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Investigation into the effect of backpressure on the mechanical behavior of methane-hydrate-bearing sediments via DEM analyses

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Abstract:

Backpressure has been extensively applied in experimental tests to improve the water saturation of samples，and its effect on the strength of saturated soils has been traditionally regarded as trivial in Soil Mechanics. However, a non-negligible influence of backpressure on the macro mechanical properties of methane-hydrate-bearing-sediments (MHBS) has been surprisingly observed in several recent experiments reported in the literature. This paper aims to shed light on this phenomenon. A theoretical analysis on the microscopic interaction between soil grains and inter-particle methane hydrate (MH) was carried out to highlight how backpressure affects the mechanical properties of the inter-particle MH which in turn affect the macroscopic mechanical behavior of MHBS. The influence of backpressure is accounted for in a new bond contact model implemented into the Distinct Element Method (DEM). Then, a series of DEM biaxial compression tests were run to investigate the link between mechanical properties of MHBS and backpressure. The DEM numerical results show that shear strength, small strain stiffness and shear dilation of MHBS increase with the level of backpressure. As the critical state is approached, the influence of backpressure ceases. Moreover, the elastic modulus and cohesion of MHBS increase linearly while the internal friction angle decreases at a decreasing rate as the backpressure increases. Simple analytical relationships were achieved so that the effect of backpressure on the mechanical properties of MHBS can be accounted in the design of laboratory tests to characterize the mechanical behavior of MHBS.

Keywords:

1 Introduction

Gas hydrates are ice-like solids composed of water and gas, in which the gas molecules, mainly methane, are trapped within a lattice formed by water molecules [1,2,3]. Hydrates, especially methane hydrates (MHs), have attracted increasing global interest in the past two decades due to their potential use as a future energy resource [4-9]. They are highly concentrated: for instance one cubic foot of hydrate normally produces 150-170 cubic feet of natural gas when dissociated [5,10]. It is estimated that the amount of gas in hydrate reservoirs worldwide exceeds the volume of known conventional gas resources [4,11]. Besides, the estimated amount of organic carbon in hydrates is twice as much as that in all conventional fossil fuel resources, including natural gas, coal and oil [12].

MHs are widely found in marine continental margin sediments and permafrost regions where temperature and pressure conditions are appropriate for the stability of MHs [13,14]. In particular, over 90% of natural MHs are found in marine sediments [1]. It has been observed that the presence of MHs changes the mechanical behavior of the bearing sediments, for instance by greatly enhancing the strength of the host sediments [15-17], especially when the MH content is high. However, MHs may dissociate as a result of changes of temperature/pressure conditions, or human interventions (e.g., installation of platforms, cables or pipelines), which in turn may change the microstructure of methane-hydrate-bearing sediments (MHBS) and deteriorate their mechanical properties. This may then compromise the stability of wellbores, pipelines, rig supports and other equipment involved in oil and gas production and in the worst case scenario lead to submarine landslides [10,18]. In order to investigate the triggering mechanisms of these geo-hazards and to establish suitable macroscopic constitutive models to solve boundary-value problems related to the production of MH from MHBS (e.g., potential instability of the walls of the drilling wells), a comprehensive investigation is needed to understand the fundamental mechanical properties of MHBS.

29   To date, various geo-laboratory tests have been performed on artificial or natural

30   MHBS samples to investigate their mechanical behavior. To replicate the pore-water

31   pressure condition beneath the ocean floor and to stabilize MHs in the soils, MHBS

32   samples are usually tested with some backpressure applied. The effect of

33   backpressure is often neglected since, according to the principle of effective stresses

34   [19], it is the effective confining pressure that controls the mechanical behavior of

35   MHBS. However, experimental results recently obtained by Miyazaki et al. [20] and

36   Hyodo et al. [21] surprisingly indicate that backpressure does influence the

37   mechanical properties of MHBS, e.g., shear strength and stiffness of MHBS increase

38   with the level of backpressure. The mechanisms at the root of this phenomenon

39   remain largely unknown. It is authors' opinion that the root causes of the

40   backpressure-dependent mechanical behavior of MHBS manifested at the

41   macroscopic scale have to be sought in the interaction between MHs and soil grains at

42   the microscopic scale. To this end, the paper aims to investigate this interaction by

43   performing a campaign of biaxial tests via the Distinct Element Method (DEM). The

44   choice of the DEM is supported by the fact that biaxial DEM tests have already

45   demonstrated to be successful at establishing quantitative correlations between bond

46   properties and the mechanical behaviour observed at the macroscopic scale for other

47   bonded granulates [22-25].

48   Although experimental tests performed for different values of backpressure are

49   reported in the literature (e.g., backpressures of 5MPa, 10MPa and 15MPa in [26-28];

50   backpressures of 6MPa, 7MPa, 8MPa and 8.5MPa in [20]; a backpressure of 8MPa in

51   [28], backpressures of 15MPa and 20MPa in [29]), no systematic study, to the best of

52   authors' knowledge, has been carried out until now. So the campaign of tests here

53   reported is the first systematic investigation into the effect of backpressure on the

54   mechanical behavior of MHBS.

55   The paper has the following structure. Firstly, the available experimental evidence

56   about the influence of backpressure on the mechanical properties of MHBS, [20,21],

57   is briefly recalled. Secondly, a bond contact model capturing the interaction between

58   soil particles and MH bonds under different levels of backpressure is introduced, [30].

59   Thirdly, the results obtained via DEM biaxial tests performed for various levels of

60   backpressure are illustrated. The numerical results are compared with experimental

61   tests to assess the predictive capability of the DEM. The influence of backpressure on

62   the macroscopic mechanical properties of MHBS in terms of elastic modulus,

63   apparent cohesion and internal friction angle, and their dependency on the level of

64   MH saturation and on the effective confining pressure is also investigated.

65

66   **2 Backpressure-dependent behavior of MHBS**

67   In geotechnical laboratory tests, backpressure is widely used to fully saturate soil

68   samples [31-37]: the samples are saturated by increasing at the same time the internal

69   pore-water pressure, σ_w (also called backpressure), and the cell pressure, σ_3, of the

70   same amount. According to the principle of effective stresses [19], the amount of

71   backpressure should not affect the mechanical behavior of fully saturated samples.

72   However, several recent experiments [20,21] unequivocally point to a non negligible

73   influence of the applied backpressure on the mechanical properties of MHBS. For

74   instance, tests reported by Miyazaki and Hyodo (see Figure 1) run for various levels

75   of backpressures with all the other variables (e.g., effective confining pressure, void

76   ratio, and temperature) being constant except for a small variation of MH saturation,

77   show that both the peak deviator stress and the secant elastic modulus of MHBS

78   increase for increasing backpressure. The small variation of MH saturation, S_{MH},

79   between the tested samples is unavoidable due to the current experimental limitations

80   in controlling the amount of MH flowing into the samples. However, the influence of

81   backpressure is evident observing that (see Fig 1a) samples with lower MH saturation

82   (S_{MH}=17%) subject to a higher backpressure ($\sigma_w = 8$ MPa) exhibit a higher shear

83   strength than samples with higher saturation (S_{MH}=19%) subject to a lower

84   backpressure ($\sigma_w = 6$ MPa) .

85   Several authors showed that MH plays an important role in the mechanical

86 behavior of MHBS [14,17,39,40], e.g., they tend to increase strength, stiffness, strain

87 softening and shear dilation of the host sand. MHs are found in the pores of MH

88 bearing sediments. Brugada et al. [14] and Waite et al. [17] identified three main

89 formation habits at the pore scale, plotted in Figure 2: (1) pore-filling, with methane

90 hydrates floating in the pore fluid without bridging any particles; (2) load-bearing,

91 with hydrate particles being part of the strong force chains of the granular assembly;

92 and (3) cementation, with methane hydrates cementing sand grains (acting as bridges

93 between grains). As suggested by Waite et al. [41] and confirmed by experimental

94 data from different sources [42,43], the bulk properties of MH bearing sediments

95 strongly depend on the formation habits of the hydrates. For instance, hydrates acting

96 as cementation agents at inter-particle contacts cause a larger increase in strength and

97 stiffness than pore-filling hydrates. This implies that hydrate morphology has to be

98 accounted for into any realistic constitutive model. In this respect, only an

99 investigation at the scale of the pores can shed light on the link between pore habit

100 and macroscopic properties [44]. Brugada et al. [14] made the first attempt to

101 simulate the pore-filling habit by 3D DEM simulations. In their simulations, soil

102 grains and MH particles get into contact and exchange forces, however without

103 exhibiting any cementation at contacts. Jung et al. [40] improved previous numerical

104 research work by characterizing the mechanical behaviour of MHBS for two cases of

105 hydrate pore habits: distributed hydrates and patchy saturation. In this paper instead,

106 habit 3 is considered (see Figure 2). This habit is of common occurrence especially in

107 unconsolidated sediments containing an abundant gas phase: for instance significant

108 deposits of cementing MHs have been discovered in the Blake Ridge, off the

109 southeast coast of the United States [45] and in the Cascadia margin [46]. In this habit,

110 hydrates form mainly at grain contacts generating interparticle bonds [41]. With

111 regard to laboratory tests, this habit is induced by percolating methane gas through

112 samples of sands partially saturated with water in the pendular regime. The formation

113 of MH generates the onset of bonds between nearby sand grains whose stiffness

114 remains unchanged since the grains are not coated by the hydrate. [21-23,30,39,41,47]

115   showed that this pore habit plays an important role in the macro-mechanical

116   behaviors of MHBS and contributes substantially to the bulk properties of MHBS.

117   For instance in case of a MH saturation of 50%, the strength of the MHBS can be

118   twice as much as that of the host sand [20-24].

119   Figure 3 presents a schematic view of a pair of soil grains immersed in water and

120   cemented by a MH inter-particle bond. MH can be envisaged as a solid block

121   surrounded by water at a pressure equal to the backpressure acting on the MHBS

122   sample. For the range of water backpressure employed in the experiments of [20] and

123   [21], the sand grains can be assumed unbreakable and of negligible deformability.

124   MHs instead are crystalline compounds whose deformability and strength are affected

125   by the level of surrounding pressure. In fact, tests on blocks of pure solid MH, i.e.,

126   without sand grains [42,48-52], show that both the strength and elastic modulus of

127   MHs increase with the level of surrounding pressure. In the next section, the bond

128   contact model incorporating the main mechanical features of the MH bonds and

129   employed in all the DEM biaxial tests presented in the paper, is illustrated.

130

131   3 MH bond contact model incorporating the effect of backpressure

132   The bond model for MH cementing pairs of sand grains was first formulated in Jiang

133   et al. [30]. The contact law here employed for the bond, experimentally derived in

134   [53,54], is presented in Figure 4. The mechanical response of the bond is featured by

135   elastic behavior until breakage followed by brittle failure. For an intact bond, the

136   normal force F_n, the shear force F_s, and the moment M are computed, respectively, by:

$$F_n = K_n(u_n - u_0) \tag{1a}$$

$$F_s \leftarrow K_s \Delta u_s + F_s \tag{1b}$$

$$M \leftarrow K_r \Delta \theta + M \tag{1c}$$

140   where u_n is the overlap of two particles; Δu_s and $\Delta \theta$ are the increments of relative

141   shear displacement and relative rotation angle, respectively; u_0 is the initial overlap

between two adjacent particles, indicating the distance between the two particles at the time of formation of the MH bond. K_n, K_s and K_r in Equation (1) are the normal, tangential and rolling bond contact stiffnesses, respectively. They are given by:

$$K_n = \frac{B E_{MH}}{t} \qquad (2a)$$

$$K_s = \frac{2}{3} K_n \qquad (2b)$$

$$K_r = \frac{1}{12} K_n B^2 \qquad (2c)$$

where E_{MH} is the Young's modulus of MH; t is the average thickness of the MH bond defined as the area of MH bond divided by its width. The bond width, B (see Figure 3), was prescribed for all bonds as a fraction of the equivalent radius of the two bonded particles, $\bar{R} = 2 R_1 R_2 / (R_1 + R_2)$, so that $B = \beta \bar{R}$.

Contact interaction between non-bonded particles (e.g. contacts formed by particles moving around after their MH bonds have broken) is linearly elastic – perfectly plastic, i.e. linear springs in both normal and tangential directions plus a frictional slider in the tangential direction only (see [30, 55]).

With regard to the strength of the bonds, in Figure 5 the bond strength envelope employed in the DEM simulations is plotted in the F_s-M-F_n space. The strength of a bond subject to pure tensile, compressive, shear or rolling actions is denoted by R_{tb}, R_{cb}, R_{sb} and R_{rb} respectively. The strength envelope in the F_s-M plane assumes an elliptical shape:

$$\frac{M^2}{R_{rb0}^{\,2}} + \frac{F_s^{\,2}}{R_{sb0}^{\,2}} = 1 \qquad (3)$$

with R_{sb0} being the tangential strength of the bonds in the absence of any moment, and R_{rb0} the rolling strength in the absence of any shear force. R_{sb0} and R_{rb0} are both a function of the applied normal force F_n and of the tensile and compressive bond strengths, R_{tb} and R_{sb}, respectively. They were determined on the basis of the

166 experimental data available on bonding MH [20,21] according to the procedure

167 outlined in [53]:

$$R_{sb0} = 1.38 \cdot 0.38 \cdot (F_n + R_{tb}) \cdot \left(\ln \frac{R_{cb} + R_{tb}}{F_n + R_{tb}} \right)^{0.59} \qquad (4a)$$

$$R_{rb0} = 1.366 \cdot 0.741 \cdot (F_n + R_{tb}) \cdot \left(\ln \frac{R_{cb} + R_{tb}}{F_n + R_{tb}} \right)^{0.59} \qquad (4b)$$

$$R_{tb} = B \times \sigma_{tMH} \qquad (4c)$$

$$R_{cb} = B \times \sigma_{cMH} \qquad (4d)$$

172 where σ_{tMH} and σ_{cMH} are the tensile and compressive strengths of pure MH,

173 respectively. The Young's modulus and the tensile and compressive strengths of MH

174 are a function of pore-water pressure temperature and MH density. Tests performed

175 on pure MH samples [30] provide the following expressions:

$$\frac{E_{MH}}{p_a} = 3\left(\frac{\sigma_w}{p_a} \right) - 1.98\left(\frac{T}{T_0} \right) + 4950.50\left(\frac{\rho}{\rho_w} \right) - 1821.78, \qquad (5a)$$

$$\frac{\sigma_{cMH}}{p_a} = 0.81\left(\frac{\sigma_w}{p_a} \right) - 2.08\left(\frac{T}{T_0} \right) + 184.16\left(\frac{\rho}{\rho_w} \right) - 134.65, \qquad (5b)$$

$$\frac{\sigma_{tMH}}{p_a} = 0.45\left(\frac{\sigma_w}{p_a} \right) - 1.15\left(\frac{T}{T_0} \right) + 101.75\left(\frac{\rho}{\rho_w} \right) - 74.39, \qquad (5c)$$

179 where $\sigma_w = \gamma_w h$ as shown in Figure 5, with h being the depth of the water table; T the

180 ambient temperature; ρ the MH density; p_a the standard atmospheric pressure

181 $(1.01 \times 10^5 \mathrm{Pa})$; T_0 a reference temperature of $1\,^{\circ}\mathrm{C}$ and ρ_w the density of water at $4\,^{\circ}\mathrm{C}$.

182   Concerning the level of MH saturation, it is convenient to introduce a

183 dimensionless parameter S_{MH}, expressing the ratio of the methane hydrate area, A_{MH},

184 over the total void area, A_V, i.e., $S_{MH} = A_{MH} / A_V$. This parameter is a 2D measure

185 of the level of MH saturation equivalent to the 3D ratio of methane volume over the

186   void volume. S_{MH} can be calculated from the geometry of all the bonds in the

187   sample (see [30]) according to the following formula:

188
$$S_{MH} = \frac{A_b}{A_v} + S_{MH_0} = \frac{(1+e_p)}{e_p A} \sum_{i=1}^{m} A_{b,i} + S_{MH_0}$$

$$= \frac{(1+e_p)}{e_p A} \sum_{i=1}^{m} \overline{R}_i^2 \left[2\beta - \beta\sqrt{1 - \frac{\beta^2}{4}} - 2\arcsin(\frac{\beta}{2}) + \frac{t_{0i}\beta}{\overline{R}_i} \right] + S_{MH_0}$$

(6)

189   where A is the total area of the sample, e_p the sample planar void ratio, m the total

190   number of MH bonds in the sample, $A_{b,i}$ the area of each bond, $t_{0,i}$ the minimum

191   thickness of each bond (see Figure 3) and $\overline{R}_i$ the equivalent radius for each bonded

192   pair of particles (the width of each bond being $B_i = \beta\overline{R}_i$). In Equation (6), S_{MH_0} is the

193   threshold value of hydrate saturation at which methane hydrates start to bond sand

194   grains which in turn is a function of the hydrate growth history. In [30] an in-depth

195   explanation of Eq (6) is provided.

196   The input parameters of the adopted bond model are four (listed in Table 1): the

197   pore-water pressure surrounding the bond, σ_w, temperature, T, the density of MH, ρ,

198   and the bond width parameter β. However, for the range of backpressures considered

199   in this study (5~20MPa), the maximum possible variation of MH density is only

200   0.02g/cm^3 [59], hence a constant MH density of 0.9 g/cm3 was assumed in all DEM

201   simulations. Then, the Young's modulus (E_{MH}), tensile strength (σ_{tMH}) and

202   compressive strength (σ_{cMH}) of the bonds can be obtained via Equations (5). Finally,

203   the stiffness (i.e., K_n, K_s, K_r) and strength parameters (i.e., R_{tb}, R_{cb}, R_{sb0} and R_{rb0}) of

204   the bonds can be calculated via Equations (2) and (4), respectively. It is important to

205   note that the density of MH is affected by the amount of backpressure applied.

206

4 DEM tests on MHBS

208   The presented bond contact model was implemented into a C++ subroutine and run

209   in the PFC-2D code [56].

210

211   **4.1 Simulation procedure**

212   In the DEM numerical tests, we set out to mimic the methane hydrate generation

213   process as reported in the experimental tests of [21] (see Figure 1). In their tests, first,

214   methane gas was pumped into the host specimens to form MHs under a backpressure

215   of 4MPa, 1℃ temperature and an effective confining pressure of 0.2MPa. After the

216   generation of MHs was completed, water was injected to fully saturate the samples

217   and backpressure was increased to the prescribed value of interest. Then, the samples

218   were consolidated under the prescribed confining pressure with both backpressure

219   and temperature kept constant. Finally, the samples were sheared in drained

220   conditions at a strain rate of 0.1%/min and constant backpressure and temperature.

221   To mimic these experiments in the DEM, first homogenous samples without

222   any cementation were generated via the 'multi-layer under-compaction method' [56],

223   then they were isotropically consolidated to an effective confining pressure of 0.2

224   MPa. Second, bonds of various thicknesses were activated between pairs of adjacent

225   particles either in contact or having a distance between their edges of less than a

226   prescribed threshold and the bond parameters being chosen to reflect the presence of

227   4 MPa of backpressure at 1℃. Then, the bond parameters were changed to reflect the

228   application of various levels of backpressure (*i.e.* 5MPa, 10MPa, 15MPa or 20MPa),

229   which remained constant during the subsequent part of the tests. Thereafter, the

230   effective confining pressure of the samples was gradually increased to a prescribed

231   value (isotropic consolidation). Lastly, the samples were axially loaded by moving

232   the top and bottom platens at a constant strain rate of 5%/min maintaining the

233   backpressure constant.

234

235   **4.2 Sample parameters**

236 Figure 6 presents the grain size distribution of Toyoura sand employed by Hyodo et al.

237 [21] and the distribution employed in our DEM simulations. The DEM material is

238 made up of disks of 10 diameter sizes ranging from 6.0mm to 9.0mm. The median

239 diameter, d_{50}, is 7.6 mm, with the uniformity coefficient of the particle size

240 distribution being 1.3. The particle size distribution of the DEM material is obtained

241 by an almost uniform upscale of the experimental particle size distribution. Particle

242 scaling is necessary to perform the simulations within an affordable runtime. Table 2

243 lists the values of the DEM parameters used in the simulations. In Figure 7, a DEM

244 MHBS sample is plotted before compression is applied. It can be observed that the

245 initial MH bonds (plotted as black solid lines in the figure), are homogenously

246 distributed in the sample.

247   Close examination of SEM images of MHBS samples shows that MH bonds are

248 limited in size, i.e., they are formed only between particles lying within a certain

249 distance (t_0^{th}) whose value depends on various factors, e.g. relative density of the

250 granular sample, level of MH saturation, etc. According to the experimental

251 observations available [30], this distance was assumed as 5% of the median particle

252 diameter, $i.e.$ t_0^{th}/d_{50}=0.05. The relationship between β and $S_{MH} - S_{MH_0}$ obtained from

253 Eq (6) for S_{MH_0}=20% (value taken according to [43, 58]) and an initial planar void

254 ratio, e_p, of 0.25 is plotted in Figure 8. The value of β employed in each DEM test to

255 reproduce a prescribed S_{MH}, was selected using the relationship in Figure 8.

256

5 DEM numerical results

5.1 Stress-strain relationships obtained at different backpressures

259 In the description of the state of stress, the following 2D invariant variables have been

260 used: $s = (\sigma_y + \sigma_x)/2$ (mean effective pressure) and $t = (\sigma_y - \sigma_x)/2$ (deviator stress).

261 In Figure 9 both the stress-strain and volumetric behaviors of MHBS from

262 experimental triaxial tests of Hyodo et al. [21] and our DEM simulations are plotted.

263 In the figure, positive and negative values of volumetric strain means dilation and

264 contraction, respectively. For each case, results for two different levels of

265 backpressure (5MPa and 10MPa) are presented. It can be observed that although the

266 numerical results do not quantitatively reproduce the experimental data, they capture

267 the essential features of the observed influence of backpressure on the stress-strain

268 response of the MHBS. These features can be listed as follows: 1) the peak deviator

269 stress manifested for a backpressure of 10MPa is higher than the peak deviator

270 manifested for 5MPa of backpressure although the MH saturation in the former test is

271 (slightly) lower than in the latter one; 2) the initial slope of the stress-strain

272 relationship for the test run at 10MPa of backpressure is higher than the test run at

273 5MPa; 3) the difference between the deviator stress of the two samples (5MPa and

274 10MPa backpressure) becomes much smaller at large strains; (4) shear dilation

275 increases with the level of backpressure.

276 To explain these findings, the stress-strain and the volumetric responses obtained

277 from DEM biaxial tests run for various level of confinement (σ_3') and backpressures

278 (σ_w) are plotted in Figure 10. It emerges that the peak deviator stress increases for

279 increasing levels of backpressure; however the deviator stress at large axial strains

280 tend to reach a unique value independently of the level of backpressure applied. So

281 the influence of backpressure on the stress-strain response of MHBS at large strains is

282 much less significant than at small strains. This is in agreement with the available

283 experimental data (see Figure 9b).

284 In order to investigate the mechanisms taking place at the microscale responsible

285 for the influence of back-pressure on the observed MHBS mechanical response, we

286 examined the pattern of bond breakages and the distribution of contact force chains

287 throughout the tests. In Figure 11 the percentage of intact MH bonds is plotted against

288 the applied axial strain. Obviously the percentage of MH bonds decreases as the axial

289 strain increases. It is evident that the percent of intact MH bonds at large strains (e.g.,

290 15%) is heavily affected by the level of applied backpressure: the higher the level of

291 backpressure, the larger the number of intact bonds. This result appears to be at odds

292 with the observation that the deviator stresses relative to these tests are very close at

293 large strains (see Figure 10). The distribution of the intact MH bonds in the sample

294 holds the key to unveil this apparent contradiction. The intact bonds of a sample

295 subject to a large level of backpressure and a sample subject to a small level, 20MPa

296 and 5MPa respectively, are plotted in Figure 12. It can be observed that in the case of

297 large backpressure (20MPa) most bonds are broken and the intact ones are roughly

298 uniformly distributed. Instead in case of low backpressure (5MPa), most bonds are

299 still intact but their distribution in the sample is highly inhomogeneous. In particular a

300 diagonal narrow band with only a few disconnected MH bonds appears running from

301 the upper to the bottom boundary of the sample. This band forms a weak zone which

302 determines the maximum vertical stress that the sample can sustain. This is confirmed

303 by observing that the contact force chains in the weak zone of the sample subject to

304 low backpressure are significantly thinner than in the other zones of the sample (see

305 Figure 12). In the two samples the maximum stress that can be sustained is similar

306 since in both cases it is governed by zones where there are no networks of

307 interconnected bonds. These observations at the microscale explain why the deviator

308 stresses of the two samples at large strains are close to each other and to the value

309 observed for samples without any MH bond.

310   So it can be concluded that the peak deviator stress in MHBS samples increases

311 with the level of backpressure due to the fact that larger applied backpressures

312 increase the strength of MH bonds; however, this strength increment progressively

313 reduces with the progression of sample shearing which causes more and more bonds

314 to break until a full diagonal band of broken bonds running from the upper to the

315 bottom boundaries of the sample is formed. This band rules the maximum vertical

316 stress that can be sustained by the sample so that the deviator stress tends to reach a

317 unique value at large strains independent of the level of backpressure applied.

318

319 **5.2 Effect of backpressure on the mechanical properties of MHBS**

320 In Figure 13 the secant elastic modulus calculated at 50% of the measured deviator

321 stress (E_{50}) is plotted against the level of backpressure applied for tests run at various

322 levels of MH saturations and effective confinement. From the figure emerges that the

323 elastic modulus increases almost linearly with the level of backpressure σ_w whatever

324 the value of effective confining pressure, σ_3', and MH saturation, S_{MH}, are. Also it

325 can be observed that the elastic modulus is higher at larger effective confinement (σ_3')

326 and MH saturation (S_{MH}) as can be expected. So the elastic modulus of MHBS can be

327 determined as a function of σ_w, σ_3' and S_{MH}:

$$E_{50} = E(\sigma_w, \sigma_3', S_{MH}) \qquad (7)$$

328

329 For sake of generality, it is convenient to use an expression for Eq. (7) in

330 dimensionless form:

$$\frac{E_{50}}{p_a} = A_1 \frac{\sigma_3'}{p_a} + (A_2 \frac{\sigma_w}{p_a} + A_3) \cdot (S_{MH} - S_{MH0})^{A_4}$$

331 $$\qquad (8)$$

332 with A_1, A_2, …A_4 being constant values to be determined by biaxial tests. In our case,

333 the constants assume the following values: A_1=6.46, A_2=1.36, A_3=1470, and A_4 =

334 0.30. The curves obtained according to Eq. (8) were reported in Figure 13 as well: it

335 is apparent that they well interpolate the data. In the figure a vertical dash line signals

336 the critical pressure for the dissociation of MH at 1 °C, $\sigma_{w(cr)}$=2.9 MPa, [60]. Since

337 MH cannot exist at a backpressure lower than the critical pressure, Equation (8) no

338 longer applies for $\sigma_w < \sigma_{w(cr)}$.

339 The peaks of the deviator stress obtained from all the tests performed are plotted

340 in the $t - s$ plane (see Figure 14) together with their linear interpolations. The slope

341 and the intercept of the linear interpolations provide the internal friction angle (ϕ)

342 and the apparent cohesion (c) of the MHBS. From the figure it emerges that ϕ and c

343 depend on both backpressure and MH saturation:

344 $$c = c(\sigma_w, S_{MH}) \qquad (9a)$$

345 $$\phi = \phi(\sigma_w, S_{MH}) \qquad (9b)$$

346 The values of apparent cohesion were plotted against backpressure in Figure 15. It

347 emerges that c increases almost linearly with the level of backpressure and the slope

348 of the line is dictated by the level of MH saturation. Therefore a convenient

349 dimensionless expression for Equation (9a) is:

350 $$\frac{c}{p_a} = (B_1 \frac{\sigma_w}{p_a} + B_2) \cdot (S_{MH} - S_{MH0})^{B_3} \qquad (10a)$$

351 with $B_1, \ldots B_3$ being constant values. In our case $B_1 = 0.115$, $B_2 = -0.10$ and $B_3 = 0.43$.

352 The lines obtained according to Eq. (10a) were reported in Figure 15: it is apparent

353 they interpolate the data very well.

354   The relationship between internal friction angle and backpressure obtained from

355 the DEM tests (Eq (9b)) is plotted in Figure 16. It can be observed that the internal

356 friction angle decreases with the level of backpressure and the rate of decrease is

357 roughly insensitive to the amount of MH saturation applied. Hence, a suitable

358 expression for Equation (9b) is:

359 $$\phi = C_0 - C_1 (\frac{\sigma_w}{p_a})^{c_2}, \qquad (10b)$$

360 with $C_0, \ldots C_2$ being constant values that in our case are: $C_0 = 23.2°$, $C_1 = -0.14$ and

361 $C_2 = 0.72$. The linear relationship obtained on the basis of Equation (10b) is reported

362 in Figure 16 (solid black line).

363

364 **5.3 Other factors affecting the mechanical properties of MHBS**

365   In addition to backpressure, both surrounding temperature and the density of

366 inter-particle methane hydrate influence deformability and strength of the inter-

367 particle methane hydrate (see Eqs. (5)) which in turn affect the mechanical properties

368   of MHBS. In the following, new equations will be proposed to account for the

369   influence of temperature (T) and methane hydrate density (ρ) on the secant elastic

370   modulus at 50% of the deviator stress (E_{50}), apparent cohesion (c), and internal

371   friction angle (ϕ) of the MHBS.

372   The values of E_{50}, calculated from the DEM tests, are plotted against temperature

373   and methane hydrate density in Figures 17 and 18 respectively. From the figures

374   emerges that the elastic modulus linearly increases with temperature decreasing and

375   methane hydrate density increasing. This indicates the dependence of E_{50} on

376   temperature and methane hydrate density may be accounted for by the following

377   expression:

378
$$\frac{E_{50}}{p_a} = A_1 \frac{\sigma_3^{'}}{p_a} + \left(A_2 \frac{\sigma_w}{p_a} + A_3 \frac{T}{T_0} + A_4 \frac{\rho}{\rho_w} + A_5\right) \cdot (S_{MH} - S_{MH0})^{A_6} \qquad (11)$$

379   Comparison between Equation (11) and Equation (5a) shows that both the elastic

380   modulus of MHBS (E_{50}) and of pure MH (E_{MH}) hold linear relationships with

381   backpressure (σ_w), temperature (T) and MH density (ρ). Assuming that E_{50} can be

382   expressed as a function of the elastic modulus of pure MH, a simple dimensional

383   expression to determine E_{50} can be derived from Equations (11) and (5a):

384
$$E_{50} = a_1 \sigma_3^{'} + a_2 E_{MH} \cdot (S_{MH} - S_{MH0})^{a_3} \qquad (12)$$

385   with a_1, a_2 and a_3 being constant parameters, and E_{MH} given in Equation (5a). In

386   our case, the constants assume the following values: $a_1 = 6.92$, $a_2 = 0.55$ and $a_3 =$

387   0.30. The linear relationships obtained by using Equation (12) are reported as dashed

388   lines in Figures 17 and 18. It emerges that Equation (12) effectively captures the

389   dependence of E_{50} on backpressure (σ_w), temperature (T), density of pure methane

390   hydrate (ρ), methane hydrate saturation (S_{MH}) and effective confining pressure ($\sigma_3^{'}$).

391   The values of apparent cohesion (c), calculated from the DEM tests, are plotted

392   against temperature and methane hydrate density in Figures 19a and 19b respectively.

393   Similarly to the secant elastic modulus E_{50}, also cohesion linearly increases with

394   temperature decreasing and methane hydrate density increasing. Hence, the

395   dependence of cohesion on temperature and methane hydrate density may be

396   accounted for by the following expression:

397
$$\frac{c}{p_a} = (B_1 \frac{\sigma_w}{p_a} + B_2 \frac{T}{T_0} + B_3 \frac{\rho}{\rho_w} + B_4) \cdot (S_{MH} - S_{MH_0})^{B_5} \qquad (13)$$

398   Assuming that the apparent cohesion can be expressed as a function of the tensile

399   strength of pure MH (σ_{tMH}), a simple dimensional expression to determine c can be

400   derived from Equations (13) and (5c):

401
$$c = b_1 \sigma_{tMH} \cdot (S_{MH} - S_{MH_0})^{b_2} \qquad (14)$$

402   where b_1 and b_2 are constant parameters (here b_1 = 0.20 and b_2 = 0.42), and σ_{tMH} is

403   given in Equation 5(c). The relationships obtained by using Equation (14) are plotted

404   as dashed lines in Figures 15 and 19. Looking at the two figures emerges that

405   Equation (14) is effective at capturing the dependence of cohesion on backpressure

406   (σ_w), temperature (T), density of pure methane hydrate (ρ) and methane hydrate

407   saturation (S_{MH}).

408   The values of internal friction angle (ϕ), calculated from the DEM tests, are

409   plotted against temperature and methane hydrate density in Figures 20a and 20b

410   respectively. Analogously to apparent cohesion, the internal frictional angle linearly

411   increases with temperature decreasing and methane hydrate density increasing

412   whereas no significant change is found associated with the variation of MH saturation.

413   Therefore, the dependence of the internal friction angle on temperature and methane

414   hydrate density may be accounted for by the following expression:

415
$$\phi = C_0 - (C_1 \frac{\sigma_w}{p_a} + C_2 \frac{T}{T_0} + C_3 \frac{\rho}{\rho_w} + C_4)^{C_5} \qquad (15)$$

416   with C_1, C_2,… C_4 being constants. This indicates that the internal friction angle can

417   be expressed as a function of the tensile strength of methane hydrate (σ_{tMH}) in a way

418   similar to that of cohesion (see Equation (14)). Hence, a simple dimensional

419   expression to determine ϕ can be derived from Equations (15) and (5c):

420          $$\phi = \phi_{host} - c_1 (\frac{\sigma_{tMH}}{p_a})^{c_2} \qquad (16)$$

421   where c_1 and c_2 are constant parameters, and ϕ_{host} is the internal friction angle of the

422   host sand (i.e., the sample without MH bonds). In our case, ϕ_{host} = 23.8^o, and c_1, c_2

423   are equal to 0.21 and 0.77, respectively. The relationships obtained by using Equation

424   (16) are plotted in Figures 16 and 20. Looking at the two figures emerges that

425   Equation (16) is effective at capturing the dependence of the internal friction angle on

426   backpressure (σ_w), temperature (*T*), density of pure methane hydrate (*ρ*) and methane

427   hydrate saturation (S_{MH}).

428

6 Conclusions

430   This paper investigates the mechanisms taking place at the microscopic level at the

431   origin of the experimentally observed influence of backpressure on the mechanical

432   properties of methane hydrate bearing sediments (MHBS) via a theoretical

433   mechanical analysis at the grain scale and a campaign of Distinct Element Method

434   tests. DEM biaxial compression tests was performed under different levels of

435   backpressure to validate the proposed bond model accounting for the influence of

436   backpressure on the MH bonds cementing the grains of the bearing sediment and to

437   investigate the relations between mechanical properties of MHBS and backpressure.

438   The main conclusions are as follows:

439   (1) in agreement with experimental tests reported in the literature, backpressure is

440   found to increase the shear strength and elastic modulus of MHBS samples in tests

441   run at constant effective confining pressure.

442   (2) DEM biaxial tests incorporating the proposed contact bond can effectively

443   capture the main features of the influence of backpressure on stress-strain and

444   volumetric behaviors of MHBS. High levels of backpressure increase shear strength,

445   elastic modulus and shear dilation of MHBS. The contribution of the backpressure to

446   the deviator stress decreases as the axial strain increases. At large axial strains, the

447   values of the deviator stress of MHBS samples with different backpressures are

448   nearly identical. This is due to the fact that weak areas without MH bonds control the

449   deviator stresses of MHBS samples at such a state.

450   (3) The DEM simulations show that the apparent cohesion and elastic modulus

451   linearly increase while the internal friction angle decreases at a decreasing rate as the

452   backpressure increases. These backpressure effects depend on the effective confining

453   pressure and MH saturation. Such dependency is formulated in Equations (8) and (10).

454

References

1. Kvenvolden KA, Ginsburg GD, and Soloviev VA. Worldwide distribution of subaquatic gas hydrates. *Geo-Marine Letters* 1993; 13:32-40.

2. Sloan ED. Gas hydrates: review of physical/chemical properties. *Energy & Fuels* 1998; 12:191-196.

3. Sloan ED. Fundamental principles and applications of natural gas hydrates. *Nature* 2003; 426:353-359.

4. Kvenvolden KA. Methane hydrate - a major reservoir of carbon in the shallow geosphere? *Chemical Geology* 1988; 71:41-51.

5. Kvenvolden KA. Gas Hydrates - Geological Perspective and Global Change. *Reviews of Geophysics* 1993; 31:173-187.

6. Kvenvolden KA, and Lorenson TD. The global occurrence of natural gas hydrate. *Geophysical Monograph* 2001; 124(322):3-18.

7. Collett TS, and Kuuskraa VA. Hydrates contain vast store of world gas resources. *Oil and Gas Journal* 1998; 96(19):90-95.

8. Max MD, and Lowrie A. Oceanic methane hydrate: a "frontier" gas resource. *Journal of Petroleum Geology* 1996; 19(1):41-56.

9. Clayton CRI, Priest JA, and Best AI. The effects of disseminated methane hydrate on the dynamic stiffness and damping of a sand. *Geotechnique* 2005; 55(6):423-434.

10. Folger P. Gas hydrates: resource and hazard. *CRS report for congress;* 2010. May 25.

11. Collett TS. Energy resource potential of natural gas hydrates. *AAPG Bulletin* 2002; 86(11):1971-1992.

12. Ayhan D. Methane hydrates as potential energy resource: Part Ⅰ - Importance, resource and recovery facilities. *Energy Conversion and Management* 2010; 51:1547-1561.

13. Kimoto S, Oka F, and Fushita T. A chemo-thermo-mechanically coupled analysis of ground deformation induced by gas hydrate dissociation. *International Journal of Mechanical Sciences* 2010; 52:365-376.

14. Brugada J, Cheng YP, Soga K, and Santamarina JC. Discrete element modelling of geomechanical behavior of methane hydrate soils with pore-filling hydrate distribution. *Granular Matter* 2010; 12:517-525.

15. Makogon YF. Hydrates of Natural Gas. *Penwell Books,* Tulsa, Oklahoma; 2008.

16. Winters WJ, Waite WF, Mason DH, Gilbert LY, and Pecher LA. Methane gas hydrate effect on sediment acoustic and strength properties. *Journal of Petroleum Science and Engineering* 2007; 56:127-135.

17. Waite WF, Santamarina JC, Cortes DD, Dugan B, Espinoza DN, Germaine J, Jang J, Jung W, Kneafsey TJ., Shin H, Soga K, Winters WJ, and Yun TS. Physical properties of hydrate-bearing sediments. *Reviews of Geophysics* 2009; 47(4): doi: 10.1029/2008RG000279.

18. Sultan N, Cochonat P, Foucher JP, and Mienert J. Effect of gas hydrates melting on seafloor slope instability. *Marine Geology* 2004; 213(1-4):379-401.

19. Terzaghi K. Theoretical soil mechanics. *John Wiley and Sons* New York; 1936. p. 11-15.

20. Miyazaki K, Masui A, Sakamoto Y, Aoki K, Tenma N, and Yamaguchi T. Triaxial compressive properties of artificial methane-hydrate-bearing sediment. *Journal of Geophysical Research: Solid Earth* 2011; 116(B6), doi: 10.1029/2010JB008049.

21. Hyodo M, Yoneda J, Yoshimoto N, and Nakata Y. Mechanical and dissociation properties of methane hydrate - bearing sand in deep seabed. *Soils and Foundations* 2013; 53(2):299-314.

22. Utili S, Nova R. DEM analysis of bonded granular geomaterials. *International Journal for Numerical and Analytical Methods in Geomechanics* 2008; 32(17):1997-2031.

23. Utili S, Crosta GB. Modelling the evolution of natural slopes subject to weathering: Part II. Discrete element approach *Journal of Geophysical Research – Earth Surface* 2001; 116, F01017, doi:10.1029/2009JF001559.

24. Jiang MJ, Yan HB, Zhu HH, and Utili S. Modelling shear behaviour and strain localisation in cemented sands by two dimensional Distinct Element Method Analyses. *Computers and Geotechnics* 2011; 38(1):14-29.

25. Jiang MJ, Zhang WC, Sun YG, and Utili S. An investigation on loose cemented granular materials via DEM analyses. *Granular matter* 2013; 15(1):65-84.

26. Hyodo M, Nakata Y, Yoshimoto N, and Yoneda J. Mechanical behavior of methane hydrate-supported sand. in *Proceedings of International Symposium on Geotechnical Engineering Ground Improvement and Geosynthetics for Human Security and Environmental Preservation* Bangkok, Thailand; 2007. p. 195-208.

27. Hyodo M, Nakata Y, Yoshimoto N, Orense R, and Yoneda J. Bonding strength by methane hydrate formed among sand particles. in *Proceedings of the 6th International Conference on Micromechanics of granular media*, edited by M. Nakagawa, and S. Luding, *AIP publishing*, Golden, Colorado; 2009. p. 79-82.

28. Masui A, Hironori H, Ogata Y, and Aoki K. Effects of methane hydrate formation on shear strength of synthetic methane hydrate sediments. in *Proceedings of the 15th International Offshore and Polar Engineering Conference*, Seoul, Korea; 2005. p. 364-369.

29. Kingston E, Clayton C, and Priest J. Gas hydrate growth morphologies and their effect on the stiffness and damping of a hydrate bearing sand. in *Proceedings of the 6th International Conference on Gas Hydrates,* Vancouver, British Columbia, Canada; 2008.

30. Jiang MJ, Zhu FY, Liu F, and Utili S. A bond contact model for methane hydrate bearing sediments with inter-particle cementation. *International Journal for Numerical and Analytical Methods in Geomechanics* 2014; 38:1823-1854.

31. Lowe J, and Johnson TC. Use of back-pressure to increase degree of saturation of triaxial test specimens. in *Proceedings of ASCE Research Conference on Shear Strength of Cohesive Soils*, Boulder; 1960. p. 819-836.

32. Black DK, and Lee KL. Saturating Laboratory Samples by Back Pressure. *Journal of the Soil Mechanics and Foundations Division* 1973; 99(1):75-93.

33. Bryant WR, Hottman W, and Trabant P. Permeability of unconsolidated and consolidated marine sediments, Gulf of Mexico. *Marine Georesources & Geotechnology* 1975; 1(1):1-14.

34. Chapuis RP, Lavoie J, and Girard D. Design, construction, performance, and repair of the soil-bentonite liners of two lagoons. *Canadian Geotechnical Journal* 1992; 29(4):638-649.

35. Haug MD, and Wong LC. Impact of molding water content on hydraulic conductivity of compacted sand-bentonite. *Canadian Geotechnical Journal* 1992; 29(2):253-262.

36. Li J, and Qubain BS. Resilient modulus variations with water content. in *Proceeding of the Symposium on Resilient Modulus Testing for Pavement Components*, edited by G. N. Durham, W. L. DeGroff, and W. A. Marr, *ASTM International*, Salt Lake City, Utah; 2003. p. 59-69.

37. Tripathi K, and Viswanadham B. Evaluation of the permeability behaviour of sand-bentonite mixtures through laboratory tests. *Indian Geotechnical Journal* 2012; 42(4):267-277.

38. Oka F. Validity and limits of the effective stress concept in geomechanics. *Mechanics of Cohesive frictional Materials* 1996; 1(2):219-234.

39. Soga K, Lee SL, Ng MYA, and Klar A. Characterisation and engineering properties of methane hydrate soils. In Characterisation and Engineering Properties of Natural Soils, edited by T. S., Tan, *Taylor and Francis*, London; 2006. p. 2591-2642.

40. Jung JW, Santamarina JC, and Soga K. Stress-strain response of hydrate-bearing sands: numerical study using discrete element method simulations. *Journal of Geophysical Research: Solid Earth* 2012; 117(B4), doi: 10.1029/2011JB009040.

41. Waite WF, Winters WJ, and Mason DH. Methane hydrate formation in partially water-saturated Ottawa sand, *American Mineralogist* 2004; 89:1202-1207.

42. Hyodo M, Nakata Y, Yoshimoto N, and Ebinuma T. Basic research on the mechanical behavior of methane hydrate-sediments mixture. *Japanese Geotechnical Society* 2005; 45(1):75-85.

43. Yun TS, Francisca FM, Santamarina JC, and Ruppel C. Compressional and shear wave velocities in uncemented sediment containing gas hydrate. *Geophysical Research Letters* 2005; *32*(10), doi: 10.1029/2005GL022607.

44. Sultan N, and Garziglia S. Geomechanical constitutive modelling of gas-hydrate-bearing sediments. in *Proceedings of the 7th International Conference on Gas Hydrates (ICGH 2011)*, Edinburgh, Scotland, United Kingdom; 2011.

45. Guerin G, Goldberg D, and Meltser A. Characterization of in situ elastic properties of gas hydrate‐bearing sediments on the Blake Ridge. *Journal of Geophysical Research: Solid Earth* 1999*;* 104(B8):17781-17795.

46. Yuan T, Spence GD, Hyndman RD, Minshull TA, and Singh SC. Seismic velocity studies of a gas hydrate bottom‐simulating reflector on the northern Cascadia continental margin: Amplitude modeling and full waveform inversion. *Journal of Geophysical Research: Solid Earth* 1999; 104(B1):1179-1191.

47. Clayton CRI, Priest JA, and Rees EVL. The effects of hydrate cement on the stiffness of some sands. *Geotechnique* 2010; 60(6): 435-445.

48. Stern LA, and Kirby SH. Polycrystalline methane hydrate: synthesis from superheated ice, and low-temperature mechanical properties. *Energy and Fuels* 1998; 12:201-211.

49. Nabeshima Y, and Matsui T. Static shear behaviors of methane hydrate and ice. in *Proceeding of the 5th Oceanic Mining Symposium*, Tsukuba, Japan; 2003. p. 156-159.

50. Song Y, Yu F, Li Y, Liu W, and Zhao J. Mechanical properties of artificial methane hydrate under triaxial compression. *Journal of Natural Gas Chemistry* 2010; 19:246-250.

51. Yu F, Song Y, Liu W, Li Y, and Lam W. Analyses of stress strain behavior and constitutive model of artificial methane hydrate. *Journal of Petroleum Science and Engineering* 2011; 77:183-188.

52. Li Y, Song Y, Liu W, and Yu F. Experimental research on the mechanical properties of methane hydrate-ice mixtures. *Energies* 2012; 5(2):181-192.

53. Jiang MJ, Sun YG, Li LQ, and Zhu HH. Contact behavior of idealized granules bonded in two different inter-particle distances: An experimental investigation. *Mechanics of Materials* 2012; 55:1-15.

54. Jiang MJ, Sun YG, and Xiao Y. An experimental investigation on the contact behavior between cemented granules. *Geotechnical Testing Journal*, 2012; 35(5): 678-690.

55. Dattola G., Di Prisco C, Redaelli I, Utili S. (2014). A distinct element method numerical investigation of compaction processes in highly porous cemented granular materials. *International Journal for Numerical and Analytical Methods in Geomechanics*, 38(11): 1101–1130.

56. Itasca Consulting Group, Particle flow code in 2 dimensions, Version 3.1, Minnesota, USA; 2004.

57. Jiang MJ, Konrad JM, and Leroueil S. An efficient technique for generating homogenous specimen for DEM studies. *Computers and Geotechnics* 2003; 30:579-597.

58. Santamarina JC, and Jiang J. Gas production from hydrate bearing sediments: geomechanical implications. *Fire in the ice* 2009; 9(4):18-22.

59. Makogon YF. Natural gas hydrates – a promising source of energy. *Journal of Natural Gas Science and Engineering* 2010; 2:49-59.

60. Stern LA, Circone S, Kirby SH, and Durham WB. Anomalous Preservation of Pure Methane Hydrate at 1 atm. *The Journal of Physical Chemistry B* 2001; 105:1756-1762.