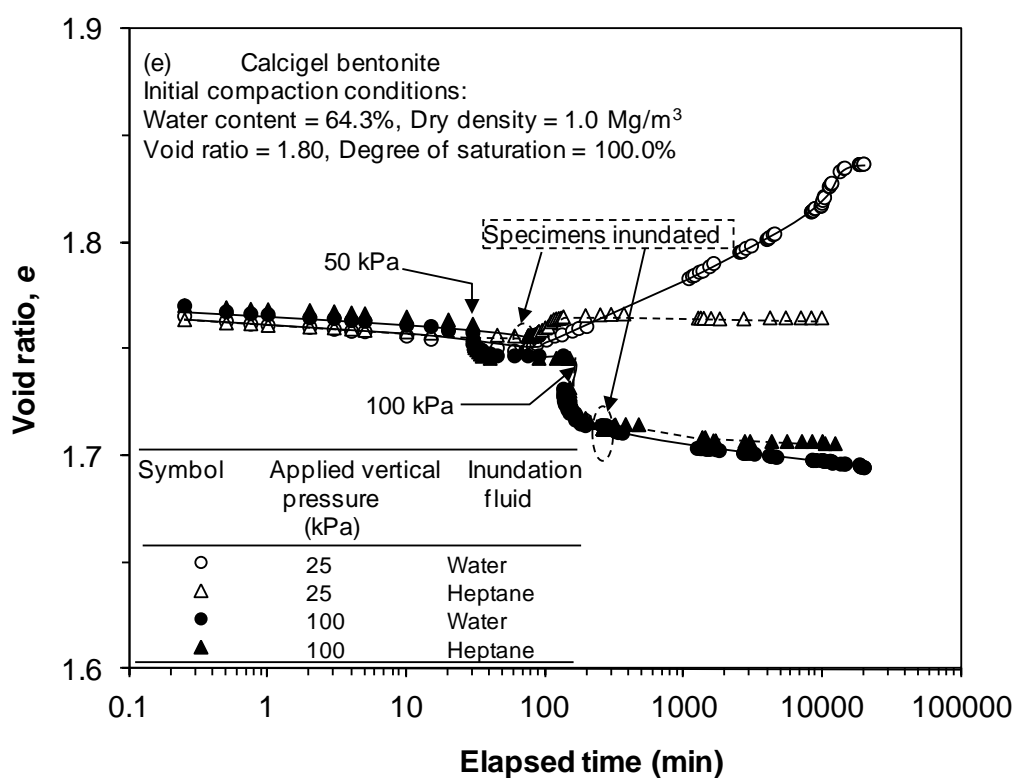| Symbol	Applied vertical pressure (kPa)	Inundation fluid
○	25	Water
△	25	Heptane
●	100	Water
▲	100	Heptane

Secimens inundated

Fig. 4 Time-deformation behaviour of Spergau kaolin specimens (initial water content = 9%)

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Fig. 5 Time-deformation behaviour of Calcigel bentonite specimens with initial water contents of (a) 55.2% and (b) 64.3%

Volume change behaviour of swelling and non-swelling clays upon inundation with water and a low dielectric constant fluid

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Highlights

- Volume change behaviour of clays with water and heptane as the inundating fluids is presented.
- Mineralogy, dielectric properties, compaction conditions, and stress impact the volume change.
- Moblised attractive and repulsive forces dictate compression and swelling in unsaturated clays.

Volume change behaviour of swelling and non-swelling clays upon inundation with water and a low dielectric constant fluid

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Abstract: Studies of the volume change behaviour of saturated and unsaturated fine-grained soils upon exposure to various types of fluid are of significant interest while dealing with contaminated soils. In this study, the one-dimensional volume change behaviour of two clays (Spergau kaolin and Calcigel bentonite) with dominant minerals as kaolinite and montmorillonite was studied by inundating several initially unsaturated and saturated clay specimens with water and a non-polar organic liquid (heptane). For both cases, applied vertical pressures of 25 and 100 kPa were considered under oedometric conditions. The magnitude of compression and swelling deformations of the clays were found to be strongly dependent upon the mineralogy of clays, dielectric properties of the pore-fluid, initial compaction conditions, and applied stress during the wetting process. The test results indicated distinct interactions between unsaturated clays and water. Specimens of Spergau kaolin exhibited compression with water, whereas specimens of Calcigel bentonite exhibited swelling. Compression and swelling deformations of the clays with heptane as the inundating liquid remained within about ± 1.0% for both clays indicating a very minor interaction between unsaturated clays and molecules of this non-polar organic liquid. The test results emphasized the significance of attractive and repulsive forces and their impact on the volume change behaviour of clays of different mineralogy.

Keywords: minerology of clays; swelling; compression; pore-fluid; waste containment; laboratory tests

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